

### Evaluation of energy production from municipal wastewater using forward osmosis process and anaerobic membrane bioreactor

Sosuke Onoda<sup>a,b</sup>, Yuta Masumori<sup>a</sup>, Taro Miyoshi<sup>a</sup>, Masahiro Yasukawa<sup>a</sup>, Tomoki Takahashi<sup>a</sup>, Hideto Matsuyama<sup>a,\*</sup>

<sup>a</sup>Center for Membrane and Film Technology, Department of Chemical Science and Engineering, Kobe University, 1-1 Rokkodai, Nada-ku, Kobe 657-8501, Japan, emails: matuyama@kobe-u.ac.jp (H. Matsuyama), yuta.mas1024@gmail.com (Y. Masumori), t-miyoshi@pegasus.kobe-u.ac.jp (T. Miyoshi), myasukawa@people.kobe-u.ac.jp (M. Yasukawa), t.takahashi@crystal.kobe-u.ac.jp (T. Takahashi) <sup>b</sup>Research and Development Planning Department, Technical Development Division, Technical Research Center, Kobelco Eco-Solutions Co., LTD., 1-1-4 Murotani, Nishi, Kobe 651-2241, Japan, email: s.onoda@kobelco-eco.co.jp

Received 6 May 2016; Accepted 22 October 2016

#### ABSTRACT

The anaerobic treatment is a potential way to realize the energy production in wastewater treatment. However, it is necessary to increase the organic matter concentration in municipal wastewater. In this study, we proposed a wastewater treatment system that involved an anaerobic membrane bioreactor (AnMBR) combined with a forward osmosis (FO) process to concentrate the wastewater. The energy balance of this system was estimated by the calculation based on the FO condensation test and batch methane fermentation test. The methane yield from the condensed municipal wastewater in the batch methane fermentation test at 40°C was  $0.22 \text{ m}^3$ -CH<sub>4</sub>/kg-COD<sub>Cr rem</sub> putting COD<sub>cr</sub> as the chemical oxygen demand was determined by chromium oxidation method. The energy generated from the yielded methane was estimated to be  $0.33 \text{ kWh/m}^3$ . Nevertheless, the total energy consumption was  $0.39 \text{ kWh/m}^3$ , which was higher than  $0.2 \text{ kWh/m}^3$  of a conventional activated sludge process. It was found that it is possible to get a net energy if the fermentation temperature in the AnMBR process can be reduced to below  $34^\circ$ C.

Keywords: Wastewater condensation; Forward osmosis; Anaerobic membrane bioreactor

#### 1. Introduction

Wastewater treatment plays a very important role with regard to public hygiene in a city. However, a huge amount of energy is devoted to wastewater treatment. The amount of energy consumed for wastewater treatment is approximately 700 GWh in Japan, which corresponds to 0.7% of the total energy consumption in Japan [1]. Furthermore, it has been reported that this figure is 3% in the USA [2]. At present, aerobic biological treatment (e.g., conventional activated sludge process), in which the organic matter presented in wastewater is converted to carbon dioxide and microbial

cells under aerobic condition, is the prime choice for treating municipal wastewater. Many wastewater treatment plants based on the conventional activated sludge process have been in operation for several decades. As a result, considerable operational knowledge of the aerobic wastewater treatment processes has already been accumulated. In such aerobic wastewater treatment processes, aeration is essential for maintaining the dissolved oxygen (DO) concentration in the bioreactor at a certain level (e.g., more than 2 mg/L [3]) to keep the aerobic microorganisms active. However, aeration generally consumes a huge amount of energy. In general, 45%–75% of the total operational cost of the aerobic biological wastewater treatment is due to the aeration in the bioreactor

<sup>\*</sup> Corresponding author.

<sup>1944-3994/1944-3986 © 2017</sup> Desalination Publications. All rights reserved.

[4]. Reducing the energy consumption associated with aeration is critical for ensuring the sustainability of wastewater treatment.

On the other hand, in addition to aerobic treatment methods, organic matter can also be removed from wastewater under anaerobic conditions. In the anaerobic treatment, organic constituents contained in wastewater are converted to methane and carbon dioxide (sometimes referred to as biogas) by anaerobic microorganisms [5]. Since aeration is not required for anaerobic treatment, the energy consumption in anaerobic treatment is generally much lower than that in aerobic treatment. The additional advantages of applying anaerobic treatment are reduced sludge production and the potential to produce methane gas, which can be used as fuel. Since municipal wastewater contains plenty of organic matter, which can be a source of methane generation, applying anaerobic treatment to municipal wastewater treatment would be an attractive option for reducing the energy demand associated with wastewater treatment. According to the estimation made by McCarty et al. [2], domestic wastewater treatment could be a net energy producer by using a completely anaerobic treatment. However, anaerobic treatment is difficult to apply for wastewater with low concentration of chemical oxygen demand (COD). It was reported that raw water with a COD determined by chromium oxidation method  $(COD_{Cr})$  concentration higher than 1,500–2,000 mg/L is preferable for anaerobic treatment [6]. However, the typical  $\mathsf{COD}_{\mathsf{Cr}}$  concentration in municipal wastewater is in the range of 250-800 mg/L [6]. Therefore, in order to perform the anaerobic treatment efficiently, the raw wastewater should be condensed prior to introducing it into the anaerobic bioreactor. Lateef et al. [7] succeeded in concentrating actual municipal wastewater to a COD<sub>c</sub>, concentration of more than 6,000 mg/L using a microfiltration (MF) membrane. Based on the COD<sub>Cr</sub> concentration in the condensed wastewater, they estimated that up to 0.5 kWh of electricity per cubic meter of wastewater could be produced by applying anaerobic treatment to the condensed wastewater. The study performed by Lateef et al. [7] clearly indicated that membrane separation is one of the most suitable technologies for enabling the recovery of energy in the form of methane, from municipal wastewater. However, the use of a MF membrane has several limitations. First, the MF membrane permeate cannot be directly discharged into a natural water body as some dissolved matters contained in the wastewater permeate through an MF membrane. Due to this, some post-treatment is required to improve the quality of treated water. The other issue is the loss of organic matter into the permeate water. Since approximately half of the organic matter contained in municipal wastewater is in dissolved state (i.e., cannot be filtered by the MF membrane), the efficiency of the MF membrane in recovering organic matter is believed to be limited. On the basis of the points mentioned above, it can be said that proper selection of membrane is essential for the successful condensation of organic matter contained in municipal wastewater.

