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Water and nutrient recovery by a novel moving sponge – Anaerobic osmotic membrane bioreactor – Membrane distillation (AnOMBR-MD) closed-loop system

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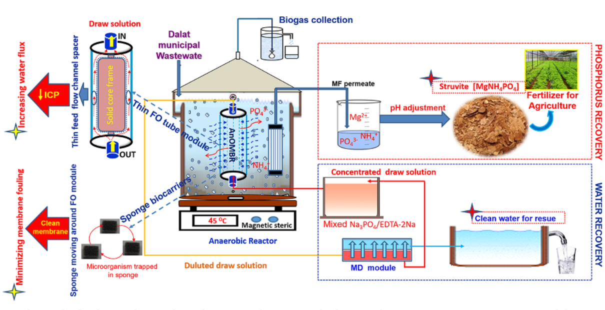
GRAPHICAL ABSTRACT



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ABSTRACT

For the first time, a novel sponge-based moving bed-anaerobic osmosis membrane bioreactor/membrane distillation (AnOMBR/MD) system using mixed Na_3PO_4/EDTA–2Na as the draw solution was employed to treat wastewater for enhanced water flux and reduced membrane fouling. Results indicated that the moving sponge-AnOMBR/MD system obtained a stable water flux of 4.01 L/m^2 h and less membrane fouling for a period lasting 45 days. Continuous moving sponge around the FO module is the main mechanism for minimizing membrane fouling during the 45-day AnOMBR operation. The proposed system's nutrient removal was almost 100%, thus showing the superiority of simultaneous FO and MD membranes. Nutrient recovery from the MF permeate was

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best when solution pH was controlled to 9.5, whereby 17.4% (wt/wt) of phosphorus was contained in precipitated components. Moreover, diluted draw solute following AnOMBR was effectively regenerated using the MD process with water flux above 2.48 L/m^2 h and salt rejection > 99.99%.

1. Introduction

Water reuse and nutrient recovery from wastewater have been considered good and rational outcomes of bolstering the world's limited freshwater and nutrient resources (Chen et al., 2018; Nguyen et al., 2016b; Nguyen et al., 2020b). Conventional activated sludge treatment processes can effectively reclaim freshwater, but they are inefficient in recovering nutrients from wastewater and generate a large physical footprint (My et al., 2017). Innovative processes to further improve water reuse and nutrient recovery from wastewater are needed to promote recycling and reuse practices (Sayi-Ucar et al., 2015). Membrane bioreactor (MBR) is an innovative process that can help advance wastewater reuse with markedly reduced space requirements (Guo et al., 2012; Ramesh et al., 2006, Luo et al., 2014). However, the traditional activated sludge-based MBR process exhibits several operational and development issues, most notably great energy consumption, membrane fouling as well as limited nutrient rejection in a single reactor (Alturki et al., 2012; Alturki et al., 2012; Jang et al., 2013; Nguyen et al., 2012).

Recently, anaerobic osmotic membrane bioreactor (AnOMBR) has been developed to overcome serious problems pertinent to the traditional activated sludge-based MBR (Gao et al., 2020). The AnOMBR process combines a conventional anaerobic bioreactor with a highly retained forward osmosis (FO) membrane in a single reactor tank to reduce the process's physical footprint compared to the traditional activated sludge method (Achilli et al., 2009; Chang et al., 2019a; Chang et al, 2019b; Cornelissen et al., 2008). In the AnOMBR process, the high retention membrane provides an additional barrier to the anaerobic biological degradation of pollutants, thus enhancing their rejection from wastewater (Li et al., 2020; Nguyen et al., 2018b; Wang et al., 2017). Moreover, the AnOMBR process can effectively decrease the energy consumption of wastewater treatment by using osmotic pressure difference between draw solute (DS) and feed solution (FS) streams across the FO membrane (Nguyen et al., 2020a).

To date, there have been a number of studies demonstrating the AnOMBR process for advanced wastewater treatment. Wang et al. (2017) and Gu et al. (2015) showed that AnOMBR system could achieve high removal efficiency with 96% of TOC, 85–98% of TP and 62.7–81.2% of NH_4^+-N. Moreover, Chang et al. (2019b) demonstrated that the up-flow anaerobic sludge blanket-osmotic membrane bioreactor/membrane distillation hybrid system could remove > 95% of NH_4^+-N and PO_4^{3-}- P. However, a major obstacle to the practical realization of AnOMBR is the lack of suitable systems designed to achieve high water flux but less membrane fouling in long term wastewater treatment operations. For example, the AnOMBR system using a submerged FO tubular module can achieve water flux of only 1.5 L/m^2 h despite deploying a high DS concentration, i.e. 1.5 M $MgSO_4$ (Chang et al., 2019a). The low water flux of this system is due to the thick feed spacer used in the traditional FO tubular design. Basically, the thick feed spacer in the FO tube module causes low cross-flow rate and low turbulence transfer nearby the surface of FO membrane, which causes a severe internal concentration polarization (ICP) effect and hence negatively affects the FO water flux (Chang et al., 2019b; Rodrigo Valladares Linares, 2014).

