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Submerged versus side-stream osmotic membrane bioreactors using an outer-selective hollow fiber osmotic membrane for desalination

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Abstract

This study investigated the comparative performances, fouling mitigation efficiencies, and operational costs of side-stream and submerged osmotic membrane bioreactors (OMBR) systems using an outer-selective hollow fiber thin-film composite forward osmosis (OSHF TFC FO) membrane. Generally, the submerged OMBR system exhibited the higher fouling mitigation efficiency and a much slower flux decline rate when compared with that of the side-stream system. The side-stream OMBR system demonstrated an initial water flux of 15.8 LMH using 35 g/L NaCl as the draw solution, which was 2-fold higher than that of the submerged system when at its optimal performance. However, salinity accumulation in the reactor of the side-stream system was at a higher rate than for the submerged OMBR system. Both OMBR systems showed comparably high pollutant removal efficiencies over the experimental period. Annual operating costs for the side-stream OMBR system has been estimated to be 38% higher (OPEX) than for the submerged system. Membrane replacement cost accounted for the majority of the OPEX, over 89%, while the energy consumption and cleaning costs only accounted for relatively small portions. Therefore, reducing the membrane replacement cost is critical to realizing the commercial viability of the submerged OMBR system.

Keywords: *Submerged module, Side-stream module, Outer-selective hollow fiber; Membrane fouling, Operating cost, Osmotic membrane bioreactor.*

1 Introduction

Osmotic membrane bioreactors (OMBR) have recently attracted significant research interests for use in wastewater reclamation and desalination applications [1, 2]. While many of obvious advantages (i.e., high quality permeate, suitable for nutrient recovery) have been confirmed by previous OMBR studies [3], membrane fouling still remains a key concern [4-6]. Membrane fouling is an unavoidable issue in any membrane filtration processes, causing water flux and water quality to decline and may also reduce membrane lifetime, requiring more frequent cleaning and replacement, thereby increasing operational cost [7]. Therefore, effective fouling mitigation methods are essential to maintain a sustainable operation of forward osmosis (FO) processes and OMBR system. Recently, membrane fouling strategies have been assessed, including: investigation of suitable protocols for effective membrane cleaning methods; optimization of operating parameters related to membrane fouling including operating flux, draw solution (DS) properties, membrane orientation, optimum cross-flow velocity (CFV), specific aeration demand (SAD); and the application of high anti-fouling membranes and creative membrane and membrane module configuration designs. [8].

A large number of recent OMBR studies have applied flat-sheet membrane module as their main configuration [2, 3, 7, 9-12] while comparatively fewer studies have utilized FO membranes with hollow configurations [13-15]. As for the hollow fiber FO membrane, the polyamide selective layer can be coated either on the lumen side, which is called inner selective hollow fiber (ISHF), or on the outer surface of the fiber to form an outer-selective hollow fiber (OSHF). Whilst Zhang et.al [13] have used ISHF FO membranes for their OMBR study, Tran et. al [14] applied an OSHF FO membrane to investigate the fouling mitigation efficacy of the OMBR system. Compared to ISHF membranes, OSHF membranes offer several merits, including larger membrane surface area, lower fouling potential, and easier cleaning under the active layer facing FS (AL-FS) orientation [16-18]. When the ISHF FO membrane is used under the AL-FS orientation, FS (activated sludge) is recirculated on the lumen side of the fiber, which can cause severe clogging and blocking inside the micro-sized hollow fiber due to the suspended flocs, particulates, and foulants present. Under this membrane orientation, the OSHF FO membrane is more suitable as the FS is circulated outside of the fiber, and therefore not only prevents clogging and blockage inside the fiber, but also offers a more favorable condition for membrane cleaning. As fouling occurs, a fouling-cake layer can be formed on the outer surface of the fiber which can be easily and effectively mitigated by physical cleaning (i.e., using high shear force generated by elevated CFV), air scouring, and/or chemical cleaning.

Side-stream (external cross-flow) modules and submerged (plate-and-frame) modules are the two main configurations in which submerged modules have been widely applied for both conventional MBR and OMBR studies [9, 19]. In the external cross-flow module, activated sludge flows through the membrane module and is recirculated back to the bioreactor while the submerged module is directly immersed into the bioreactor, and therefore, in direct contact with the activated sludge. The application of the external cross-flow module in OMBR systems is expected to provide favorable conditions for controlling membrane fouling and reducing the effect of external concentration polarization (ECP) by simply enhancing the CFV of the FS stream. Also, chemical cleaning can be carried out in-place (CIP) in the side-stream module with minimum interruption and downtime. Nonetheless, the disadvantages of side-stream OMBR system is the high operational cost due to the additional pumping energy required for the activated sludge. Further, the strong pumping shear could break the flocs formed in the bioreactor resulting in a lower sludge yield and reduced chemical oxygen demand (COD) removal rate [20].

Submerged OMBR systems do not require circulation of activated sludge, thereby eliminating the pumping energy and also the adverse impact on microorganism growth and treatment activities [20]. Although the fouling potential of submerged MBR system is higher than the crossflow side-stream system, however, submerged OMBR systems can utilize the air bubbles used for aeration as a measure of continuous air scouring to minimize fouling and ECP mitigation [9, 21]. Nevertheless, CIP is not feasible for submerged OMBR and membrane cleaning will definitely require the interruption of the system's operation.

Application of external cross-flow (side-stream) or submerged membrane modules for OMBR systems has both pros and cons that significantly influence the fouling mitigation efficacy and overall system performance. It is therefore essential to systematically investigate and compare the performance of these two OMBR system configurations. This study aims to evaluate and compare the fouling mitigation efficiency and operational costs of the two OMBR systems using side-stream and submerged membrane modules. Performance was assessed based on water flux, salt accumulation in the activated sludge, and the pollutant removal efficacy. Fouling mitigation efficiency was evaluated according to the flux decline rate over the testing period and flux recovery after the implementation of a fouling cleaning method. Membrane autopsies were then conducted on the fouled membranes with a scanning electron microscope (SEM) and an energy diffusive X-ray (EDX) to obtain the surface and cross-sectional morphologies, and the elemental compositions of fouling cake-layer for a better understanding of the membrane fouling and the mitigation efficacy.

2 Materials and methods

2.1 Side-stream and submerged membrane module.

This study used submerged and side-stream modules with the same membrane surface areas, comprised of OSHF TFC FO membranes, fabricated and developed at the Center for Technology in Water and Wastewater, University of Technology Sydney, Australia. Specifications of the lab-scale side-stream and submerged membrane modules are described in **Fig. S1** and **Table S1**. The property of the OSHF TFC FO membrane have also described elsewhere in our previous studies [14, 22].

2.2 Draw solution and synthetic wastewater.

The chemicals used were all of reagent grade, supplied by Merck, Australia. Sodium chloride (NaCl) solution with a concentration of 35 g/L was used as the DS. Synthetic wastewater was prepared using the same recipe as the one described in our previous study [14].

2.3 Experiment protocols

2.3.1 **Experiment setup for baseline tests**

Schematics of the FO testing systems for the side-stream and submerged membrane modules are depicted in **Fig. S2**. The objective of these tests was to verify the performance of each module before installation into the OMBR systems to allow for comparison of the performance of the two membrane module configurations. These tests used sodium chloride (NaCl) solution with a concentration of 35 g/L as DS and deionized (DI) water as the FS. Water flux (J_w) and specific reverse solute flux (SRSF) were two main parameters used for verifying modules' performance. Testing conditions are tabulated in **Table. S2**.