This study focused on the application of forward osmosis (FO) membrane for concentrating the organic matter contained in municipal wastewater. FO membrane filtration process has been drawing attention due to its unique characteristic (i.e., applying hydraulic pressure is not required during the membrane filtration [8]). In FO membrane filtration process, water molecules contained in the feed solution (FS) with a relatively low osmotic pressure are spontaneously transported into the draw solution (DS), which has an osmotic pressure higher than the FS, on the basis of the osmotic pressure gradient. Since the FO membrane generally has a rejection capacity similar to the reverse osmosis (RO) membrane, almost all the dissolved components are expected to be rejected by applying the FO membrane. This feature of the FO membrane is thought to be a clear advantage of this technology from the point of view of the quality of treated water, especially in comparison with porous membranes (i.e., MF and ultrafiltration membranes). In addition, several researchers have reported that the FO membrane filtration process is one of the most suitable technologies for concentrating products containing high organic matter or suspended solid condensations [9–11], because the structure of the fouling layer developed during the process of FO membrane filtration is looser than during the RO membrane filtration process, making the recovery of membrane fouling easier [12]. By applying direct FO membrane filtration, the treatment of wastewater and the production of condensed wastewater for the anaerobic digester were achieved simultaneously. Gu et al. [13] proposed an anaerobic membrane bioreactor (AnMBR) system, in which an FO membrane is directly submerged in the anaerobic bioreactor. In their study, however, elevated salt concentration in the bioreactor as a result of the back-diffusion of salt from the DS caused inhibitory effects on the activity of microorganisms responsible for anaerobic treatment. The increase in salt concentration in the bioreactor also resulted in a decrease in the driving force of the FO membrane filtration (i.e., the effective osmotic pressure difference between the bioreactor and DS). Zhang et al. [14] achieved a condensation of more than 300% by using FO membrane to treat municipal wastewater. However, they did not evaluate the significance of the concentrate ratio obtained in this study for anaerobic treatment.

Based on the background mentioned above, this study investigated the performance of an FO membrane with regard to concentrating actual municipal wastewater in terms of both, the rejection of constituents contained in the wastewater and the development of membrane fouling. The concentrate was subsequently subjected to a batch methane fermentation test, to evaluate the methane production potential of condensed municipal wastewater. The results obtained in these tests were used for evaluating the energy balance in the wastewater treatment system comprised up-concentrating wastewater, using an FO membrane, followed by anaerobic treatment (AnMBR). On the basis of the results obtained in this study, the technical feasibility of net energy-producing wastewater treatment systems is also discussed.

#### 2. Experimental setup

#### 2.1. Condensation of municipal wastewater using FO membrane

In this experiment, effluent from the primary sedimentation tank in the Port Island wastewater treatment plant in Kobe city, Japan, was used as feed water for an FO membrane filtration test. The characteristics of the wastewater used in this study have been summarized in Table 1. A schematic illustration of the experimental apparatus is shown in Fig. 1. The condensation tank, in which a small FO membrane module was directly submerged, was made of transparent acrylic resin, with an effective volume of 1,500 mL. During the experiment, the condensation tank was hermetically sealed. The FO membrane used in this experiment was an asymmetrically hollow fiber membrane made of cellulose triacetate (Toyobo, Osaka, Japan). This FO membrane had an active layer on the outer surface. In a small FO membrane module used in this study, 350 fibers were bundled, resulting in a total effective membrane surface area of 0.0846 m<sup>2</sup>. The membrane orientation was active layer facing the FS (AL-FS). This was because membrane fouling is generally less significant in this orientation [15]. 0.6 M NaCl solution was used as the DS, with an initial volume of 2,000 mL. The cross flow velocity of DS in the hollow fiber was set to 5.5 cm/s. During the FO membrane filtration test, the salt concentration of the DS was maintained at a constant by supplying saturated NaCl solution (approximately 4.0 M), based on the decrease in electrical conductivity, using a conductivity control device (CM-31P, Toa-DKK, Tokyo, Japan). The wastewater was added to the condensation tank using a peristaltic pump (MP-1000, Eyela, Tokyo, Japan). The feed pump was monitored using a water level sensor, to keep the water level constant.

In the FO membrane filtration test mentioned above, the flow rate of water across the FO membrane (Q) was evaluated

#### Table 1

Water quality of the effluent from the primary sedimentation tank in the Port Island wastewater treatment plant (feed water for the concentration test)

Item	Value
TOC, mg/L	88.5
COD <sub>cr</sub> mg/L	300
$PO_4$ –P, mg-P/L	4.2
NH <sub>4</sub> –N, mg-N/L	34.0

from the rate of decrease in the mass of the feed water in the feed water tank. The water flux  $J_w$  was calculated using the following equation:

$$J_W = \frac{Q}{A} = \frac{m_0 - m_t}{A \cdot t} \tag{1}$$

where Q is the flow rate through the FO membrane (g/h); Ais the effective membrane surface area  $(m^2)$ ; *t* is the operation time (h);  $m_0$  is the initial weight of the feed tank (g); and  $m_t$  is the weight of the feed tank at operation time t(g). To maintain the water permeability of the FO membrane during the condensation operation, a simple physical cleaning was carried out every 22 h, by stirring the inside part of the condensation tank firmly with a magnetic stirrer. The reason why the stirring is not kept on inside part of the condensation tank is to prevent stirred up the suspension. We pre-experimentally confirmed that the operation without stirring inside part of the condensation tank could keep flux better. Generally, the membrane cleaning is carried out based on the water flux decline by membrane fouling to keep a stable operation of the membrane filtration. In a normal membrane filtration system for water treatment, the membrane cleaning will be carried out based on about 20% flux decline. However, in this study, it was important to understand how much the water flux decline was. Then, cleaning was carried out once a day. It took 2 h to start filtration after the beginning of cleaning. Thus, we set the physical cleaning interval as 22 h. Then, cleaning was carried out once a day. After the physical cleaning, it was subjected to pure water permeation test using a Milli-Q (as FS) and 0.6 M NaCl (as DS) for the assessment of the degree of irreversible fouling.