Besides ICP, membrane fouling is another factor limiting water flux of the AnOMBR process for the wastewater treatment applications. Membrane fouling increases the membrane resistance to water transport across the FO membrane and aggravates the external concentration polarization (ECP) effect, hence reducing the water flux through the FO membrane. During AnOMBR treatment of wastewater, biogas produced in the anaerobic biological degradation of organic pollutants can be exploited to mitigate but fails to eradicate all membrane fouling (Zou et al., 2011). Indeed, Gu et al. (2015) reported that water flux of an AnOMBR system treating a low-strength wastewater declined quickly by 74% after 16 days' operation due to membrane fouling despite continuous biogas bubbling. Recently, an up-flow anaerobic sludge-osmotic membrane bioreactor (UAS-OMBR) design combining AnOMBR with an up-flow anaerobic sludge blanket was used for municipal wastewater treatment to reduce membrane fouling (Chang et al., 2019a). However, the membrane fouling still occurred resulting in a water flux reduction by 59% after operating for 25 days (Chang et al., 2019a). Therefore, research on innovative designs to solve membrane fouling and enhance water flux is critical to realize the AnOMBR process for wastewater treatment applications.

In this study, an innovative integrated moving sponge-AnOMBR/membrane distillation (MD) system was designed to enhance water flux and minimize membrane fouling during the treatment of a real municipal wastewater. The innovative features of the system center on the

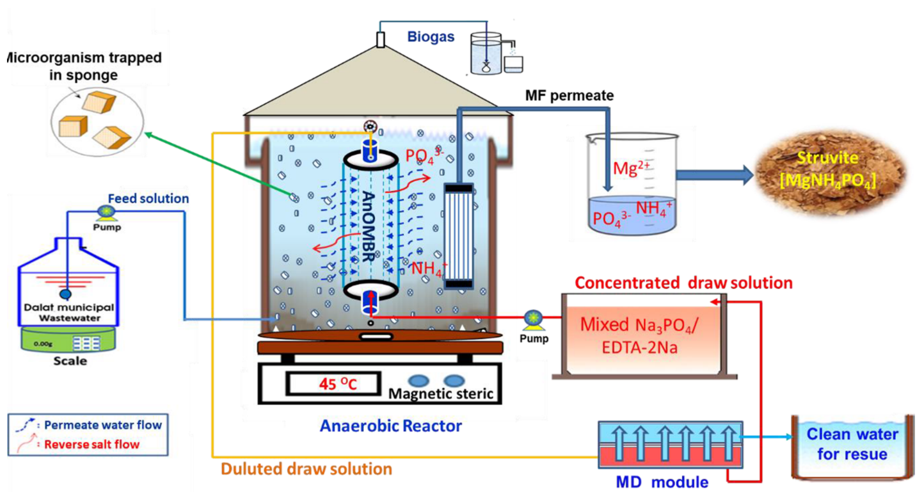

Fig. 1. A schematic of the lab scale sponge-based moving bed-AnOMBR/MD system.

new submerged FO tubular membrane module with thin feed spacers and the moving sponge. The thin feed spacers help promote turbulent water adjacent to the FO membrane to curtail the negative impact of ICP, while moving sponge carriers added into the anaerobic reactor of the AnOMBR system help clean the FO membrane. Doing so will minimize membrane fouling, thus promoting the process water flux. Moreover, the moving sponge-AnOMBR/MD system enables the wastewater to be treated simultaneously with the production of freshwater. The FO DS diluted by water permeating through the FO membrane from the wastewater is directly treated in the MD process, whereby the diluted DS is recovered in tandem with the extraction of freshwater for beneficial reuse. The heat produced from the anaerobic reactor of the integrated AnOMBR/MD system is utilized to power the MD process to decrease the energy consumption for the FO DS recovery. In other words, the moving sponge-AnOMBR/MD might be a membrane fouling-resistant and energy-efficiency to the traditional MBR system.

To the best knowledge, hardly any researchers have examined the novel moving sponge-AnOMBR/MD system using mixed Na_3PO_4/EDTA-2Na as the DS for simultaneous handling of wastewater and recovery of nutrient resource (i.e. phosphorous). Therefore, in this study the performance of the novel moving sponge-AnOMBR/MD process for wastewater treatment and nutrient recovery was symmetrically investigated. To enable the recovery of phosphorous, the AnOMBR/MD process was incorporated with a micro-filtration (MF) method to extract phosphorous in the form of struvite from wastewater. First, the positive influence of using thin spacers in the newly designed submerged FO membrane module on the process water flux was elucidated in the FO tests with deionized (DI) water as the FS. Then, an extended operation of the moving sponge-AnOMBR/MD process of a real municipal wastewater feed with the periodic MF extraction was conducted. The objective was to evaluate the process's performance with respects to permeate flux, contaminants removal, and the recovery of biogas and phosphorous.

2. Materials and methods

2.1. The moving sponge-AnOMBR/MD hybrid system

A lab-scale moving sponge-AnOMBR/MD system was designed in this study (Fig. 1). The system primarily comprised a moving sponge anaerobic reactor, a thin FO tubular module, a plate-and-frame MD module, and a hollow fiber MF module. The FO and MF membrane modules were submerged in the anaerobic reactor, while the MD membrane module was placed outside the reactor for the recovery of FO diluted DS and freshwater extraction simultaneously.

The moving sponge anaerobic reactor had an active volume of 6.0 L (internal diameter 11.3 cm and height 60 cm) and contained cubic-shaped polyurethane sponge carriers with a filling rate of 25% (by volume of the anaerobic reactor). Prior to the AnOMBR/MD hybrid process, the anaerobic reactor was acclimatized for 90 days and the biomass attached on sponge was 1.32 g biomass/g sponge. The properties of sponge carriers used in the sponge-based moving bed-AnOMBR/MD was described in the supplementary material. During the acclimatization, the wastewater was continuously pumped into the anaerobic reactor. The mixed liquor suspended solid (MLSS) in the moving sponge anaerobic reactor was maintained at 10 g/L. A magnetic stirrer was deployed to provide constant agitation in the moving sponge anaerobic reactor. The chemical oxygen demand (COD) removal by the anaerobic reactor was measured daily, and the acclimatization was terminated when the reactor COD removal stabilized and exceeded 80%.

