Secret underneath: fouling of membrane support layer in anaerobic osmotic membrane bioreactor (AnOMBR)

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Abstract

Anaerobic osmotic membrane bioreactor (AnOMBR) holds promise for simultaneous wastewater purification and biogas production, allowing for an energy and carbon-neutral treatment facility. In a typical AnOMBR, reverse osmosis (RO) is employed for re-concentrating draw solution for continuous operation and cost saving. We compared membrane fouling behaviors between AnOMBR-RO hybrid system and AnOMBR without RO unit. We concluded that the porous support layer was susceptible to both inorganic scaling and biofouling, in the closed-loop AnOMBR-RO system. We also explored two cleaning approaches to mitigate inorganic scaling and biofouling. Specifically, ethylenediaminetetraacetic acid (EDTA) was introduced into draw solution for minimizing inorganic scaling, but biofouling was deteriorated as EDTA provided extra nutrients for bacterial proliferation and biofouling. On the other hand, chemical cleaning of membrane support layer was performed using NaClO solution for biofouling control, but such cleaning efficacy attenuated after several cleaning cycles, because inorganic minerals accumulated and grew within membrane porous layer which could not be flushed by NaClO cleaning. Our finding highlighted the complexity and courter intuitive perspective to membrane fouling and cleaning in AnOMBR-RO hybrid system for inorganic scaling and biofouling management.

Keywords: membrane fouling, biofouling, support layer, forward osmosis, osmotic membrane bioreactor, wastewater treatment

1. Introduction

With urbanization and industrial advancement of modern society, water shortage has become a serious problem. In response to this water crisis and fulfilling United Nation Sustainable Development Goal of ensuring availability and sustainable management of water and sanitation for all, novel and high efficiency wastewater treatment technologies are required to purify and recycle water sources. Forward osmosis (FO) is one such promising technology that can bridge the gap in wastewater treatment and reuse. Previous studies demonstrated that fouling trend of FO membrane was less severe compared to the pressure-driven membrane processes [1-4].

Coupling FO process with biological treatment give birth to the development of anaerobic membrane bioreactor (AnOMBR) that possesses superiority in water quality and bioresource reclamation, as organic matters can be recovered in the form of biogas while nutrients such as nitrogen, phosphorus in the form of struvite precipitation [5, 6]. However, severe membrane fouling hinders the sustainable operation and deployment of AnOMBR as water flux decreased sharply; and at the same time, the effluent quality was compromised. In an early effort, Chen et al. [7] replaced traditional microfiltration (MF) membrane and ultrafiltration (UF) membrane in AnOMBR by FO membrane, with the aim of obtaining high quality water and reducing energy cost. Satisfactory phosphate (100%) and organic (>96.7%) removal were achieved by FO membrane for municipal wastewater treatment. However, periodical supernatant replacement was required for the feed side as the conductivity increased to 22 mS/cm, driven by reverse solute diffusion from the draw side. In addition, severe membrane fouling hindered the long-term, stable operation of the AnOMBR. For instance, Wang et al. [8] identified

membrane fouling in the active layer of FO membrane after 101-day AnOMBR operation. They found that biofoulants, especially polysaccharides, proteins and microorganisms, were firmly attached on membrane surface, thereby reducing water flux and shortening operation time.