# 2.2. Batch methane fermentation test using condensed municipal wastewater

A batch methane fermentation test was carried out to evaluate the methane gas yield from condensed wastewater. The schematic system of batch methane fermentation test is shown in Fig. 2. 0.4 L of anaerobically digested sludge,



Fig. 1. Schematic system of the condensation system using hollow fiber FO membrane.



Fig. 2. Schematic system of the batch methane fermentation test.

collected from the Higashinada wastewater treatment plant in Kobe city, Japan, was placed in a conical flask. 0.4 L of condensed wastewater was added to the flask, and then, the flask was hermetically sealed. During the test, the water temperature was maintained at 40°C, while the mixture of the digested sludge and condensed wastewater was continuously agitated using a magnetic stirrer. The biogas thus generated was collected in a gas holder, and its volume was recorded. In order to subtract the gas generated from the organic matter originally contained in the anaerobically digested sludge, a blank test was carried out using exactly the same digested sludge, collected at the same place and at the same time. In the blank test, 0.4 L NaCl solution, which had an electrical conductivity comparable with the condensed wastewater subjected to the batch methane fermentation test, was added to the anaerobically digested sludge instead of the condensed wastewater. The experimental conditions were the same in both the flasks, except for the characteristics of the added liquid (i.e., the condensed wastewater and the NaCl solution). Since the NaCl solution added did not have any organic matter, which could be a source of methane generation, any gas generated in the blank flask is considered as having originated from the organic matter originally contained in the anaerobically digested sludge. Therefore, the difference in the amount of biogas generated between the two flasks can be considered as the amount of biogas generated from the organic matter contained in the condensed wastewater.

Based on the assumption that methane occupied 70% of the biogas generated [16], methane yield  $\eta_{CH4}$  was calculated by the following equation:

$$\eta_{\rm CH4} = \frac{\left(V_1 - V_0\right) \cdot 0.7}{V_{\rm mix} \cdot \left\{ \left(C_1^{\rm start} - C_1^{\rm end}\right) - \left(C_0^{\rm start} - C_0^{\rm end}\right) \right\}}$$
(2)

where  $\eta_{CH4}$  is the methane yield (m<sup>3</sup>-CH<sub>4</sub>/kg-COD<sub>C</sub>);  $V_1$  is the volume of biogas generated from the digested sludge mixed with the condensed wastewater (m<sup>3</sup>);  $V_0$  is the volume of biogas generated from blank (m<sup>3</sup>);  $V_{mix}$  is the total volume of the digested sludge and condensed wastewater (m<sup>3</sup>);  $C_1^{\text{start}}$  and  $C_1^{\text{end}}$  are the COD<sub>Cr</sub> concentrations of the digested sludge mixed with the condensed wastewater at start and end of the batch fermentation test (kg-COD<sub>C</sub>/m<sup>3</sup>), respectively; and

 $C_0^{\text{start}}$  and  $C_0^{\text{end}}$  are the COD<sub>Cr</sub> concentration at start and end of the test for blank, respectively.

#### 2.3. Analytical methods

The concentration of  $\text{COD}_{cr}$  was determined by chromium oxidation method (DRB200, Hach, Colorado, USA). The concentrations of the total organic carbon (TOC) and the total inorganic carbon (TIC) were determined using a TOC analyzer (TOC-VCSH, Shimadzu, Kyoto, Japan). The concentration of phosphate ion PO<sub>4</sub>–P was determined by the molybdenum blue method [17], while the concentration of ammonium ion was determined by the closed salicylate-chlorine method (Hach, Colorado, USA) using a Hach spectrometer (DR900).

#### 3. Material and energy balance

### 3.1. Calculation of the material balance of organic matter during up-condensation using FO membrane

The material balance of the organic matters, which were fed to the condensation tank during the municipal wastewater condensation test, was calculated by dividing the organic matter into the following parts: accumulated in the condensation tank, permeated through the FO membrane and lost due to some experimental limitations (e.g., mineralization by biodegradation, accumulation into a dead-space, such as adherence to the inner wall of the condensation tank or loss at the time of physical cleaning). Each quantity was calculated by the following equations:

$$Q_{\text{Feed}}^{\text{TOC}} = C_{\text{waste}}^{\text{TOC}} \cdot \left( \sum V_{\text{permeate}} + V_{\text{tank}} \right)$$
(3)

$$Q_{\text{accumulate}}^{\text{TOC}} = C_{\text{conc}}^{\text{TOC}} \cdot V_{\text{tank}}$$
(4)

$$Q_{\text{permeate}}^{\text{TOC}} = \sum \left( C_{\text{permeate}}^{\text{TOC}} \cdot V_{\text{permeate}} \right)$$
(5)

$$Q_{\text{lost}}^{\text{TOC}} = Q_{\text{Feed}}^{\text{TOC}} - Q_{\text{accumulate}}^{\text{TOC}} - Q_{\text{permeate}}^{\text{TOC}}$$
(6)

where  $Q_{\text{Feed}}^{\text{TOC}}$  is the quantity of the TOC, which was fed into the condensation tank during the condensation test (mg);

 $C_{\text{waste}}^{\text{TOC}}$  is the TOC concentration of municipal wastewater (mg-C/L);  $V_{\text{permeate}}$  is the volume of water that permeates the FO membrane measured every 12 h during the condensation test (L);  $V_{\text{tank}}$  is the volume of condensation tank (L);  $Q_{\text{accumlate}}^{\text{TOC}}$  is the quantity of the TOC accumulated in the condensation tank (mg);  $C_{\text{conc}}^{\text{TOC}}$  is the TOC concentration in the condensed wastewater at end of the condensation test (mg-C/L);  $Q_{\text{permeate}}^{\text{TOC}}$  is the TOC, which permeated the FO membrane (mg); and  $C_{\text{permeate}}^{\text{TOC}}$  is the TOC concentration of the permeate measured every 12 h during condensation test (mg-C/L).