The most notable component of the moving sponge-AnOMBR/MD hybrid system was the FO tubular membrane module. This membrane module was designed to have a thin flow channel for the DS while maintaining a large surface area of FO membrane for water to permeate through (Fig. 2). The membrane module had a solid core frame placed inside a membrane tube (i.e. made from CTA-ES membrane provided by Hydration Technologies, Inc., Albany, OR, USA) having an active membrane surface area of 120 cm^2 (diameter 2.6 cm and height 15 cm). The thickness of flat sheet FO membrane was 50 µm. The contact angles of the membrane support and active layer were 70° and 78°, respectively. The solid core frame was placed inside the flat-sheet FO membrane tube, and a plastic spacer with different thickness was inserted between the solid core frame and the membrane tube. This was done to create the thin DS flow channel of the FO membrane module.

The MD module consisted of two acrylic semi-cells and a microporous polytetrafluoroethylene (PTFE) membrane coupon inserted between them to form the feed and distillate channels. The length, width, and height of each channel were 30.0, 5.0, and 0.2 cm, respectively and the surface area of the MD membrane module was 200 cm^2. The MD PTFE membrane with pore size of 0.45 µm and contact angle of 114° provided by Ray-E Creative Co., Ltd., Taiwan. The MF membrane module used polyvinylidene fluoride (PVDF) hollow fiber membrane with pore size of 0.45 µm. The MF membrane was also provided from Ray-E Creative Co., Ltd., Taipei, Taiwan.

During the moving sponge-AnOMBR/MD process, the municipal

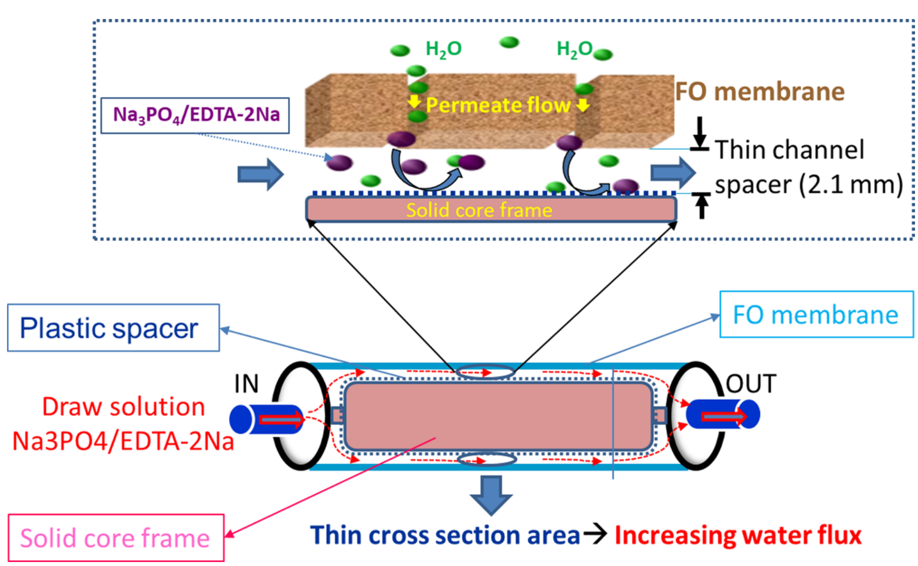

Fig. 2. The schematic diagram of the FO tubular membrane module.

wastewater from the feed tank pumped into the anaerobic reactor, but the water level in the reactor remained constant due to the float-controller. The temperature of the anaerobic reactor was maintained at 45 °C. Hydraulic retention time (HRT) is calculated by the moving sponge-AnOMBR/MD water fluxes and MF permeate fluxes with the range of 40–50 h. In this study, most of anaerobic bacterial communities attached on sponge and amount of sludge was generated in anaerobic reactor was not significant. During 45-day moving sponge-AnOMBR/MD operation, sludge was not discharged and system was operated under high solids retention time (SRT).

The mixed 0.2 M Na_3PO_4/0.25 M EDTA-2Na DS was pumped into the submerged FO tubular membrane at cross-flow rate ranging from 0.5 to 3.0 cm/s. Based on different osmotic pressures across the FO membrane, water permeated from the anaerobic reactor through the FO membrane and diluted the DS. The dilution flow of FO DS was circulated to the MD membrane module whereby it was regenerated together with freshwater extraction. The small amount of DS loss was replenished by adding 2 L of mixed 0.2 M Na_3PO_4/0.25 M EDTA-2Na DS after 10-day AnOMBR operation to maintain the DS concentration. The MD recovery of the diluted DS was controlled at distillate and feed temperature of 25 °C and 45 °C, respectively, with the heat sourced from the anaerobic reactor. Given the FO membrane's high retention, dissolved salts accumulated in the anaerobic reactor, seriously affecting the reactor's microbial activity. To mitigate this, the MF module was submerged in the anaerobic reactor to periodically extract accumulated nutrient and phosphorous from the wastewater. During the moving sponge-AnOMBR/MD experiments, characteristics (e.g.COD, total dissolved solids (TDS), NH_4^+-N, and PO_4^{3-}P) of the waters in the anaerobic reactor and the effluent tank were measured daily to evaluate the process treatment's efficiency. Water fluxes of the AnOMBR and the MD process were determined using digital balances to record the changing weight of the wastewater feed tank and the MD distillate tank.