2.4 *The bench-scale submerged and side-stream OMBR systems*

This research work used two lab-scale OMBR systems, with side-stream and submerged membrane modules, as depicted in **Fig. 1**.

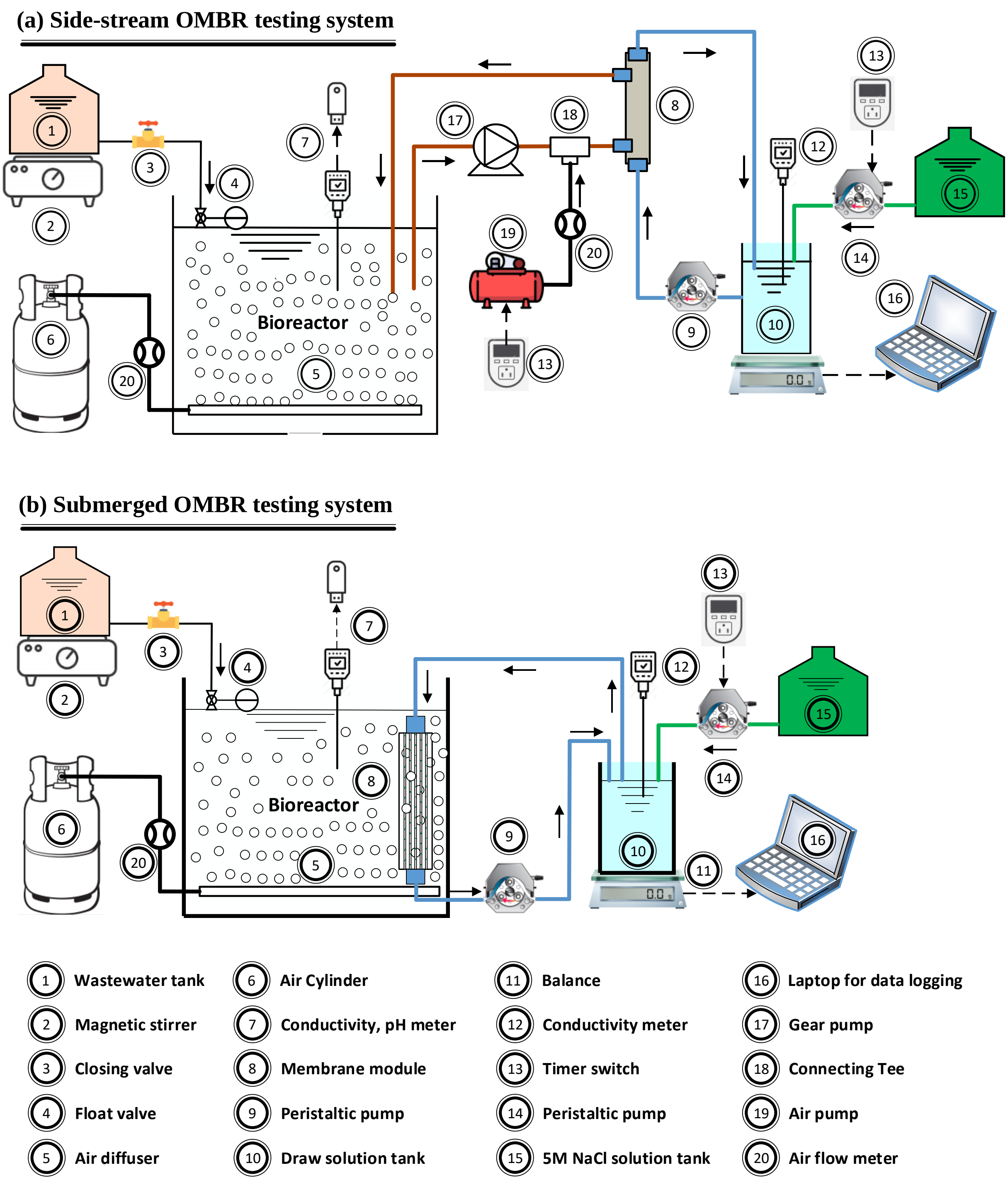


Figure 1. Bench-scale OMBR systems using side-stream and submerged membrane modules.

2.4.1 Operations of submerged and side-stream OMBR systems

The two OMBR systems in this study employed acclimatized activated sludge obtained from the water recycling facility at Sydney Central Park. Before being used for the experiments, activated sludge was acclimatized for more than 6 months, until the achieved TOC removal efficiency was consistently over 90%. A volume of 1.5 L with a mixed liquor suspended solids (MLSS) concentration of 6.5 ± 0.2 g/L, and total dissolved solids (TDS) of 0.53 ± 0.05 g/L was poured into the reactor of each OMBR system. A floating valve was installed in the reactor of each system to control the incoming volume of synthetic wastewater influent and to maintain a constant water level in the reactor. An air diffuser (Aqua One, Australia) was employed in each OMBR system reactor to provide aeration with an intensity of 3 L/min, maintaining the dissolved oxygen concentration in the bioreactor at a level of more than 3 mg/L for microorganisms. A portable pH and conductivity meter - HQ40D (HACH, Germany) was used in each system to regularly monitor the salinity and pH of the activated sludge in each reactor.

DS was recirculated from the membrane modules back to the DS tank by a peristaltic pump (LongerPump, USA). For the submerged OMBR system, DS was circulated at a flowrate of 10 ml/min through the submerged membrane module in sucking mode (the submerged module was connected to the suction line of the pump). Regarding the side-stream OMBR system, DS was pumped at a flowrate of 21 ml/min into the cross-flow module under the pushing mode, (side-stream module was connected to the pushing line of the pump). A DS concentration-controlling unit, including a conductivity probe and a programmable timer switch connected to a peristaltic dosing pump (LongerPump, USA) was used for maintaining a constant DS concentration at 35 ±

1 g/L by automatically supplementing a highly concentrated (5M) NaCl stock solution into the DS tank. As for the side-stream OMBR system, a gear pump (Cole-Parmer, USA) was used to circulate the activated sludge at a flowrate of 1 L/min from the reactor to the side-stream module which was then returned to the reactor. The solid retention time (SRT) was maintained at 30 days for both OMBR systems, with a daily discharge of 50 ml of mixed liquor. As for the side-stream OMBR system, aeration of 3 L/min was injected together with the cross-flow of the activated sludge to the membrane module for 5 minutes every two hours as a fouling mitigation method. All experiments were conducted in the laboratory, under a highly controlled environment with an ambient temperature of 22 ± 1 ˚C.

Daily physical cleaning was applied to the side-stream membrane module for 15 minutes over the course of the experiment. Physical cleaning involved a crossflow circulation of DI water into the side-stream module at 1.5 L/min with simultaneous aeration at 3 L/min to remove any fouling cake layer that may have formed on the outer surface of the hollow fiber membrane. Physical cleaning was also applied to the submerged module only when water flux dropped by 40% since the surface of the submerged membrane was in constant contact with air bubbles in the submerged OMBR system. The submerged membrane module was removed from the system and was installed in a cleaning tank containing DI water with continuous aeration at a rate of 3 L/min for 15 minutes.