#### 3.2. Estimation of energy balance

The proposed system, involving the condensation of wastewater and the AnMBR, is schematically shown in Fig. 3. The net energy generation,  $E_{nst'}$  for treating a unit volume of municipal wastewater in the proposed system, was calculated by the following equation:

$$E_{net} = E_g - E_C \tag{7}$$

where  $E_{nst}$  is the net energy generation (kWh/m<sup>3</sup>);  $E_g$  is the energy generation (kWh/m<sup>3</sup>); and  $E_c$  is the energy consumption (kWh/m<sup>3</sup>).

Energy consumption ( $E_c$ ) can be further divided into energy for operation (i.e., energy required for pumping etc.) and energy for heating (required for maintaining the temperature of the mixed liquor suspension during the anaerobic treatment). With regard to the operational energy, the energy consumption for operating the FO membrane filtration device and the AnMBR were assumed to be 0.1 kWh/m<sup>3</sup> [18] and 0.06 kWh/m<sup>3</sup> [19], respectively. For calculating the energy required for heating the anaerobic bioreactor, the following assumptions were made: the bioreactor is thermally insulated (i.e., negligible heat exchange with the outside), and heat is exchanged between the effluent and the influent of an AnMBR. Under the assumptions mentioned above,  $E_c$  was calculated by the following equation:

$$E_{c} = 0.1 + \frac{\left\{C_{P} \cdot \left(T_{An} - T_{in}^{'}\right) + 0.06 + E_{C_{Post}}\right\}}{R_{FO}}$$

$$= 0.1 + \frac{C_{P} \cdot \left[T_{An} - \left\{\left(\frac{T_{in} + T_{An}}{2} - T_{in}\right) \cdot \eta_{HEx} + T_{in}\right\}\right] + 0.06 + E_{C_{Post}}}{R_{FO}}$$
(8)

where  $C_p$  is the specific heat of water (kWh/m<sup>3</sup>/K);  $T_{An}$  is the operational temperature of AnMBR (K),  $T_{in}$  is the temperature of the influent in the heat exchanger (K);  $T_{in}$  is the temperature of the influent in the AnMBR (i.e., the effluent of the heat exchanger; K);  $\eta_{HEx}$  is the efficiency of the heat exchanger;  $E_{CPost}$  is the energy consumption during post-treatment after AnMBR (kWh/m<sup>3</sup>); and  $R_{FO}$  is the volume basis condensation rate of wastewater by FO membrane. The post-treatment system after AnMBR and its energy consumption  $E_{CPost}$  have been discussed later in this paper, based on the experimental results obtained.

The energy generated by the combustion of methane gas obtained from the anaerobic treatment  $E_g$  was calculated by the following equation:

$$E_{g} = \frac{C_{\rm in} \cdot \operatorname{Rem}_{AnMBR}^{\rm COD_{-}Cr} \cdot \eta_{\rm CH4} \cdot H_{\rm CH4} \cdot \eta_{\rm CG}}{R_{\rm FO}}$$
(9)

where  $C_{in}$  is the COD<sub>Cr</sub> concentration of the influent of AnMBR (i.e., the condensed wastewater; kg-COD<sub>Cr</sub>/m<sup>3</sup>), and Rem<sup>COD\_Cr</sup><sub>AnMBR</sub>. is removal ratio of COD<sub>Cr</sub> at AnMBR. In this study, the removal ratio of COD<sub>Cr</sub> from the batch methane fermentation test mentioned above was applied to Rem<sup>COD\_Cr</sup><sub>AnMBR</sub>.  $\eta_{CH4}$  is the methane yield (m<sup>3</sup>-CH<sub>4</sub>/kg-COD<sub>Cr</sub><sub>-rem</sub>);  $H_{CH4}$  is the heat quantity of methane gas (kWh/m<sup>3</sup>-CH<sub>4</sub>); and  $\eta_{CG}$  is the energy conversion efficiency of methane gas.  $\eta_{CG}$  is approximately 0.5 in conventional thermal power generation processes, although this efficiency can be increased up to approximately 0.8, when combined with an exhaust heat recovery system [18].



Fig. 3. Schematic system of the proposed system.

#### 4. Results and discussion

#### 4.1. Membrane fouling in hollow fiber FO membrane

Variation in water flux during the condensation of wastewater using hollow fiber FO membrane module is shown in Fig. 4. The closed plots show the water flux during the FO membrane filtration of wastewater, while the open plots show the water flux determined by filtering the Milli-Q water after physical cleaning. Although the water flux in the FO filtration of wastewater clearly decreased during the continuous filtration for 36 h due to membrane fouling, most of the membrane fouling that developed was reduced by the physical cleaning. The decrease in the Milli-Q water flux determined after physical cleaning was not pronounced throughout the experiment. This indicates that the majority of the membrane fouling that developed during the FO filtration of wastewater for 36 h was reversible using physical methods. It also suggests that a stable, long-term operation of the FO membrane filtration for concentrating municipal wastewater can be achieved by adopting appropriate physical cleaning methods during the filtration. In the pressure-driven membrane filtration processes, such as an MF membrane filtration, the foulant layer is compacted by mechanical pressure and deposited on the membrane surface, which increases the resistance to filtration [12]. In contrast, in the FO membrane filtration process, the compaction of the fouling layer is expected to be less significant because of the absence of extensive hydraulic pressure. Therefore, it is difficult to consider the compaction of the fouling layer as a dominant factor for increasing the resistance to filtration in FO membranes. One of the reasons for the decrease in the FO flux is the reduction of the effective osmotic pressure difference caused by the NaCl leaked from the DS side. The leaked NaCl accumulates in the cake layer on the membrane surface formed by the foulant and leads to "cake enhanced osmotic pressure" [19].