2.2. The wastewater feed and the FO draw solution

A real municipal wastewater (i.e. collected from Dalat wastewater treatment facility in Vietnam) served as the FS for the moving sponge-AnOMBR/MD hybrid system. Properties of the municipal wastewater were as follows: 880 ± 2 mg/L of COD, 47.25 ± 0.75 mg/L of NH_4^+-N, 16.32 ± 0.18 mg/L of PO_4^{3-}P, 459 ± 5 mg/L of TSS, 825 ± 1 mg/L of TDS, 25.4 ± 0.3 mg/L of Mg^{2+} and pH of 7.3 ± 0.5. The DS was prepared by dissolving the mixture of 0.2 M Na_3PO_4 and 0.25 M EDTA-2Na in DI water at room temperature, and then stirred at least 24 h prior to the AnOMBR tests. Laboratory-grade $Na_3PO_4{\cdot}12H_2O$ and EDTA–2Na·$2H_2O$ were purchased from Sigma-Aldrich Co., Ltd., Germany.

2.3. Nutrient recovery protocol

Phosphorous and ammonium in the wastewater were recovered using the submerged MF. Due to the high FO membrane rejection, phosphorus and ammonium salts accumulated in the anaerobic reactor during sponge-based moving bed-AnOMBR/MD operation, negatively affecting the reactor microbial activity and the FO performance. On days 16, 26, 36 of the moving sponge-AnOMBR/MD operation with the real municipal wastewater, 3.0 L water was extracted from the anaerobic reactor using the MF membrane module to maintain TDS less than 6 g/L in anaerobic reactor. The MF permeate rich in phosphorus and ammonium was pH adjusted (i.e. using a 2 M NaOH solution), and slightly stirred for 90 min before proceeding through filters (pore size of 0.45 µm) to recover the struvite precipitation ($MgNH_4PO_4{\cdot}6H_2O$). These precipitates were air dried at room temperature while the filtrates were stored at 4 °C for later examination.

2.4. Calculation methods

The water flux J_w (L/m^2 h), reverse salt flux J_s (g/m^2 h) and specific reverse salt flux (J_s/J_w, g/L) were calculated according to our previous studies (Nguyen et al., 2016a, Nguyen et al., 2020a).

$$\text{Biogasproduction} = \frac{V_{biogas}}{1000xV_{eff}(COD^{inf} - COD^{eff})} \quad (1)$$

where $V_{biogass}$ is the volume of biogas; V_{eff} is the volume of treated wastewater; COD_{inf} is the COD concentration of influent stream; and COD_{eff} is the COD concentration of effluent stream.

The rejection of TDS, PO_4^{3-}-P, and NH_4^+-N and COD in moving sponge-AnOMBR/MD system was calculated according to the following equation:

$$R = (1 - \frac{C_{eff}}{C_{inf}}).\ 100\% \quad (2)$$

where R is the rejection; C_{eff} is the concentration of TDS, PO_4^{3-}-P, and NH_4^+-N and COD at the effluent stream; and C_{inf} is is the concentration of TDS, PO_4^{3-}-P, and NH_4^+-N and COD at the influent stream.

2.5. Analytical methods

The concentrations of NH_4^+-N and PO_4^{3-}-P were analyzed in an ultraviolet–visible spectrophotometer (DR-4000, Hach, Japan). COD, Mg and MLSS analyses were conducted following Standard Methods

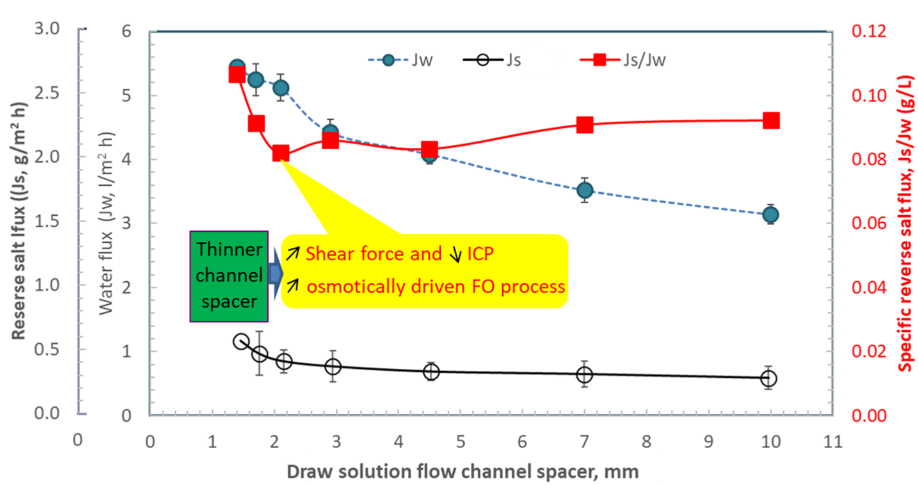

Fig. 3. Effect of the draw solution channel height on new submerged-tubular FO performance (Draw solution: mixed 0.2 M Na_3PO_4/0.25 M EDTA-2Na, Feed solution: DI, Temperature: 45 ± 2 °C, Membrane orientation: Active layer facing with feed solution. Error bars were based on the standard deviations of three replicate tests.

(APHA, 2005). pH meter (HI 9025, Hanna Instruments) was used to measure the pH in the anaerobic reactor every day and the value of ORP was measured by using a MULTI 3630 IDS –WTW, Germany. The osmometric (Model 3320, Advanced Instruments, Inc., USA) was used to measure the osmolality of DS, then using Morse equation to convert the measured osmolality of the DS to osmotic pressure as follows:

$$p = (Sf.\;\; n.\, C).\, R.\, T \tag{3}$$

where, $(\Sigma\phi.N.C)$ is total osmolality of DS, T and R are the absolute temperature *and* the universal gas constant, respectively.