2.5 Analytical methods

2.5.1 **Determination of water flux and specific reverse solute flux**

Water flux - J_w (L/m^2 h - LMH) was calculated by Equation [1]:

$$J_w = \frac{\Delta V}{A_m \times \Delta t} \qquad [1]$$

Where: A_m (m^2) is an effective area of FO membrane; Δt (h) is time interval; ΔV is the net volume change of DS solution (L). When DI water is used as FS, reverse solute flux - J_s (g/m^2 h – gMH) was calculated using Equation [2]:

$$J_s = \frac{\Delta V \times \Delta C_t}{A_m \times \Delta t} \qquad [2]$$

Where: ΔV and ΔC_t are the net changes in the FS volume (L) and FS's salt concentration; Δt (h) and A_m (m^2) are the same as in Equation [1]. Subsequently, the specific reverse solute flux (SRSF) was determined by Equation [3]:

$$SRSF = \frac{J_s}{J_w} \quad [3]$$

2.5.2 **Determination of water quality parameters and pollutant removal efficiency**

COD, MLSS, and mixed liquor volatile suspended solids (MLVSS) were measured according to standardized methods for the examination of water and wastewater [23]. Measurement of dissolved oxygen (DO) was carried out using a DO meter (Vernier, USA). Samples were regularly collected from the synthetic wastewater tank, the reactor, and the DS tank for analysis of the basic pollutant concentrations. Measurements of TOC concentrations in the collected samples were carried out using the TOC analyzer (Multi N/C 2000, Analytik Jena GmbH, Germany). Concentrations of NH_4^+, TN, and PO_4^{3-} were measured using the corresponding test kits and photometer - Spectroquant, NOVA 60 (Merck, Australia). In order to attain accurate analytical values, samples were pretreated, followed by dilution, if necessary, to minimize the interference of chloride and to ensure the proper range of analytes. Pollutant removal efficiencies (R_{eff}) of the OMBR systems were calculated using the following equations:

$$R_{eff}(\%) = \left(1 - \frac{C_{DS} \times DF}{C_{ww}}\right) \times 100 \quad [4]$$

where C_{DS} and C_{ww} are the concentrations of pollutants in mg/L of draw solution and synthetic wastewater, respectively. DF is the dilution factor taking into account the volume of permeate, which is calculated by Equation [5].

$$DF = \left(1 + \frac{V_{Per}}{V_{DS}^{i}}\right) \qquad [5]$$

where V_{DS}^{i} and V_{Per} are, respectively, the initial volume of DS volume of permeated water through the FO membrane at the time when samples are collected.

2.6 Membrane and fouling cake layer characterization

Fouled membrane samples were taken from the submerged and cross-flow module at the end of each experiment for analysis of their morphological structures and elemental compositions. Samples were dried at ambient temperature (22 ± 1 ˚C) before being coated with platinum in a high vacuum sputter coater (EM ACE600, Leica). Subsequently, membrane samples were analyzed in a field emission scanning microscopy and an energy diffusive X-ray (EDX) analyzer (FE-SEM, Zeiss Supra 55VP, Carl Zeiss AG).

2.7 Operating cost analysis

Operating costs (OPEXs) of the two OMBR systems were estimated and analyzed for economic comparison between the two OMBR membrane module configurations. The boundary of the OPEX calculation is presented in **Fig. S3**. OPEXs were calculated based on the following considerations and assumptions:

- OPEX cost were calculated assuming full-scale OMBR systems with the same capacity of 24,000 m^3/day, with effluent from both OMBR systems having a similar TDS of 3.6 ± 0.1 g/L.
- OPEXs in this study were comprised three main costs components: energy consumption (EC); membrane replacement (MR); and cleaning chemicals (CC).

- EC costs included pumping energy for the circulation of FS and regular injection of air bubbles into the membrane module (for the side-stream OMBR system only) and DS pumping for both OMBR systems. MR cost is calculated with an assumed lifetime of 7 years for submerged membrane module and 5 years for side-stream membrane module, (membrane lifetime is reduced because of additional shear force and high CFV with pressure in the side-stream module). CC cost includes energy consumption for pumping water, recirculating water during physical and chemical cleaning processes, air pumps, water cost, and the chemical costs for NaOH and citric acid.
- OPEX cost calculation in this study excludes the following costs: construction, infrastructure system, accessories, transport, used membranes, maintenance costs, aeration cost for maintaining dissolved oxygen for microorganism, and waste management.
- Energy costs were calculated assuming an electricity price of AU $0.29/kWh [24], a water price of AU $1.97/1kL, prices of NaOH and Citric acid are AU $100/ton and AU $840/ton, respectively.
- The costs of the outer selective hollow fiber FO membrane module were assumed to be AU $1,250 for the side-stream membrane module [24], and AU $625 for the submerged one. The higher cost for the side-stream membrane module is due to the costs for housing materials and the additional complexity in production. Specifications of the assumed commercial membrane modules are presented in Table S3.
- Average water fluxes, used for calculation of operational cost, were taken from the performance of each membrane module in each OMBR system.

- Daily physical cleaning is assumed to be applied to the side-stream membrane module, and a fortnightly physical cleaning is applied to the submerged membrane module. A quarterly chemical cleaning is carried out for both side-stream and submerged membrane modules.
- Chemical cleaning was assumed to be performed with an alkaline-acidic cleaning protocol for both the submerged and side-stream membrane modules. Initially, physical cleaning using clean water and aeration with an intensity of 3 L/min was carried out for 5 minutes. Next, NaOH solution (0.1% w/v, at pH 12) was then used for alkaline cleaning for 30 minutes. The first step was then repeated for 5 minutes. Subsequently, acidic cleaning was carried out using a citric acid solution (2% w/v, pH 3) for 30 minutes. Finally, the first step was repeated for 5 minutes for cleaning the membrane module and remove residual of the citric acid solution before membrane modules are brought back to operation.