Although membrane fouling can be almost completely reduced by physical cleaning carried out once every 36 h, the water flux measured immediately after physical cleaning gradually decreased as the number of cycles increased. The gradual decrease in water flux after the physical cleaning can be explained by the decrease in the effective osmotic pressure difference between the feed wastewater and the DS. The potential causes of this decrease might be the increase



Fig. 4. Variation in flux during the concentration of municipal wastewater. The arrow shows the physical cleaning of membrane.

in the concentration of salts originally contained in the raw wastewater (the final volumetric condensation ratio after 180 h of operation was approximately 19 times) and the diffusion of salts contained in the DS as a result of the so-called reverse solute diffusion (i.e., the solute contained in a DS diffuses into an FS based on a condensation gradient). According to the previous report of our research group [20], the reverse salt flux of the FO membrane in this study is approximately 0.004 mol-NaCl/m<sup>2</sup>/h. Considering the membrane area and the operation time in this study, the amount of NaCl diffused from DS side to feed side after each batch is estimated approximately at 0.43 g. And the NaCl concentration of condensed wastewater after 180 h operation is estimated approximately at 2.3 g-NaCl/L. However, this NaCl concentration will be overestimated. As the salt concentration difference between feed side and DS side decreases with operation time, accordingly, the reverse salt flux will become smaller. Therefore, the NaCl concentration of condensed wastewater at the end of 180 h operation will be lower than this estimated value. And this is consistent with decrease in water flux. To minimize the reduction in water flux during the up-condensation of municipal wastewater using FO membrane, it is important to develop an FO membrane with a low reverse solute flux from DS to FS.

#### 4.2. Water qualities of permeate in DS side

The concentrations of TOC,  $PO_4-P$  and  $NH_4-N$  of feed, permeate and concentrate after 180 h operation in FO condensation test are shown in Table 2. The concentrations of TOC,  $PO_4-P$  and  $NH_4-N$  of the permeate are shown in Fig. 5 as function of operation time. In Fig. 5, the  $COD_{Cr}$  concentration in the permeate could not be measured, because the diluted DS with the permeate had a high salt concentration. For this reason, it was substituted with TOC, as an indicator of organic matter. The initial TOC and the phosphate ion concentrations of the permeate were 11.5 mg-C/L and less than 0.1 mg-P/L, respectively. The rejections of TOC and phosphate ion were more than 90% and 98%, respectively. These results indicate that sufficient rejection was achieved for the organic matter and the phosphorus, by the FO membrane filtration alone.

In contrast, the concentration of ammonium ions exceeds the discharge standards of the total nitrogen in Hyogo Prefecture, Japan (20 mg/L). In the filtration process using an FO membrane, the concentration of ammonium ions in the feed side of the FO membrane increases as the wastewater gets condensed. The gradual increase in the concentration of ammonium ions in the feed water results in its increase in

Table 2

Water qualities of feed, permeate and condensate in FO condensation test. The value of "Permeate" and "Feed" show the concentration at the end of 180 h operation

Item, mg/L	Feed	Permeate	Condensate
TOC	88.5	5.5	1,409
PO <sub>4</sub> -P	4.2	ND	ND
NH <sub>4</sub> -N	34.0	29.7	130

Note: ND = No data.



Fig. 5. Variation in TOC,  $NH_4$ –N and  $PO_4$ –P of permeate during the municipal wastewater condensation test.

the permeate. At the end of this experiment, when the wastewater was condensed up to 19 times based on volume, the concentration of ammonium ions in the permeate reached 29.7 mg/L. To make the proposed process feasible, a significant improvement is essential, in the rejection of ammonium ions by the FO membrane. On the other hand, if ammonia rejection by the FO membrane is improved, the concentration of ammonia in the condensed wastewater will increase. A high concentration of ammonia is known to inhibit the methane fermentation in anaerobic treatment. In mesophilic methane fermentation, it is necessary to control the concentration of ammonium nitrogen to less than 2,000 mg/L [21]. Considering the typical concentration of 10-40 mg/L [22] of total nitrogen (most of the nitrogen species, including organic nitrogen, are generally transformed into ammonium ions during anaerobic digestion) in municipal wastewater, a condensation of more than 50 times, this value is required to increase the concentration of ammonium ions to a level at which methane fermentation would be inhibited. On the other hand, as discussed later, the results obtained in this study indicate that a condensation of 19 times by an FO membrane is sufficient for achieving a wastewater treatment system with net energy production. Therefore, the elevated concentration of ammonium ions in the condensed wastewater would not be problematic from the point of view of the efficiency of methane fermentation. In fact, the decrease in the ammonium ion concentration in the FO membrane permeate would be more important, especially when a stringent discharge standard needs to be met, considering the problems associated with eutrophication.

## 4.3. Accumulation of organic matter in condensed municipal wastewater

Fig. 6 shows the variation in the concentrations of TOC and TIC in the condensed municipal wastewater during the up-condensation of municipal wastewater using the FO membrane. As seen in Fig. 6, the concentration of the total carbon (TC; i.e., the sum of TOC and TIC) gradually increased. On the other hand, the ratio of TIC to TC increased from 0.3 at the beginning to 0.4 at the end of the experiment, indicating that a portion of the organic matter rejected by



Fig. 6. Variation in the TIC and TOC concentration of condensate during the municipal wastewater condensation test.

the FO membrane was mineralized during the experiment. This result suggests that further improvement in the concentration of organic matter is possible by reducing the time required for condensation.

It was not possible to accurately measure the COD<sub>cr</sub> due to the inhibition by the salt concentration in the condensed wastewater. However, if the COD<sub>C</sub>/TOC ratio were kept constant during the condensation test, the COD<sub>c</sub>, concentration of the condensed wastewater at 180 h might be about 4,800 mg/L (i.e., the concentrate factor for organic matter is approximately 16 times). This value is apparently higher than that required for efficient anaerobic treatment (i.e., 1,500–2,000 mg/L [6]). The value of COD<sub>Cr</sub> concentration achieved in the experiment in this study was higher than that reported by Zhang et al. (i.e., 1,642.3 mg/L), while the results obtained in this study reinforce the findings obtained by Zhang et al. [14], which suggested that pre-condensation using an FO membrane is highly feasible when applying an anaerobic treatment to the main stream of wastewater treatment for potential energy production.