Counter-intuitively, membrane fouling in AnOMBR can also occur within the membrane support layer, particularly in a closed-loop system. For instance, significant fouling within membrane support layer was reported by Kim et al. [9], and membrane physical flushing proved futile as foulants occurred and trapped inside the porous cellulose triacetate support layer. Such detrimental membrane fouling was driven by the contaminant accumulation within draw solution in a closed-loop system where draw solution was re-concentrated through a wide range of separation processes, such as, nano-filtration (NF) [10], membrane distillation (MD) [11] and reverse osmosis (RO) [12]. For instance, when treating digested sludge feed by an FO-RO membrane system, Xie et al. [13] found that protein-like substance was the major constituent that accumulated in the draw solution, which could pass through FO membrane while be rejected by RO membrane. In addition, Choi et al. [14] also reported that membrane fouling shifted from RO membrane to FO membrane in treatment of secondary wastewater effluent in the FO-RO hybrid system, confirming the transportation of low molecule weight substance to the DS side. In their long-term, pilot-scale FO-RO osmotic dilution process to treat wastewater from coal-fired power station [15], they observed contaminants accumulation in the draw solution which was detrimental to downstream RO membrane. As a result, in a closed-loop AnOMBR, there exists an unavoidable issue associated with contaminant buildup in the draw solution; and consequently, membrane fouling in the support layer of the AnOMBR system was hypothesized due to constant accumulation of inorganic and organic compounds in draw

solution.

In this study, we investigated membrane fouling in a closed-loop AnOMBR system, coupling with the RO unit to re-concentrate draw solution. Filtration performance and fouling behavior of AnOMBR-RO hybrid system were examined and analyzed. Varying approaches for membrane cleaning were performed and compared to alleviate membrane fouling within the membrane support layer (i.e., the side facing draw solution). Findings reported here shed insights into membrane fouling mechanisms in the AnOMBR-RO hybrid system, and offered effective membrane fouling mitigation strategies for sustainable AnOMBR operation.

2. Materials and methods

2.1 Bioreactor set-up and operating conditions

Details of the AnOMBR were reported in our previous literatures [8, 16]. Briefly, the effective volume of the AnOMBR unit was 5 L; two pieces of cellulose triacetate (CTA) membrane (provided by Hydration Technologies Inc.) with the total area of 0.025 m^2 were placed in the FO module; the FO module as well as an MF membrane with the identical membrane area were immersed in the bioreactor. 0.5 M NaCl was used as draw solution during the operation and its concentration was maintained in the comparatively stable level by the conductivity controller (OKD-650, Shenzhen OK Instrument Technology, China) connected to the concentrated NaCl solution of 5 M. Permeate water flux of the MF membrane was adjusted to achieve the low salinity environment in the feed side and the conductivity of the mixed sludge was controlled around 2-4 mS/cm.

An RO unit (STM-0021-HP, Starmen Scitechnology, Xia'men, China) was used to concentrate draw solution for AnOMBR. The RO unit was operated in batch mode with SW30

RO membrane (Dow, Filmtec) with well documented NaCl rejection above 99.4%: when the draw solution volume reached 1.3 times of its initial volume, RO was operated at 4 MPa until the draw solution had been concentrated to its initial volume. The start and stop of RO unit were automatically controlled through water level sensor. The draw solution temperature was maintained as 25 ± 1℃.

Two AnOMBR systems were used for fair comparison of membrane fouling: AnOMBR was operated with draw solution being discarded, which was denoted as R1; the AnOMBR hybrid with RO was denoted as R2. Specifically, synthetic municipal wastewater was used as influent and its composition can be found in previous literatures [8, 15]. The initial mixed liquor suspended solids (MLSS) concentration was 2.8 g/L and the MLVSS/MLSS value was 0.6 at the beginning. This selection of initial MLSS was reflected as a low strength feed wastewater, and was consistent with ample previous literatures [7, 8, 16-19].

During the operation, the sludge retention time (SRT) was set as 100 days while the hydraulic retention time was decided by the FO and MF permeate fluxes.

Membrane fouling mitigation was carried out via two approaches: introducing ethylenediaminetetraacetic acid (EDTA) into draw solution or performing chemical cleaning by NaClO solution. Specifically, 20 mg/L EDTA was added into 0.5 M NaCl draw solution in AnOMBR-RO hybrid system, which was denoted as R3. Periodical chemical cleaning was performed every five days. During NaClO cleaning, 2 L NaClO solution (50 mg/L) was circulated in the module through a peristaltic pump for 15 min followed by DI water rising for 15 min. The AnOMBR-RO hybrid system with periodical chemical cleaning was denoted as R4.