3 Results and discussion

3.1 Baseline performance of side-stream and submerged modules

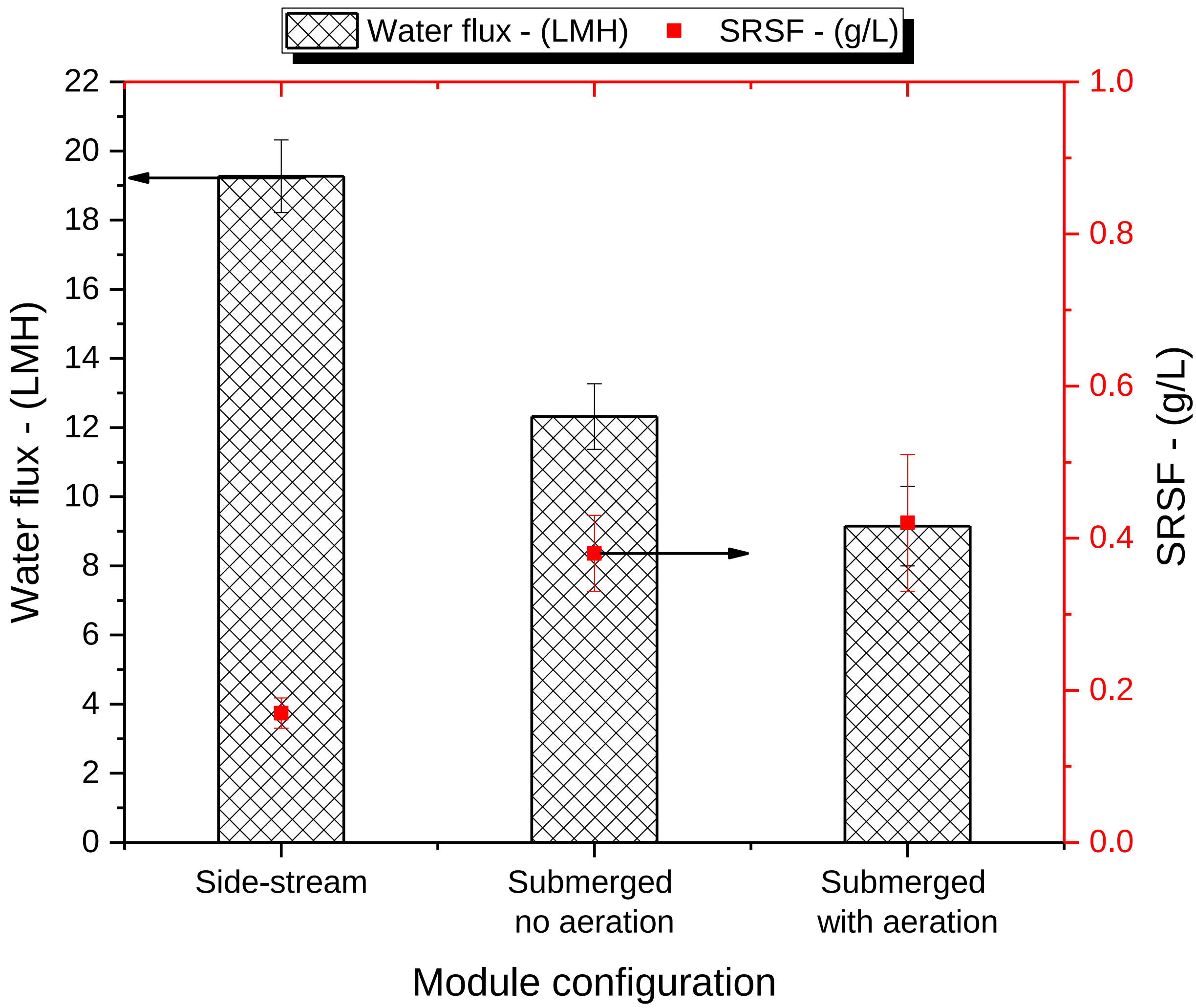


Figure 2. Performance (J_w and SRSF) of the OSHF TFC FO membrane module with side-stream and submerged configurations. Testing conditions: FS = DI water; DS = 35 g/L NaCl; AL – FS orientation; Ambient temperature (22 ± 1℃).

Fig. 2 shows the performance (J_w and SRSF) of the two membrane modules under cross-flow and submerged configurations using the OSHF TFC FO membrane under the initial test with DI water as FS. As clearly indicated in the figure, the side-stream module exhibited higher water flux (19.3 LMH) and an SRSF (0.17 g/L) when compared to the submerged membrane module with a J_w of 12.3 LMH and an SRSF of 0.38 g/L. This result is in agreement with the one observed by Blandin et. al [25] in their performance comparison of submerged and cross-flow membrane modules using a flat-sheet TFC FO membrane. The lower performance of the submerged membrane module could be ascribed to a more severe concentrative external concentration polarization (CECP) effect on the submerged system. Different operating conditions between the two systems resulted in distinct hydrodynamic behaviors in each system, which resulted in the dissimilar CECP effects. Moreover, recirculation of FS inside the side-stream module with a CFV generated turbulence and a shear force on the outer surface of each fiber, mitigating the severity of the CECP effect. As for the submerged system, while stirring was carried out in the FS tank, the effectiveness of CECP mitigation might not be as high as that in the side-stream system because less shear force and turbulence were made under the absence of CFV. CECP is therefore likely to be a more serious nearby the outer surface of hollow fiber where the reverse diffusion of draw solutes occurs.

Subsequently, continuous aeration with the same intensity as the one used in OMBR systems was sparged into the FS tank to investigate the effect on the performance of the submerged membrane module. As can be seen in **Fig. 2**, interestingly, J_w was only 9.2 LMH, which is 33% lower than without aeration. Under the continuously aerated condition, CECP might have a reduced impact on the performance of the membrane module since aeration generates secondary flows with wakes and shear forces near the membrane surface, destabilizing CECP. Moreover, air bubbles make direct contact, scouring onto the membrane surface, causing hollow fibers to vibrate, thereby mitigating the CECP effect [26]. However, intensive aeration might result in the substantial presence of air bubbles rather than the water on the membrane surface, reducing the contact area between water and membrane surface, deteriorating water flux [25]. This could be the main factor contributing to the lower J_w as aeration was introduced.

A lower SRSF value was observed with the side-stream module, which can be ascribed to the combined effect of increased water permeability and hindrance of reverse solute flux (J_s - RSF) in the side-stream system. SRSF is the ratio of J_s over J_w; SRSF will therefore be lower with a higher J_w. Additionally, reverse diffusion of draw solutes to the FS might also be impeded by the slightly higher hydraulic pressure generated by the recirculation pump on the FS side of the hollow fiber membrane in the side-stream module [9]. The decreased SRSF was in agreement with the findings reported in previous FO study conducted by Morrow et. al [9] and other pressure-assisted forward osmosis research works [27-29].