As mentioned above, there is some potential for improving the accumulation of COD<sub>Cr</sub> by minimizing the mineralization during the up-condensation of municipal wastewater using an FO membrane. To minimize the mineralization, the retention time of the condensed municipal wastewater in the condensation chamber should be as short as possible. In other words, it is important to draw the FO membrane permeate from the raw wastewater as quickly as possible. In this study, the packing density of the FO membrane in the condensation chamber (i.e., the membrane surface area available in a unit volume of the chamber) was arbitrarily selected. It is certainly possible to further increase the surface area of the membrane installed in the chamber. An appropriate selection of the packing density of the FO membrane is apparently an important research topic for future. In addition, an effective membrane cleaning method for maintaining the membrane flux during the condensation should also be investigated. In the experiment carried out in this study, although the development of the physically irreversible fouling was marginal, as shown in Fig. 4, the membrane flux gradually decreased during the FO membrane filtration of the municipal wastewater. If an effective membrane cleaning method is applied during the FO membrane filtration, the membrane flux can be kept at higher level, resulting in a rapid withdrawal of the FO membrane permeate from the municipal wastewater.

## 4.4. Material balance of organic matter during up-condensation using FO membrane

The material balance of the organic matter determined at the end of up-condensation using the FO membrane is shown in Fig. 7. Of the total organic matter subjected to the up-condensation, 84% was accumulated in the concentrate, while 8% was lost into the permeate through the FO membrane. In addition, 8% of the organic matter was likely to be lost due to some experimental limitations (e.g., mineralization by biodegradation, accumulation into a dead-space, such as adherence to the inner wall of the condensation tank or loss at the time of physical cleaning). In the study performed by Lateef et al. [7], the fraction of organic matter that could be recovered through condensation using an MF membrane was limited to slightly more than half of the organic matter introduced into the condensation chamber. In the present study, however, a larger fraction of the organic matter contained in the actual municipal wastewater can be recovered through the up-condensation of wastewater using an FO membrane. This difference could be attributed to the difference in the type of membrane used for concentrating wastewater. Generally, an FO membrane has a rejection capability almost equal to that of the RO membrane. Therefore, the rejection of dissolved organic matter by the FO membrane is expected to be much higher than that by the MF membrane. Such a high rejection of organic matter by the FO membrane filtration would make this process suitable as a pre-treatment, prior to the anaerobic treatment for producing methane gas.

With regard to the organic matter mineralized by biodegradation during the up-condensation of the municipal wastewater, a significant reduction in mineralization can be expected by decreasing the retention time in the condensation tank (approximately 10 h in the experiment carried out in this study). As the condensation chamber used in this study was arbitrarily constructed, it is expected that there is some potential for increasing the packing density of the membrane further. By increasing the packing density of the FO membrane in the condensation chamber, the volume of water filtrated through the FO membrane in a unit time increases, which results in the decrease in the retention time required in the condensation chamber.



Fig. 7. Material balance of the organic matter during the condensation of municipal wastewater by the FO membrane.

#### 4.5. Batch methane fermentation test

The condensed wastewater used in this test is different from the condensed wastewater obtained for the FO membrane filtration test from the mentioned above. The COD<sub>cr</sub> concentration of the condensed wastewater used in this test was 2,600 mg/L. The biogas generated in the blank test originated from the organic matter contained in the seed sludge (i.e., digested sludge collected from a full-scale anaerobic digester). Therefore, the difference between the volume of biogas generated in the blank test and the batch fermentation test (in which the condensed municipal wastewater was used) was the volume of biogas generated from the organic matter contained in the condensed wastewater. The result of the methane fermentation test indicates that the volume of biogas from the reactor, which was fed into a 0.4-L condensed wastewater, is 0.3 L, while it is 0.1 L from the blank reactor. On the basis of the assumption mentioned above, the volume of biogas generated from 0.4 L of the condensed municipal wastewater was estimated to be 0.2 L. In addition, the amount of COD<sub>Cr</sub> removal at the end of the operation was  $0.64 \text{ g-COD}_{Cr}$  (the removal rate of COD<sub>Cr</sub> was 0.62). Taking the above into consideration, the methane yield from the organic matter contained in the condensed municipal wastewater was calculated to be 0.22  $m^3$ -CH<sub>4</sub>/kg-COD<sub>Cr.rem</sub> (assuming methane occupied 70% of the biogas produced [23]). This value is similar to the methane yield from the anaerobic fermentation of organic matter contained in municipal wastewater reported in previous studies [24,25]. The methane yield is dependent on the composition of raw water. However, the composition of municipal wastewater should not greatly vary like the industrial wastewater. Thus, we believe that it is reasonable to compare our result with the references [24,25]. As mentioned above, the volumetric up-condensation factor of the condensed wastewater used in this test was approximately 10. Therefore, the amount of biogas that can be generated from 1 L of raw wastewater without condensation is estimated to be 0.035 L. This roughly corresponds to the generation of 0.28 kWh of electricity by treating 1 m<sup>3</sup> of wastewater, based on the assumption that the heat quantity of methane gas  $H_{\rm CH4}$  is 36 MJ/m<sup>3</sup>-CH<sub>4</sub> [26], and the energy conversion efficiency of methane gas  $\eta_{CG}$  is 0.8 [21].

#### 4.6. Estimation of net energy balance

Based on the energy generation potential of the condensed wastewater obtained in the experiment described in the previous sections, the energy balance of the wastewater treatment system involving the up-condensation of wastewater using an FO membrane and AnMBR was evaluated. The results of the tests are shown in Table 3.

In the estimation, the method for post-treatment after AnMBR should also be considered. Considering that the  $COD_{Cr}$  concentration of the condensed wastewater was 4,800 mg- $COD_{Cr}$ /L and the removal rate of  $COD_{Cr}$  obtained from the batch methane fermentation test was 0.62, the  $COD_{Cr}$ concentration of the effluent from the AnMBR would be approximately 1,800 mg/L. The reduction in the concentrations of the ammonium and phosphate ions is expected to be negligible. As a result, the concentrations of these ions in the effluent of the AnMBR are expected to be the same as those in the condensed wastewater (i.e., 130 mg/L for ammonium Table 3

Energy balance of the system combining the wastewater condensation by the FO membrane and the anaerobic MBR

	Consumption (kWh/m³)	Generation (kWh/m³)
Condensation of wastewater	0.1	-
by FO membrane		
AnMBR (operation)	0.0032	-
AnMBR (heating)	0.60	-
Biogas power generation	-	0.33
Post-treatment (MAP and AO)	0.017	-
Total	0.72	0.33