2.2 Analytical methods

Influent, effluent and sludge supernatant samples were analyzed for NH_4^+-N, total nitrogen (TN) and total phosphorus (TP) concentrations using standard method. TOC analyzer (TOC-

V_{CPH}, Shimadzu, Japan) was used to measure the TOC concentrations by combustion oxidation method. Inductively coupled plasma optical emission spectrometer (ICP-OES, Optima 8300, PerkinElmer, USA) was used to analyze ion profile of draw solution samples at the conclusion of each batch operation. Concentration in the draw solution was corrected by taking concentration factor into account due to the batch mode operation in RO unit. The corrected concentration was then used for calculation of the removal efficiency of the whole AnOMBR system. Morphology observation of the fouled membranes was carried out by a HITACHI S3400 scanning electron microscope (SEM).

In order to analyze the membrane biofouling, membrane samples were stained by Concanavalin A (ConA), Calcofluor white (CW), Fluorescein isothiocyanate (FITC) and SYTO^{TM} 63 in order to analyze the spatial distribution of α-D-glucopyranose and β-D-glucopyranose polysaccharides, proteins and microorganisms, with a confocal laser scanning microscope (CLSM, TCS SP8, Leica, Germany). Detailed information about sample preparation and data calculation can be found in our previous publications [20, 21]. Briefly, the fouled FO membranes were taken out from the bioreactor at the conclusion of the operation; and then three pieces with size of about 1 × 1 cm were randomly cut from each fouled FO membrane. The fouled membrane samples were first stained with SYTO 63 (20 μM), and then the FITC solution (10 g L^{-1}) was dripped onto the samples after 1 M sodium bicarbonate ($NaHCO_3$) buffer was applied for keeping the amine group in a non-protonated form. Subsequently, the ConA (0.25 g L^{-1}) and CW (0.3 g L^{-1}) solutions were added to the samples, respectively. After each stage of the labelling process, the samples were incubated for 30 min at room temperature in the dark, and

157 then were washed twice with phosphate buffered saline (PBS) solution to remove the
158 extra probes. The three-dimensional reconstructions were obtained with ZEISS confocal
159 software, and the images were analyzed by PHLIP and Image J to calculate the quantitative
160 parameters, including biovolume, and average biofouling thickness.

161 For cleaning and analyzing inorganic scaling of the fouled membrane, 0.5% hydrochloric
162 (HCl) as well as 0.5% hydrogen peroxide (H_2O_2) was used to rinse the membrane support layers,
163 respectively, to remove inorganic and organic contaminants [16]. After that, membrane samples
164 were cut into 1 cm × 1 cm, and were dried in the oven at 60 ℃ for 12 h. Dried samples were
165 heated to 600℃ in the furnace for two hours, the weight loss before and after calcination
166 represented organic compounds, and the main components of the residue were inorganic scale.

167 **3. Results and discussion**

168 **3.1 Bioreactor performance: contaminant removal and water production**

169 Stable operating performance was achieved in both R1 and R2 reactors (Figure 1). Owing
170 to the addition of MF membrane, salinity of the two bioreactors was well controlled between 2
171 and 5 mS/cm during operations, which was one order of magnitude lower than a typical
172 AnOMBR [7]. It further corroborated that OMBR coupled with MF membrane could obtain
173 long-term operation under lower salinity environment. Effluent of higher quality was produced
174 from R2 due to the dual membrane barrier.