3.2 Performance of submerged and side-stream modules in OMBR systems

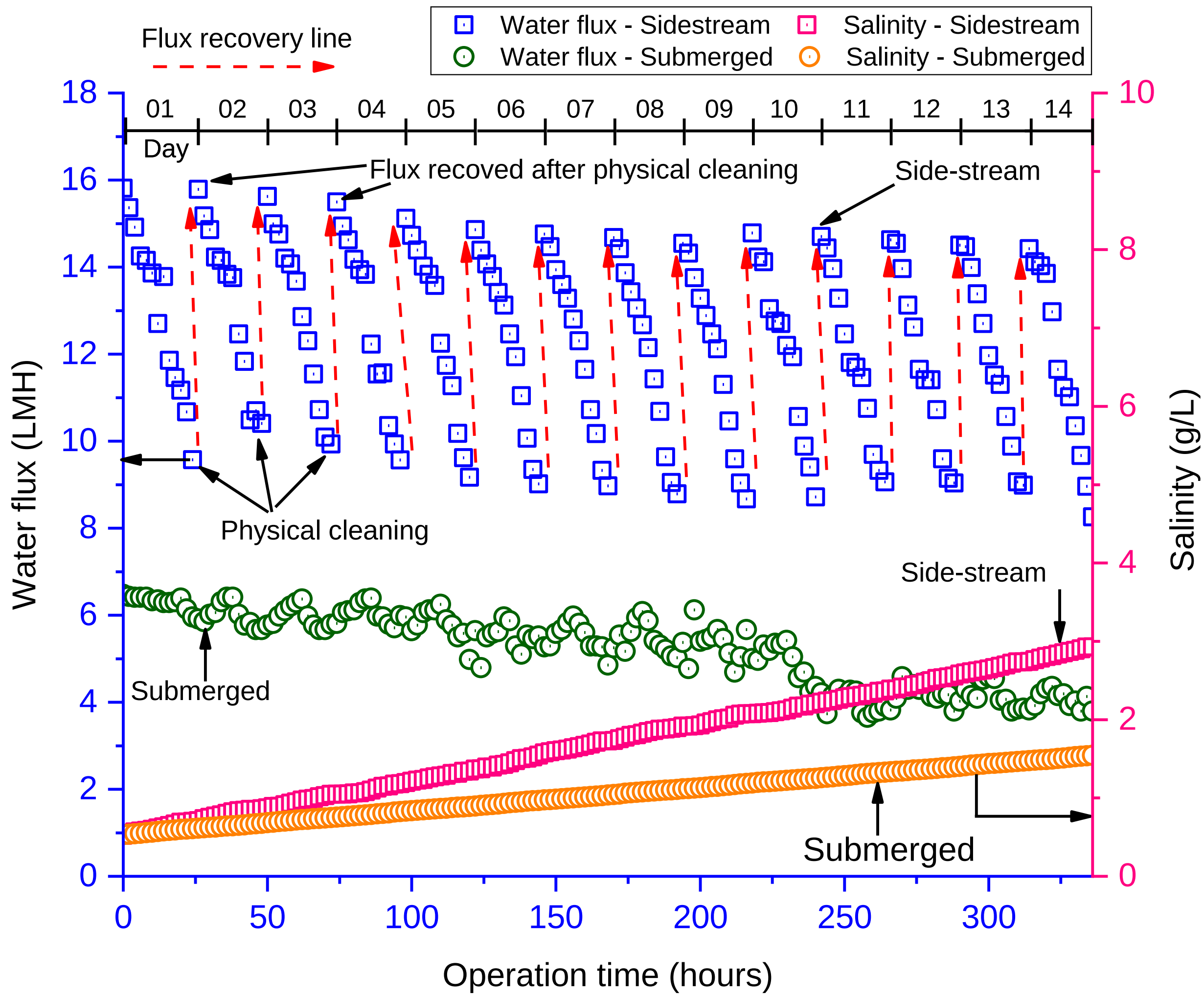


Figure 3. Water flux and salinity profiles of the reactor in two submerged and side-stream OMBR systems. Testing conditions: DS = 35 g/L NaCl; FS = activated sludge; AL – FS orientation; Ambient temperature (22 ± 1 ℃).

The profiles of J_w and the bioreactor's salinity in the submerged and side-stream OMBR systems over the experiment period are illustrated in **Fig. 3**. Similar to the earlier results from the initial baseline tests, the side-stream OMBR system produced a much higher J_w when compared to that of the submerged system. Initial J_w in the crossflow OMBR system was 15.8 LMH, about 2.5 times higher than that of the submerged OMBR system which demonstrated an initial J_w of 6.5 LMH.

The higher J_w in the side-stream system might be due partly to the effect of the slightly higher hydraulic pressure induced by the recirculation pump on the FS side, facilitating the permeation of water to the DS side. Furthermore, the intermittent injection of air bubbles into the side-stream module also likely avoided the presence and direct contact of air bubbles directly on the hollow fiber membrane surface, thereby maintaining the effective membrane surface intact. It is difficult to provide a fair comparison of the effect of CECP between these two systems, however, it is undeniable that wall shear force and the turbulence generated by the circulation with periodic aeration of FS, and continuous aeration in the side-stream module and submerged module, respectively, are different. However, both were found to have positive impacts on alleviating CECP and membrane fouling.

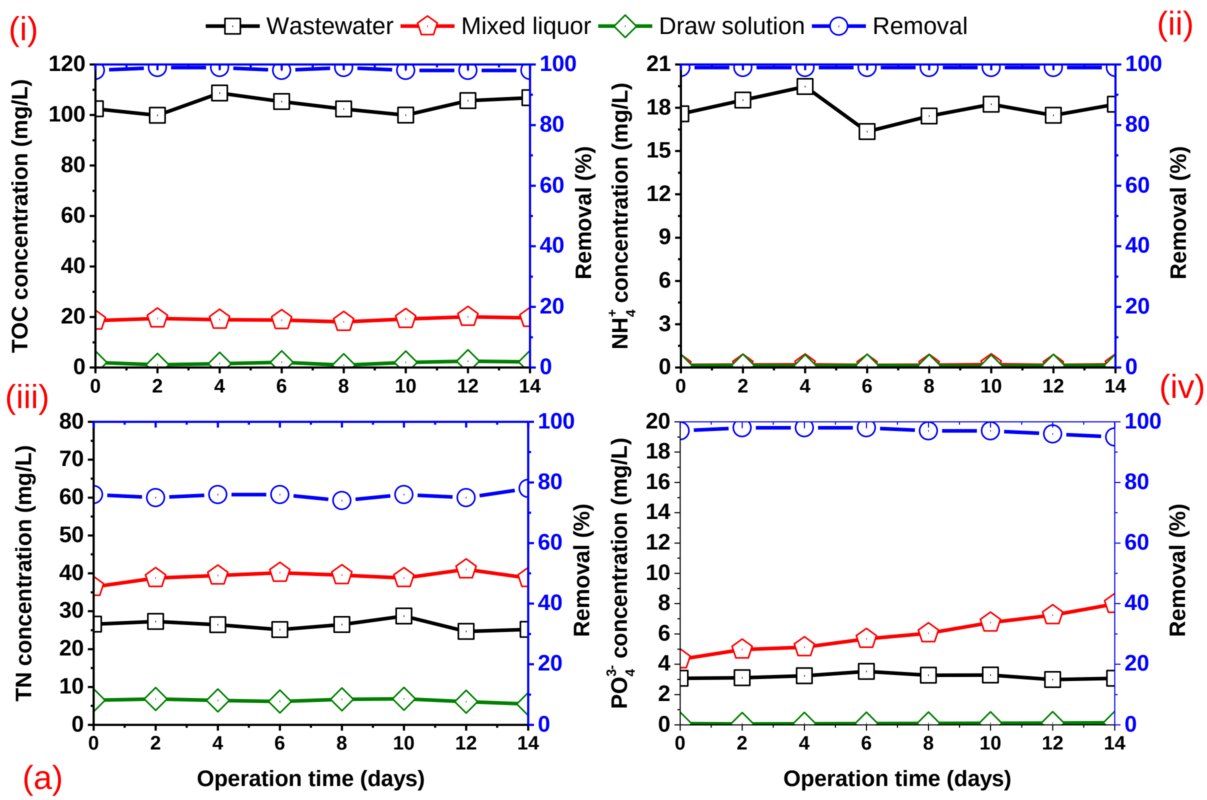
The J_w in the side-stream system significantly declined by 40% over 24 hours of operation, while the drop was only 8% for the immersed OMBR system. The difference in the flux decline rate between the two OMBR systems was attributed to the different fouling behaviors and probable intense aeration caused by the different hydrodynamic conditions, such as the initial J_w and aeration rate in these two systems. The rate of membrane fouling was significantly dependent on the initial J_w and as fouling rate was is generally more severe at higher initial J_w values, membranes are generally operated below this critical flux to lower the fouling rate [30]. As a larger volume of water permeates through the membrane, under the effect of convection, more foulants are carried towards the membrane surface, thereby inducing a greater hydrodynamic drag force towards the membrane active surface [31]. Additionally, the side-stream module might have been operated with the initial J_w above the critical flux which perhaps exacerbated the rate of membrane fouling [32].