Note: All energy values correspond to energy per 1 m<sup>3</sup> of raw wastewater without condensation. The volumetric condense factor of wastewater by FO membrane was assumed to be 19 times, and the AnMBR operational temperature was assumed to be 40°C.

ions and 78 mg/L for phosphate ions). As the concentrations of COD<sub>Cr</sub>, ammonium and phosphate ions exceed the discharge standards, applying appropriate post-treatment is indispensable. In this study, the magnesium ammonium phosphate (MAP) crystallization process [27] was selected for the recovery of phosphorous, while the anaerobic-oxic (AO) process [6] was selected for the removal of nitrogen and organic matter. According to some previous publications, the energy consumption of MAP and AO processes were reported to be 0.02 kWh/m<sup>3</sup> [28] and 0.3 kWh/m<sup>3</sup> [29], respectively. On this basis, the energy consumption for post-treatment ( $E_{C_{Post}}$ ) is estimated to be 0.32 kWh/m<sup>3</sup> (0.017 kWh/m<sup>3</sup> of raw wastewater).

The energy consumption during the up-condensation of municipal wastewater using an FO membrane was assumed to be 0.1 kWh/m<sup>3</sup> [18]. In the estimation, it was also assumed that the energy consumption of the AnMBR, excluding the energy required for heating, is 0.06 kWh/m<sup>3</sup> (0.0032 kWh/m<sup>3</sup> of raw wastewater [19]). The other parameters required for calculating the energy consumption were assumed as follows: specific heat of water ( $C_n$ ) as 1.16 kWh/m<sup>3</sup>/K; the temperature in the bioreactor of AnMBR ( $T_{AN}$ ) as 313 K; the temperature of the condensed wastewater  $(T_{in})$  as 298 K; and the efficiency of heat exchanger  $(\eta_{HE})$  as 0.7. The energy consumption of the proposed system ( $E_c$ ) was estimated to be 0.72 kWh/m<sup>3</sup> by using Eq. (8). Similarly, it was assumed that the  $COD_{Cr}$  concentration in a municipal wastewater (before condensation) is 300 mg-COD $_{\rm Cr}/L;$  the COD $_{\rm Cr}$  concentration in condensed wastewater  $C_{in}$  is 4,800 kg-COD /m<sup>3</sup>; the methane yield ( $\eta_{CH4}$ ) is 0.22 m<sup>3</sup>-CH<sub>4</sub>/kg-COD<sub>Cr rem</sub>; the heat quantity of methane gas  $(H_{CH4})$  is 36 MJ/m<sup>3</sup>-CH<sub>4</sub> [26]; and the energy conversion efficiency from the combustion of methane gas ( $\eta_{cc}$ ) is 0.8. On the basis of the assumption mentioned above, the energy generated while treating a unit volume of wastewater by the proposed system ( $E_a$ ) was estimated to be 0.33 kWh/m<sup>3</sup> by using Eq. (9). Therefore, the net energy generation  $(E_{ref})$  was calculated to be -0.39 kWh/m<sup>3</sup>, by using Eq. (7).

Unfortunately, the net energy consumption of the proposed system was larger than the typical energy consumption of conventional activated sludge process (i.e., 0.2 kWh/m<sup>3</sup> [30]). Therefore, it is necessary to reduce the net energy consumption of the proposed system. It is especially important to improve the removal rate of  $\text{COD}_{\text{Cr}}$  (i.e., increasing the amount of biogas generated) and to reduce the energy required for heating up the bioreactor of AnMBR. Watanabe et al. [31] reported that more than 95% removal of  $\text{COD}_{\text{Cr}}$  can be achieved in an anaerobic treatment process operated at a fermentation temperature of 25°C. Even if the anaerobic sludge was acclimated gradually, this  $\text{COD}_{\text{Cr}}$  removal rate can be achieved. Assuming that a 95% removal rate of  $\text{COD}_{\text{Cr}}$  was achieved and that the AnMBR could be stably operated below 34°C, the proposed system can become a net energy generation system. In their study, the feed water introduced to the anaerobic reactor was artificial wastewater. It is still unclear whether a similar performance can be achieved when actual condensed wastewater was used as feed water. However, it is believed that the proposed energy generation system will be realized in the near future.

#### 5. Conclusions

The energy balance of a wastewater treatment system involving the up-condensation of municipal wastewater by the FO membrane and the AnMBR of the condensed wastewater was evaluated. For this evaluation, condensation test using municipal wastewater by the FO membrane and batch methane fermentation test using condensed municipal wastewater were carried out. The COD<sub>Cr</sub> concentration in the wastewater reached 4,800 mg/L. It is a concentration that will be applied to anaerobic treatment. The methane yield from the condensed municipal wastewater in the batch methane fermentation test at 40°C was 0.22 m3-CH4/kg-CODCr rem. From these results, the net energy consumption of proposed system was calculated to be 0.39 kWh/m3. This value is higher than conventional municipal wastewater treatment by activated sludge process (0.2 kWh/m<sup>3</sup>). To reduce the energy consumption of proposed system, the fermentation temperature in the AnMBR should be low, because the energy of heating the AnMBR is large (0.6 kWh/m<sup>3</sup>). By our estimation, the proposed system could be a net energy producer if the fermentation temperature in the AnMBR process can be reduced to below 34°C.

#### Acknowledgments

This work has been supported by the Regional Innovation Strategy Support Program, Ministry of Education, Cultures, Sports, Science and Technology. We are deeply grateful for the kind assistance given by the Public Construction Projects Bureau of Kobe city.

#### References

- Japan for Sustainability, Japanese Municipalities Targeting Energy Self-Sufficiency at Sewage Treatment Plants, JFS Newsletter No. 71, 2008.
- [2] P.L. McCarty, J. Bae, J. Kim, Domestic wastewater treatment as a net energy producer—can this be achieved? Environ. Sci. Technol., 45 (2011) 7100–7106.
- [3] D.S. Parker, M.S. Merrill, Oxygen and air activated sludge: another view, J. Water Pollut. Control Fed., 48 (1976) 2511–2528.
- [4] D.J. Reardon, Turning down the power, Civ. Eng., 65 (1995) 54–56.
- [5] J.J. Heijnen, A. Mulder, W. Enger, F. Hoeks, Review on the application of anaerobic fluidized bed reactors in waste-water treatment, Chem. Eng. J., 41 (1989) B37–B50.