175 Both bioreactors achieved high product water quality. Nearly no phosphorus was detected
176 in the RO permeate of R2 (Figure S3). In R2, noticeable ammonia nitrogen (up to 59 mg/L)
177 accumulated in the draw solution and was due to Donnan effect [22, 23]. This phenomena
178 was mainly attributed to bidirectional diffusion of ammonium of feed solution and

sodium cations of draw solution in forward osmosis process; and on the other hand, ammonia nitrogen was relatively well rejected by RO unit, thereby resulting in the final concentration of ammonia nitrogen in the product water from R2 lower than 8.8 mg/L (Figure S4). For organic removal in R2, the TOC concentration in RO permeate was 4% lower than that in FO permeate. It was mainly driven by the similar solute-rejecting capacity for FO and RO membrane used in the experiment whose molecular weight cutoffs were 250 Da and 180 Da, respectively [14]. Amino acids as well as proteins with low molecule weight could easily pass through both FO and RO membranes.

In addition, as AnOMBR featured in biogas production during anaerobic process, the methane yield ranged from 0.25 to 0.30 L CH_4/g COD in both R1 and R2 reactors, which was consistent with previous data on AnOMBR [7, 8, 16].

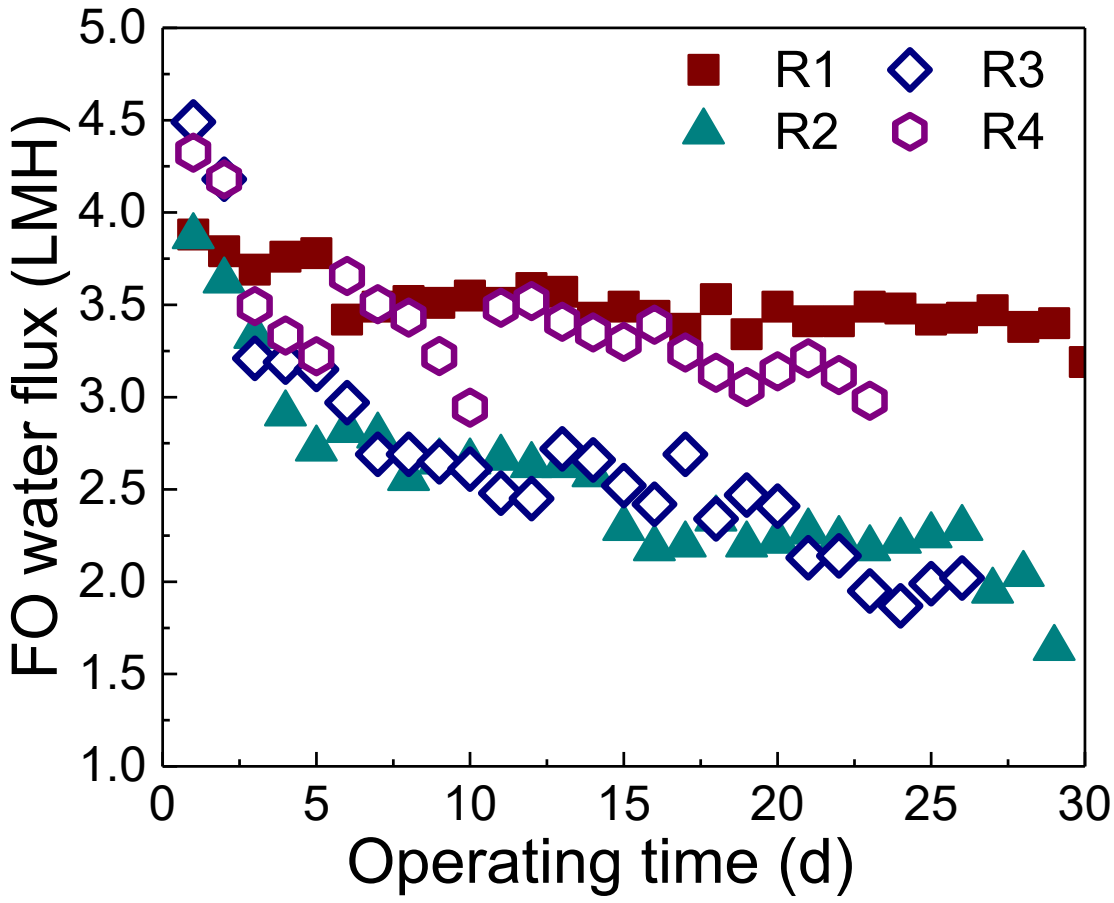


Figure 1: FO permeate flux for different bioreactors as a function of operating time. R1: AnOMBR without RO unit; R2: AnOMBR hybrid system with RO unit; R3: AnOMBR hybrid system with RO unit, and EDTA was added to the draw solution; R4: AnOMBR hybrid with RO unit, and periodically NaClO chemical cleaning was performed every 5 days.