The conductivity of solutions was measured by using a conductivity meter (Sension156, Hach, China) and the viscosity of solutions was determined using the Vibro Viscometer (AD Company, Japan). The MD and FO membrane's contact angle was recorded by using CAM 100 (KSV Instruments Inc., USA). Scanning electron microscopy and energy dispersive X-ray spectroscopy (SEM–EDX) were used to observe membrane fouling. The crystals were examined and identified as struvite by single X-ray diffraction (XRD).

3. Results and discussions

3.1. The positive impacts of the thin spacer on water flux of the FO process

The submerged FO module has often been used in AnOMBR application instead of external FO module since it is simple and inexpensive to operate. However, current submerged-tubular FO modules were designed with a thick channel spacer leading to low FO water flux. Hence, a thin-submerged FO tube module was designed to enhance water flux in this study. The new submerged-tubular FO module was analyzed at different thicknesses of the DS channel spacer from 10 to 1.4 mm to evaluate the positive impacts of the thin channel spacer on FO water flux. At the same flow velocity, a significant change occurred in the water flux when the thickness of DS flow channel spacer decreased from 10 to 1.4 mm using mixed 0.2 M Na_3PO_4/0.25 M EDTA-2Na as the DS (pH = 7.3 ± 0.5) and deionized (DI) water as FS (Fig. 3). Results show that water flux had increasing trends from 3.14 to 5.44 L/m^2 h (i.e. 73.25% increase) and reverse solute flux raised slightly from 0.29 to 0.58 g/m^2 h with decreasing in thickness of DS flow channel spacer from 10 to 1.4 mm. This result is consistent with Linares et al. (2014), who demonstrated that the higher water flux maintained using thicker feed spacers (1.2 mm per 46 mil) compared to thinner feed spacers (0.7 mm per 28 mil). Two interesting reasons could explain the increase in water flux as follows. Firstly, the thinner DS flow channel spacer in FO tube module leading to an increase in shear stress on membrane support layer could reduce the internal concentration polarization (ICP) of the membrane. Secondly, the narrow membrane channel in the FO tube module creating higher cross-flow velocity of DS retained more effective osmotic pressure at the boundary layer due to fast dilution of permeable flux (Abdulwahab et al., 2013). The mechanism through which an increase in water flux occurs when reducing thickness of DS flow channel spacer in the FO tube module is described in Fig. 2. Nevertheless, there was no noteworthy raise in the water flux (5.12 to 5.44 L/m^2 h) as the thickness of DS flow channel spacer dropped from 2.1 to 1.4 mm. Moreover, the narrower membrane channel in FO tube module creating higher turbulence of DS made higher salt diffusion leading to increase in salt leakage. However, the increasing reverse salt flux in this study (average of 41.38% increase) was lower than that of in increasing water flux (average of 63.06% increase). This was an interesting exploration from the thin FO tube module. As shown in Fig. 3, the lowest specific reverse salt flux of mixed 0.2 M Na_3PO_4/0.25 M EDTA-2Na DS (J_s/J_w = 0.08 g/L) at thickness of DS flow channel spacer of 2.1 mm, which demonstrated that mixed high charge salt of $EDTA^{4-}$ and PO_4^{3-} is a promising DS in AnOMBR system, particularly to curtail salt accumulation.

3.2. Membrane fouling during sponge-based moving bed-AnOMBR/MD process

Fig. 4 illustrates the various water fluxes of AnOMBR with time during the testing of the moving sponge-AnOMBR/MD system by using a mixture of 0.2 M Na_3PO_4/0.25 M EDTA-2Na as the DS and the real municipal wastewater as the FS. The observation result shows that the water flux declined from 4.55 to 3.19 L/m^2 h during 45-day AnOMBR operation due to decreased driving force as well as membrane fouling. This result agreed well with Nguyen et al. (2016) that the water flux decline from the first day to the 90th day was 11.49%. Basically, the driving force for AnOMBR water flux dropped as the salt accumulation in anaerobic reactor steadily raised due to diffusion of salts from the DS into the reactor and significant FO retention of influent salinity. The results show that the TDS in the reactor rose from 0.72 to 5.12 g/L after 45 days of AnOMBR operation, which reduced osmotic gradient between FS and DS (π_{DS}-π_{FS}) leading to dropping the water flux as equation follows:

$$J_w = A_w * (p_{DS} - p_{FS})\; withp\_DCRT \tag{4}$$

where A_w is membrane permeability coefficient, R is gas constant (R = 0.082 L atm/(mol K)), ΔC is concentration gradient of solution (mol/L), and T is absolute temperature (K).

Nevertheless, an approximately different water flux of 29.89% was recorded between the first day (4.55 L/m^2 h) and the 45th day (3.19 L/m^2 h). The slight decrease in water flux recommended that fouling of FO membrane in the moving sponge-AnOMBR is not significant because most of the bacterial cells are attached to the sponge biocarriers rather than membrane, therefore avoiding membrane fouling. Moreover,