The aeration rate also has considerable influence on fouling tendency of both conventional MBR [26, 33-35] and OMBR systems [15, 21]. Qin et. al [21] reported that for a continuously aerated OMBR system the flux decline was 30% lower compared to an OMBR without aeration. While aeration can help to mitigate membrane fouling, however, intense aeration might unintentionally cause a decrease in the effective membrane area thereby compromising the water flux.

Rapid flux decline and high flux recovery happened repeatedly over 14 days of the experimental period for the side-stream OMBR. **Fig 3** shows that the OMBR water almost fully recovered (99% of initial flux) for the side-stream module after physical cleaning at the end of each cycle. The high flux recovery rate in the side-stream system indicates that fouling in this system is reversible, and physical cleaning was effective in mitigating membrane fouling, which agrees well with our previous side-stream OMBR study [15]. Meanwhile, J_w for the submerged OMBR system continuously and slowly decreased without any physical cleaning and this indicates that continuous aeration at an optimum rate could be used as an effective measure of fouling control in a submerged OMBR system.

Fig. 3 also illustrates profiles of the reactor's salinity for the two OMBR systems over the 14-day operation. In general, the salinity of the activated sludge in both OMBR systems successively and slowly increased with a faster build-up rate of the reactor's salinity in the side-stream system. The initial values for the total dissolved solids (TDS) in both systems were comparatively similar at around 0.53 g/L at the beginning, but the salinities of their mixed liquors increased significantly throughout the testing period. The TDS value of activated sludge in the submerged system was almost 2-fold lower than that of the side-stream system at the end of the 14-day operation. This faster rate of draw solute accumulation in the side-stream system might be primarily attributed to the higher J_w of the system. The build-up of salinity in reactors is an unavoidable phenomenon in any OMBR system, caused by two main factors: cumulative accumulation of rejected feed salt or brine in the bioreactor as only pure water is drawn out by the high rejection FO membrane, and partly the reverse diffusion of the draw solute towards the bioreactor feed [3, 8, 14]. The rise in the reactor's salinity not only lowers the net osmotic driving force, exacerbating the flux decline, but can also impede the growth and functional activities of microorganisms, deteriorating the microbiological activities for wastewater treatment [8].

3.3 *Pollutants removal efficiency*



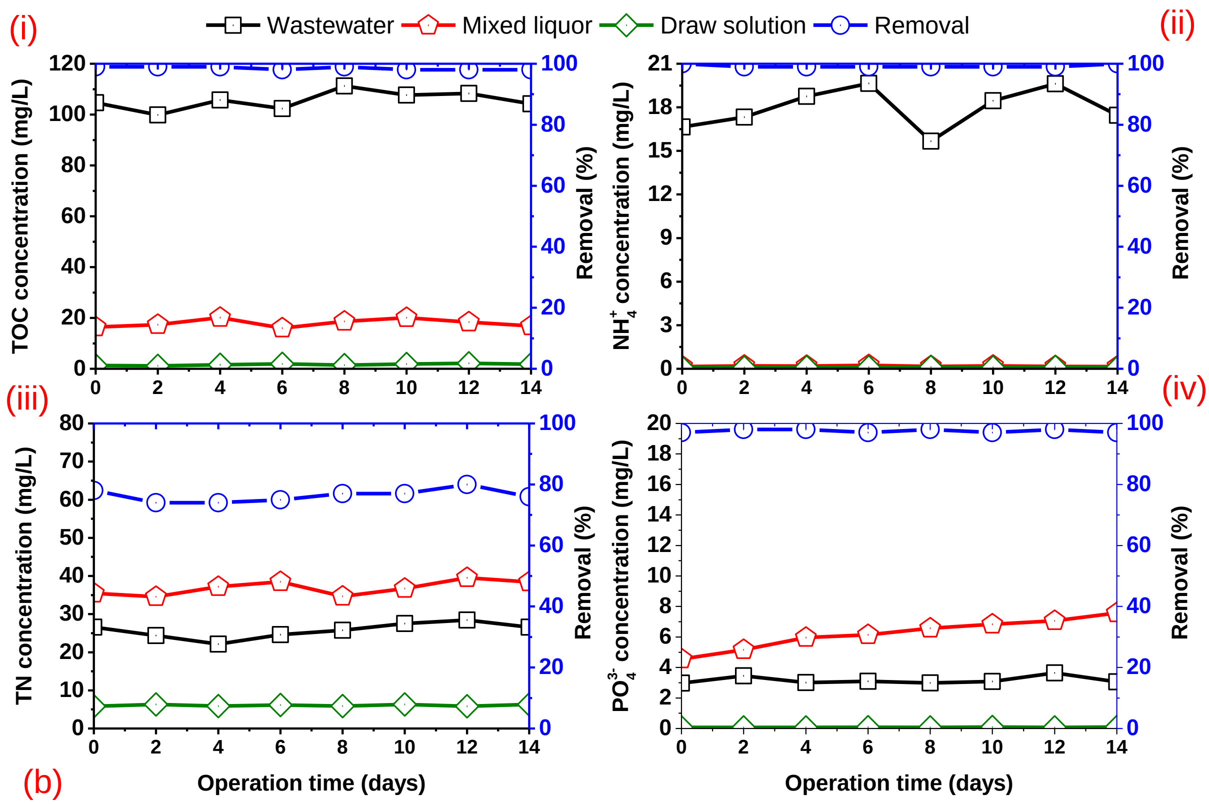


Figure 4. Concentrations and overall pollutant removal efficacies of (a) the submerged and (b) side-stream OMBR systems including (i) TOC; (ii) NH_4^+; (iii) TN removal; and (iv) PO_4^{3-}. Testing conditions: DS = 35 g/L NaCl; FS = activated sludge; AL – FS orientation; Ambient temperature (22 ± 1 ℃).

The removal efficiencies for TOC, ammonium (NH_4^+), total nitrogen (TN), and phosphate (PO_4^{3-}) by the two OMBR systems over the 14-day experiment period are presented in **Fig. 4**. In general, both the submerged and side-stream OMBR systems exhibited high and stable removal of the four selected pollutants over the experiment period, with no significant differences in the removal rates. Synergistic effects between microbial degradation and the high rejection of the FO membrane are the two main contributors to these high pollutant and nutrient removals from the wastewater [11].

As shown in Fig. 4a-(i) and Fig. 4b-(i), the two OMBR systems demonstrated consistent TOC removal, over 98% during the 14 day experiment, similar to results observed in previous OMBR studies [14, 15]. Achilli et. al [1] reported a 99% overall TOC removal in their OMBR work while Qiu and Ting [36] noticed a consistent TOC removal efficiency of up to 98%. Some recent OMBR studies have also presented high overall TOC removal with over 98% [37] and 96% [37].

Consistently high NH_4^+ removal by the two OMBR systems was also observed over 14 days as illustrated in Fig. 4a-(ii) and Fig. 4b-(ii) which agrees well with our previous OMBR studies using external side-stream modules that reported excellent NH_4^+ removal [14, 15]. Some other OMBR research works have also reported high removal efficacies of NH_4^+ [1, 3, 11, 36]. The high overall NH_4^+ removal by the two OMBR systems is due to the synergistic effects between the biological nitrification by ammonium-oxidizing bacteria and the high rejection of the FO membrane [14].