- [6] G. Tchobanoglous, H. D. Stensel, R. Tsuchihashi and F. Burton, Wastewater Engineering Treatment and Reuse, 4th ed., Metcalf and Eddy, New York, 2003.
- [7] S.K. Lateef, B.Z. Soh, K. Kimura, Direct membrane filtration of municipal wastewater with chemically enhanced backwash for recovery of organic matter, Bioresour. Technol., 150 (2013) 149–155.
- [8] T.Y. Cath, A.E. Childress, M. Elimelech, Forward osmosis: principles, applications, and recent developments, J. Membr. Sci., 281 (2006) 70–87.
- [9] K.B. Petrotos, P. Quantick, H. Petropakis, A study of the direct osmotic concentration of tomato juice in tubular membrane module configuration. I. The effect of certain basic process parameters on the process performance, J. Membr. Sci., 150 (1998) 99–110.
- [10] E.M. Garcia-Castello, J.R. McCutcheon, M. Elimelech, Performance evaluation of sucrose concentration using forward osmosis, J. Membr. Sci., 338 (2009) 61–66.
- [11] B.R. Babu, N.K. Rastogi, K.S.M.S. Raghavarao, Effect of process parameters on transmembrane flux during direct osmosis, J. Membr. Sci., 280 (2006) 185–194.
- [12] S. Lee, C. Boo, M. Elimelech, S. Hong, Comparison of fouling behavior in forward osmosis (FO) and reverse osmosis (RO), J. Membr. Sci., 365 (2010) 34–39.
- [13] Y. Gu, L. Chen, J.W. Ng, C. Lee, V.W.C. Chang, C.Y. Tang, Development of anaerobic osmotic membrane bioreactor for low-strength wastewater treatment at mesophilic condition, J. Membr. Sci., 490 (2015) 197–208.
- [14] X. Zhang, Z. Ning, D.K. Wang, J.C.D. da Costa, Processing municipal wastewaters by forward osmosis using CTA membrane, J. Membr. Sci., 468 (2014) 269–275.
- [15] S. Zhao, L. Zou, D. Mulcahy, Effects of membrane orientation on process performance in forward osmosis applications, J. Membr. Sci., 382 (2011) 308–315.
- [16] A.L. Smith, L.B. Stadler, N.G. Love, S.J. Skerlos, L. Raskin, Perspectives on anaerobic membrane bioreactor treatment of domestic wastewater: a critical review, Bioresour. Technol., 122 (2012) 149–159.
- [17] D.W. Menzel, N. Corwin, The measurement of total phosphorus in seawater based on the liberation of organically bound fractions by persulfate oxidation, Limnol. Oceanogr., 10 (1965) 280–282.
- [18] R.K. McGovern, On the potential of forward osmosis to energetically outperform reverse osmosis desalination, J. Membr. Sci., 469 (2014) 245–250.
- [19] H. Lin, J. Chen, F. Wang, L. Ding, H. Hong, Feasibility evaluation of submerged anaerobic membrane bioreactor for municipal secondary wastewater treatment, Desalination, 280 (2011) 120–126.

- [20] M. Shibya, M. Yasukawa, T. Takahashi, T. Miyoshi, M. Higa, H. Matsuyama, Effect of operating conditions on osmotic-driven membrane performances, Desalination, 362 (2015) 34–42.
- [21] G.D. Zupančič, N. Uranjek-Ževart, M. Roš, Full-scale anaerobic co-digestion of organic waste and municipal sludge, Biomass Bioenergy, 32 (2008) 162–167.
- [22] K. Kimura, R. Nishisako, T. Miyoshi, R. Shimada, Y. Watanabe, Baffled membrane bioreactor (BMBR) for efficient nutrient removal from municipal wastewater, Water Res., 42 (2008) 625–632.
- [23] G. Lettinga, A. De Man, A.R.M. Van der Last, W. Wiegant, K. Van Knippenberg, J. Frijns, J.C.L. Van Buuren, Anaerobic treatment of domestic sewage and wastewater, Water Sci. Technol., 27 (1993) 67–73.
- [24] A. Saddoud, M. Ellouze, A. Dhouib, S. Sayadi, Anaerobic membrane bioreactor treatment of domestic wastewater in Tunisia, Desalination, 207 (2007) 205–215.
- [25] D. Martinez-Sosa, B. Helmreich, T. Netter, S. Paris, F. Bischof, H. Horn, Anaerobic submerged membrane bioreactor (AnSMBR) for municipal wastewater treatment under mesophilic and psychrophilic temperature conditions, Bioresour. Technol., 102 (2011) 10377–10385.
- [26] A.D. Cuéllar, M.E. Webber, Cow power: the energy and emissions benefits of converting manure to biogas, Environ. Res. Lett., 3 (2008) 1–8.
- [27] J. Wang, Y. Song, P. Yuan, J. Peng, M. Fan, Modeling the crystallization of magnesium ammonium phosphate for phosphorus recovery, Chemosphere, 65 (2006) 1182–1187.
- [28] J.C. Van Dijk, H. Braakensiek, Phosphate removal by crystallization in a fluidized bed, Water Sci. Technol., 17 (1984) 133–142.
- [29] K. Xiao, Y. Xu, S. Liang, T. Lei, J. Sun, X. Wen, X. Huang, Engineering application of membrane bioreactor for wastewater treatment in China: current state and future prospect, Front. Environ. Sci. Eng., 8 (2014) 805–819.
- [30] R. Gnirss, J. Dittrich, Microfiltration of municipal wastewater for disinfection and advanced phosphorus removal: results from trials with different small-scale pilot plants, Water Environ. Res., 72 (2000) 602–609.
- [31] R. Watanabe, T. Sugo, S. Wakahara, Y.Y. Li, Influence of temperature on treatment performance of synthetic sewage by anaerobic membrane bioreactor (AnMBR), J. Japan Soc. Water Environ., 37 (2104) 197–204.