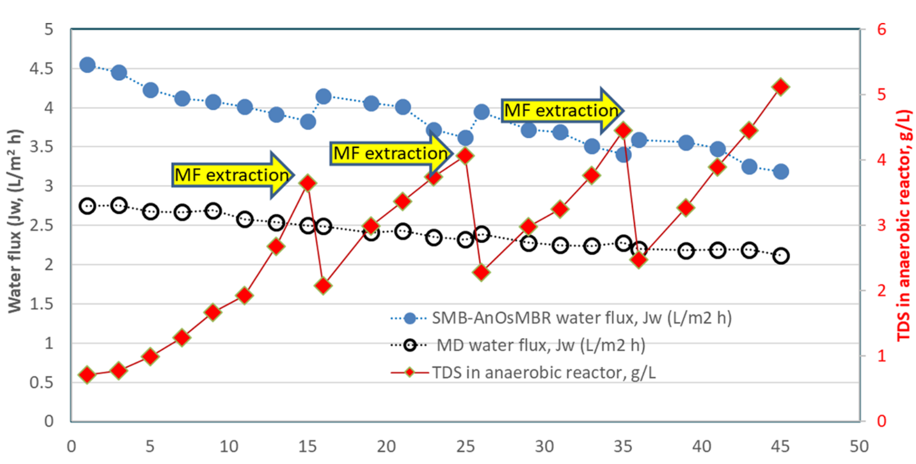

Fig. 4. Variation of water flux during sponge-based moving bed-AnOMBR/MD operation process for wastewater treatment. (Draw solution: 0.2 M Na_3PO_4 coupled with 0.25 M EDTA-2Na, Feed solution: Real municipal wastewater, Temperature: 45 ± 2 ^{0}C, Membrane orientation: Active layer facing with feed solution, Thickness of draw solution flow channel spacer of 2.1 mm).

because the moving sponge-AnOMBR system was conducted with the membrane orientation of active layer facing with FS (FO mode), deposition of foulants onto the FO surface active layer, in which it could be cleaned and removed by sponge biocarriers continuously moved around FO tube module (Mi & Elimelech, 2008; Nguyen et al., 2016a). This is an excellent way of exploring the reduction of membrane fouling in AnOMBR configuration. The water flux increased slightly at day 16 (J_w = 4.15 L/m^2 h), 26 (J_w = 3.95 L/m^2 h) and 36 (J_w = 3.59 L/m^2 h), as compared to the day before 15 (J_w = 3.83 L/m^2 h), 25 (J_w = 3.62 L/m^2 h) and 35 (J_w = 3.41 L/m^2 h), respectively, because extracting 3L of high TDS solution in anaerobic reactor and replacing 3L of low TDS solution from influent wastewater led to raising osmotic gradient between feed and DS. As compared to the previous group study, the water flux of traditional AnOMBR system dropped quickly from 1.48 L/m^2 h to 1.07 L/m^2 h after only 10 days of operation and the membrane had to be cleaned (Chang et al., 2019a). This issue demonstrated the benefits of using moving sponge-AnOMBR in reduction of membrane fouling, which is the most difficult problem of AnOMBR operation. In the continuous moving sponge-AnOMBR/MD system, diluted DS was directly regenarated by the MD process to reuse DS in AnOMBR process, whereby a waste heat from the biorector of AnOMBR hybrid system was ultized to freely supply heat for the MD recovery process. Diluted DS (mixed EDTA-2Na/Na_3PO_4) was regenerated through this process, with distillate and feed temperature of 25 °C and 45 °C, respectively. Fig. 4 shows that the water flux of MD process dropped gradually from 2.75 to 2.12 L/m^2 h after a 45-day operation. The relatively low MD water flux was due to utilize the heat source from the AnOMBR's bioreactor with low transmembrane temperature ΔT = 20 °C (Duong et al., 2015; Nguyen et al., 2018a). As seen in Fig. 4, the water fluxes of moving sponge-AnOMBR and MD process had similar decreasing trends which helped to keep the DS concentration constant.

3.3. Contaminants removal during sponge-based moving bed-AnOMBR/MD process

A novel moving sponge-AnOMBR/MD system was designed to not only reduce membrane fouling but also produce good quality water for reuse. Fig. 5 shows the variations in the rejection of TDS, NH_4^+-N, PO_4^{3-}-P and COD during moving sponge-AnOMBR/MD performance. In the hybrid moving sponge-AnOMBR/MD system, the integration between biological treatment and high retention membrane processes could achieve the high contaminants removal (> 97%) and produce high quality recycled water. As can be seen in Fig. 5A, the TDS concentration in effluent stream varied within the range of 6.8 mg/L to 12.8 mg/L with TDS removal > 97% during 45 operational days. It is worth noting that the TDS concentration in effluent stream during moving sponge-AnOMBR/MD process was less than 15 mg/L, which is regarded as good quality and reusable water. Moreover, Fig. 5B depicts the variation of NH_4^+-N concentrations in influent wastewater, bioreactor, effluent, and NH_4^+-N removal. Compared to the influent NH_4^+-N concentration of around 50 mg/L, the NH_4^+-N bulk concentrations in the anaerobic reactor were much higher (NH_4^+-N concentration ranging from 62 to 90 mg/L), indicating significant accumulation of NH_4^+-N in the bioreactor. This observation could explain that a part of the organic nitrogen in the bulk was converted and presented in the form of NH_4^+ as a consequence of the biodegradation of organic N under anaerobic conditions. This subsequently increased the NH_4^+-N concentration in the bioreactor. The NH_4^+ rejection efficiency using the FO membrane was around 75%, which retained a considerable NH_4^+-N concentration in the reactor. Additionally, the NH_4^+-N removal of moving sponge-AnOMBR/MD system was over 98% and the NH_4^+-N effluent concentration was 0.95 mg/L.