Fig. 4a-(iii) and Fig. 4b-(iii) present the removal rates of TN by the two OMBR systems, which demonstrated consistent removal rates of up to 80% and TN concentrations in the reactors marginally increased over the experiment time. Continuous accumulation of nitrogen compounds in the reactors, which were strongly rejected by the FO membrane, might be attributed to the slight increase of TN concentration in the reactors. Microbial degradation of nitrogen compounds by nitrification and denitrification processes is the main contributor to the TN removal process in the OMBR systems. Nitrification normally is an effective process in the aerobic OMBR system, with sufficient dissolved oxygen, converting ammonium into nitrite and nitrate. However, the denitrification process is not likely to occur in the bioreactor due to the presence of dissolved oxygen and this resulted in the retention and increased accumulation of TN, mostly in the form of nitrates in the bioreactors because of high rejection by FO membrane [38]. Previous OMBR studies using different types of flat-sheet Aquaporin, TFC, and CTA FO membranes also found similar results [3, 19, 39].

Fig. 4a-(iv) and Fig. 4b-(iv) demonstrate that both OMBR systems achieved consistently high removal of phosphate (PO_4^{3-}), of up to 98%. The high rejection of the FO membrane is expected to be the primary contributor to the effectiveness of PO_4^{3-}. Negatively charged orthophosphate ions have a large hydrated radius diameter of 0.49 nm, which were almost retained by the FO membrane [10]. The high PO_4^{3-} removal rates were in agreement with other OMBR studies that have reported removal rates of 99% and 98% [19]. In conclusion, these two OMBR systems using side-stream and submerged membrane modules with an OSHF TFC FO membrane exhibited stable and high removal efficacies of the four investigated pollutants.

3.4 Membrane and fouling cake layer characterization.

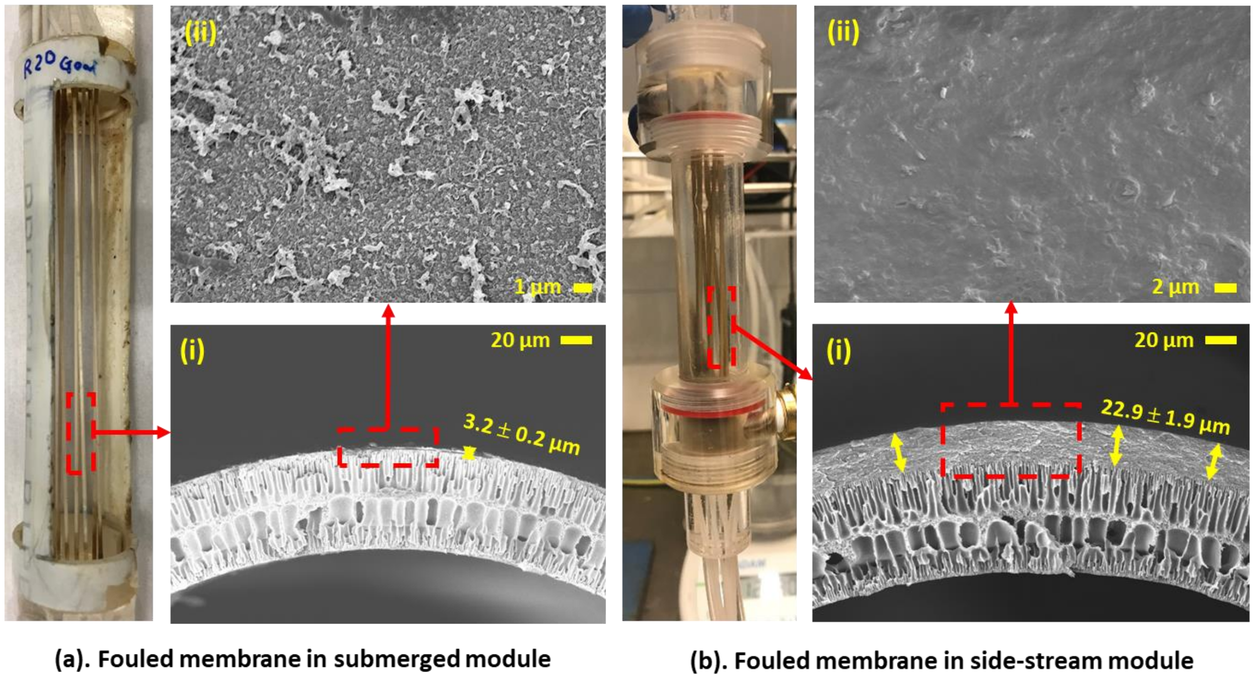


Figure 5. SEM images of fouled OSHF TFC FO membranes (without cleaning) in (a) submerged module and (b) side-stream module; (i) cross-sectional morphology; and (ii) outer surface morphology.

Table 1 The elemental compositions of pristine and fouled membranes in two operation regimes by energy-dispersive X-ray (EDX) analysis.

Weight %	*C*	*O*	*N*	*S*	*Na*	*Cl*	*Mg*	*Al*	*Si*	*P*	*Ca*	*Fe*
Submerged	55.42	17.56	4.75	3.08	6.24	8.15	0.24	0.35	0.19	1.15	1.02	1.85
Side-stream	47.21	15.34	5.94	4.24	7.34	10.46	0.93	0.96	0.73	2.34	1.95	2.56

Fig. 5 shows images of submerged and side-stream membrane modules and SEM images of the outer surface and cross-sectional morphologies of fouled hollow fiber FO membrane at the end of the experiment period without washing. Cross-sectional SEM images in Fig. 5a-(i) and Fig. 5b-(i) demonstrate the significant difference in thickness for the fouling cake layers. The thickness of the cake layer of a fouled membrane in the side-stream module was found to be 22.9 ± 1.9 µm, 7-fold thicker than the one in submerged module, with a thickness of 3.2 ± 0.2 μm. The outer surface morphologies shown Fig. 5a-(ii) and Fig. 5b-(ii) further confirm that the outer surface of the fouled hollow membrane in the side-stream module was completely covered by a dense and thick foulant layer. However, only a small portion of the outer surface of the fouled membrane in the submerged module was covered by foulants. There was a large area with typical ridge-and-valley morphology of the polyamide selective layer that can be visually seen on the outer surface of the fouled membrane in the submerged module that was not observed for the side-stream membrane. This result is in contrast to the one observed by Morrow et. al [9] in their OMBR study in which they reported that the cake layer of a fouled flat-sheet CTA membrane in the submerged module was thicker than the one in the side-stream configuration.

The scattering of foulants on the outer surface of the fouled submerged hollow fibre membrane indicates that foulants could not be deposited onto the membrane surface under continuous aeration in this study. The direct contact of air bubbles onto the membrane surface generated air scouring and hindered the initial interaction between foulants and the membrane surface, inhibiting subsequent foulant-foulant interactions need to form a thicker cake layer. This finding confirms why the flux decline rate in the submerged OMBR system was significantly slower when compared to the side-stream module. Similarly, previous research studies on conventional submerged MBR and OMBR systems reported the effectiveness of air bubbles as a fouling control method [35, 40-42].