Fig. 5C confirms that the concentration of PO_4^{3-}-P in the reactor increased gradually and reached 102 mg/L which was approximately five times higher than that of PO_4^{3-}-P concentration in influent wastewater. This phenomenon could have two key explanations: (1) the salt leakage from DS (Na_3PO_4) into the anaerobic reactor increased the PO_4^{3-}-P in the reactor; and (2) the FO membrane with small pore radius of 0.37 nmled to a high rejection of PO_4^{3-}-P from wastewater, which accumulated significant amounts of PO_4^{3-}-P in the bioreactor. The moving sponge-AnOMBR/MD system removed > 95% of PO_4^{3-}-P, and this result agreed with Holloway et al. (2014). Fig. 5D presents the various COD concentration in influent, anaerobic reactor and COD removal. With influent COD ranging from 880 to 1120 mg/L, the COD concentration in the anaerobic reactor varied from 124 mg/L to 318 mg/L. Relatively high COD in the bioreactor was due to the reverse salt flux of DS containing EDTA (organic compound), hence increasing COD in the reactor. The additional-external carbon source from reverse EDTA flux is expected to supply a suitable carbon-to-nitrogen ratio for anaerobic bacterial to treat wastewater during moving sponge-

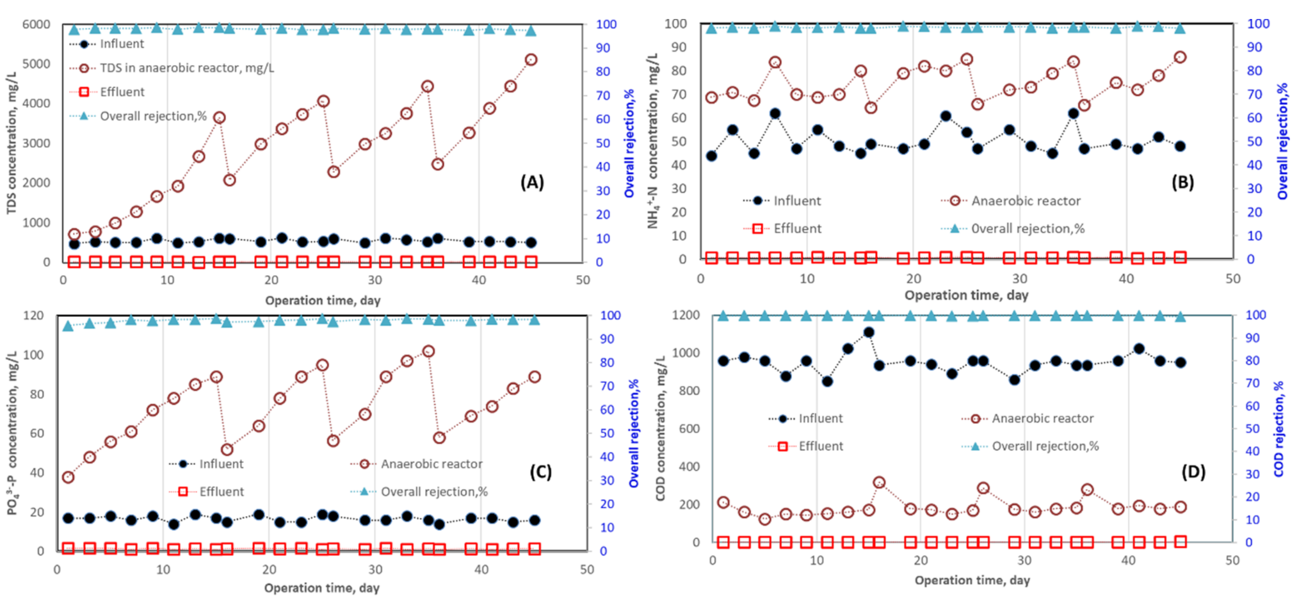

Fig. 5. Variation of contaminants removal during sponge-based moving bed-AnOMBR/MD operation process: A. TDS removal; B. NH_4^+-N removal; C. PO_4^{3-}-P removal; and D. COD removal. (Draw solution: 0.2 M Na_3PO_4 coupled with 0.25 M EDTA-2Na, Feed solution: Real municipal wastewater, Thickness of draw solution flow channel spacer of 2.1 mm).

AnOMBR operation process. Meanwhile, the COD concentration in the influent stream was relatively low, generally under 5 mg/L. Overall, the whole system demonstrated a high COD rejection of > 99% for most of the time.

3.4. Biogas and phosphorus recovery during sponge-based moving bed-AnOMBR/MD process

Fig. 6 exhibits the variation of biogas yields during the sponge based moving bed-AnOMBR/MD operation. The biogas production had decreasing trends from 0.18 to 0.11 $LCH_4/gCOD_{removed}$ within the first 15 days and then increased from 0.11 to 0.14 from day 16 onward. Within the first 15 days, the biogas yield was high with an average production of 0.16 L CH_4/g COD being removed, but this fell from day 16 onward. The main reason for reduced methane gas production was due to high salt accumulation (the salt build-up was up to 3.65 g/L). This finding agrees well with Li et al. (2017) who demonstrated that the higher reverse salt flux (or higher TDS in bioreactor) led to less methane being produced. However, the biogas yields slightly increased from day 26 onward. This phenomenon may be due to the anaerobic bacterial community in moving sponge-AnOMBR able to adapt to a high TDS state. This record demonstrated that the activity of bacterial was good and remained stable with ORP in the range of −310 to −330 mV.

As shown in section 3.3, the concentrations of NH_4^+-N, PO_4^{3-}-P were enriched in the reactor during moving sponge-AnOMBR/MD operation, leading to an increase in TDS in the reactor from 0.72 to 5.12 g/L, which is evidence of bacterial activity. MF extraction was used to blow out the high TDS solution in the anaerobic reactor at days 16, 26 and 36 with a constant MF permeate flux of 2.1 L/m^2 h. The volume was extracted of 3 L, then pH of solution was controlled to 9.5 by using 2 M NaOH, after 90-minute stirring at 180 rpm. The result recorded that the phosphorus recovery from MF permeate flux was significantly higher than nitrogen recovery. After precipitation, the concentration of PO_4^{3-}-P was quite low (around 15 mg/L) with phosphorus recovery of 82% in Table 1. This phenomenon could be explained that phosphate was easily predicated on many forms (Fig. S2). The first form was struvite ($MgNH_4PO_4.6H_2O$) and it can be observed in the XRD image as revealed in Fig. S3. Secondly, Mg^{2+} and PO_4^{3-} could be precipitated in the form of $(Mg)_3(PO_4)_2{\cdot}22H_2O$ at pH > 9, as demonstrated by Tansel et al. (2018). Thirdly, trivalent negative charge of PO_4^{3-} was easily absorbed on positively charged $Mg(OH)_2$ (s) surface at pH = 9.5, which results in an increase in phosphorus recovery (Kim et al., 2017). The precipitated solids were further employed to analyze elements using the EDX as shown in Fig. S3. The elemental composition of precipitation also highlighted a higher percentage weight of phosphorus (17.4 wt%) compared to nitrogen (1.3 wt%). Besides, the Mg element (18.7 wt%) appeared in precipitation that was possibly due to precipitations of struvite and $Mg_3(PO_4)_2{\cdot}22H_2O$ The highest precipitation amount was recorded at day 35 (0.285 g/L), followed by day 15 (0.279 g/L) and then day 25 (0.268 g/L).

4. Conclusions

A novel sponge-based moving bed-AnOMBR/MD system was successfully employed to treat wastewater for water and nutrient recovery. The study found that channel spacer of 1.4 mm facilitated a high water flux and stable mechanical of thin FO tube module. The proposed system indicated an excellent ability to reject NH_4^+-N, PO_4^{3-}-P and COD (> 99%), and this was regarded as good reuse of water. Moreover, AnOMBR attained a stable water flux of 4.01 L/m^2 h and minimal fouling because the sponge's performance as a free actively moving biocarrier in the reactor cleaned the membrane continuously. Lastly, phosphorus recovery was recorded with the highest efficiency occurring at extraction day 35 (17,4 wt%).

CRediT authorship contribution statement

Nguyen Cong Nguyen: Investigation, Writing - original draft, Methodology, Formal analysis, Data curation, Project administration. **Hung Cong Duong:** Formal analysis, Resources, Writing - review & editing. **Shiao-Shing Chen:** Supervision, Investigation, Writing - review & editing. **Hau Thi Nguyen:** Methodology, Formal analysis, Resources. **Huu Hao Ngo:** Supervision, Investigation, Conceptualization, Writing - review & editing. **Wenshan Guo:** Investigation, Writing - review & editing. **Huy Quang Le:** Methodology, Resources, Writing - review & editing. **Chinh Cong Duong:** Methodology, Data curation, Writing - review & editing. **Le Thuy Trang:** Methodology, Data curation, Writing - review & editing. **Anh Hoang Le:** Methodology, Formal analysis. **Xuan Thanh Bui:** Investigation, Validation. **Phuoc Dan Nguyen:** Methodology, Validation.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

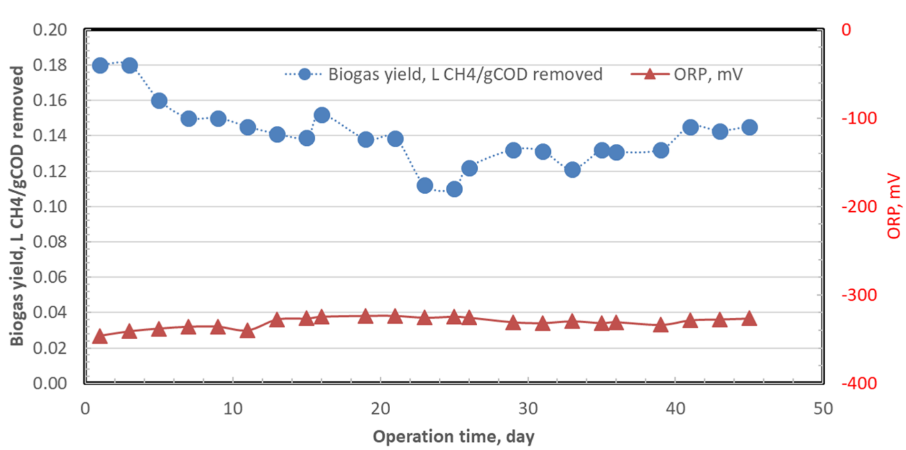

Fig. 6. Variation of biogas yield and ORP during sponge-based moving bed-AnOMBR/MD system. (Draw solution: 0.2 M Na_3PO_4 coupled with 0.25 M EDTA-2Na, Feed solution: Real municipal wastewater, Thickness of draw solution flow channel spacer of 2.1 mm).

Table 1
The variation of NH_4^+-N and PO_4^{3-}-P in bioreactor, MF effluent and after precipitation.

Extraction time, day	NH_4^+-N (mg/L)			PO_4^{3-}-P (mg/L)			Weight of precipitation, g/L
	Enriched in reactor	MF effluent	After precipitation	Enriched in reactor	MF effluent	After precipitation	
15	80.16 ± 0.95	75.12 ± 0.25	58.16 ± 0.39	89.39 ± 0.44	70.23 ± 0.45	13.22 ± 0.68	0.279 ± 0.026
25	85.24 ± 0.56	82.32 ± 0.28	63.25 ± 0.95	95.13 ± 0.38	76.32 ± 0.48	15.24 ± 0.79	0.268 ± 0.022
35	**87.15** ± 0.27	**79.21** ± 0.72	**61.27** ± 0.45	**102.34** ± 0.98	**89.11** ± 0.95	**16.15** ± 0.22	**0.285** ± **0.024**

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.biortech.2020.123573.

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