The elemental compositions of the fouling cake layers taken from the two membrane modules were analyzed using EDX. According to the analytical results presented in Table 1, both fouling cake layers contained various inorganic elements including Na, Cl, Mg, Al, Si, P, Ca, and Fe, and four main elements (C, O, N, and S) that come from the functional groups of the FO membrane itself. Results in Table 1 also show that the fouled membrane from the side-stream module contained higher percentages of the analyzed elements than the one in the submerged module. This is a reasonable, as the fouling cake layer in the side-stream module was thicker and more compacted with foulants. The presence of Ca, Mg, and Si might be an indicator of inorganic scaling in addition to organic and biofouling, and other elements such as Fe and P might originally come from the synthetic wastewater. A high amount of Na and Cl detected in the fouling cake layer could be related to the reversely diffused draw solutes which accumulated in the fouling cake layer.

3.5 Operating costs comparison between submerged and side-stream module configurations

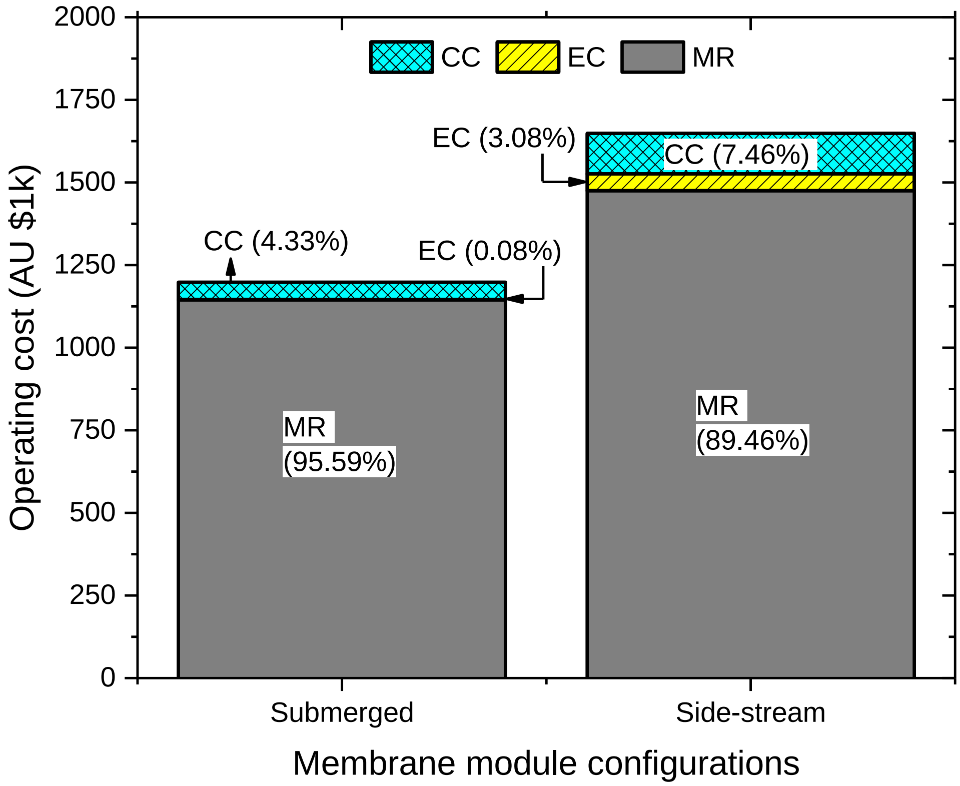


Figure 6. Annual OPEXs and cost contributions of membrane replacement (MR), energy consumption (EC), and cleaning cost (CC) to operating costs (OPEX) of the submerged and side-stream OMBR systems for a plant capacity of 24,000 m^3/day.

Fig. 6 presents the annual operating costs (OPEX) and cost contributions of the three main operating components, including membrane replacement (MR), energy consumption (EC), and cleaning cost (CC) to the overall OPEX of the submerged and side-stream OMBR systems. In general, the submerged OMBR system demonstrated a lower operating cost (AU $1.20 million per annum) than that of the side-stream OMBR system (AU $1.65 million per annum) to produce a similar volume of diluted DS. This result demonstrates that the submerged OMBR system is more economically viable compared to the side-stream system. MR was the major cost contributor, while the EC and CC were relatively minor ones for the two OMBR systems. The cost of MR in the side-stream system was AU $1.48 million, accounting for 89.46% of the OPEX, which is 1.29 times higher the MR cost (AU $1.14 million – 95.59%) for the submerged system. This is mainly due to the lower cost of the submerged membrane module even the submerged system requires double the number of membrane modules compared to the side-stream to produce a similar volume of diluted DS. The higher cost of the side-stream membrane module is due to the additional cost of housing materials and increased complexity in its production.

EC and CC in the submerged OMBR system were much lower than that of the side-stream system. For instance, EC cost contributed only 0.08% to the overall OPEX of the submerged system, which is 50 times lower than that of the side-stream system, contributing 3.08% to the overall OPEX. The main reason for this difference is that the side-stream OMBR system requires much more power for the circulation of mixed liquor into the cross-flow membrane module, and the regular injection of air bubbles into the membrane module, while there was no recirculation of activated sludge carried out in the submerged OMBR system. Similarly, the CC in the immersed system contributed 4.33% to the overall OPEX, which is two-fold lower than CC cost for the side-stream system, which accounted for 7.46% of its overall OPEX. This is due to the higher frequency of physical and chemical cleaning required for the cross-flow modules compared to the submerged ones, even though the number of submerged modules was almost double.

Results of the economic analysis demonstrate that the submerged OMBR system is more viable when compared to the side-stream. Further, the utilization of submerged membrane modules does not require circulation of activated sludge, which will minimize harmful impacts on the growth and biodegradation activities of microorganism. Circulation of activated sludge with high CFV generates strong shear force resulting in detrimental damage and breakage of microbial flocs, lower sludge yield, and reduced removal efficiency of pollutants [20]. Furthermore, more fine colloidal particles are created with the breakages of flocs, which releases extracellular polymeric substances (EPS), increasing the resistance of the fouling cake layer, exacerbating membrane fouling. The submerged OMBR system is therefore more preferable for a long-term operation as it has can sustain its flux for longer periods. However, to make the submerged system more viable, a reduction in OPEX cost is required. Based on the OPEX cost components of the two OMBR systems, the most direct way is to lower the cost of MR. Logically, the two obvious approaches to lower the MR cost is to develop OHF FO membranes with better performance in terms of water flux and fouling control [24]. While membrane price depends upon the market's maturity and mass production, higher water flux can be achieved by optimizing operating conditions and using new generations of membrane modules possessing better properties.

4 Conclusions

This study conducted an evaluation of the comparative performances of two different OMBR module configurations (submerged and side-stream) using an outer-selective hollow fiber thin-film composite forward osmosis membrane in terms of their fouling potential and fouling mitigation efficiencies. In general, the cross-flow side-stream OMBR module demonstrated a higher water flux but experienced more severe fouling compared to the submerged membrane module, thereby requiring more frequent physical cleaning. Both the OMBR systems, however, achieved similarly high removal efficiencies of both pollutants and nutrients (TOC, NH_4^+, TN, and PO_4^{3-}) over the entire experimental period. Normal aeration used in the bioreactor can also be utilized as an effective fouling mitigation method in the submerged OMBR system without the need for additional aeration as a fouling control. The submerged OMBR system demonstrated lower OPEX cost compared to the side-stream system and membrane replacement cost formed major cost component of the both the OMBR systems. Therefore, employing higher performance outer selective hollow fiber FO membrane and low-cost submerged module will reduce the OPEX cost of the OMBR system.

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