Water fluxes in both R1 and R2 were stable during the operation, although CTA FO membranes used in AnOMBR possessed relatively low water flux. Indeed, water flux of FO membrane in R1 was 3.5 LMH in average during the 30-day operation (solid square in Figure 1) and was similar to Wang et al. [8] whose water flux ranged from 2.5 to 3.5 LMH during the stable stage. However, water flux for CTA FO membrane in R2 hybrid system decreased sharply and the stable water flux was around 2 LMH (solid triangle in Figure 1), which was 42% less in water production. The key difference between R1 and R2 was the addition of downstream RO unit for a closed-loop system. It was hypothesized that such significant discrepancy in water flux profile was mainly driven by membrane fouling. Particularly, bio-fouling and inorganic scaling were substantially different for these two bioreactors as membrane fouling occurred not only in the membrane active layer but also in the membrane support layer in the closed-loop bioreactor as R2. In another word, the biofouling occurred on the membrane active layer was not the key reason for water flux deterioration, but the biofouling and scaling developed within the membrane porous support layer that dictated the system performance.

3.2 Membrane fouling in AnOMBR

3.2.1 Membrane fouling in membrane active layer revealed negligible difference

Key characteristics of biofouling layer in two AnOMBRs (R1 and R2) were imaged and compared (Figure S6). In generally, the biofouling on membrane active layers were similar for

R1 and R2 in terms of biofouling layer thickness and biovolume. There were negligible difference in composition of biofouling layer for either AnOMBR (R1 and R2). Specifically, the total thickness of biofouling layer was 61.40 ± 3.84 μm and 61.40 ± 1.54 μm for R1 and R2, respectively. In addition, average thickness of β-D-glucopyranose polysaccharides and proteins was 32.01 ± 0.05 μm and 37.89 ± 3.97 μm for R1; and was 26.44 ± 3.65 μm and 31.16 ± 3.69 μm for R2, respectively. Moreover, similar inorganic composition of the fouling layer on the membrane active layer was observed. Weight percentage of inorganic mineral accounted for 5.6% and 6.1% for R1 and R2, respectively. As a result, it was concluded that membrane fouling in membrane active layer was similar in both bioreactors, and cannot sufficiently explain the discrepancy in system performance.

3.2.2 Membrane fouling in membrane support layer was critical and substantially different

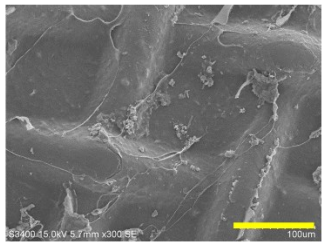
Biofouling and inorganic scaling within membrane support layer were systematically analyzed and compared in different AnOMBRs (R1 and R2). Evidence for bacteria proliferation and biofilm development within membrane support layer in both AnOMBR was highlighted in element analysis where strong peak of sulfur (Figure 2E) existed due to bacterial metabolism. Indeed, CLSM images of the membrane support layers illustrated the occurrence of biofouling (Figures 2 C and F). More importantly, biofouling in R2 AnOMBR was more severe than that in R1 one. The thickness of the fouling layers of R1 and R2 were similar of 61.68 ± 1.92 μm and 57.33 ± 0.38 μm, respectively. In addition, biovolume of biofilm was quantified and summarized in Table 1. For R2 AnOMBR, volumes of β-D-glucopyranose, proteins and total cells were 111.04%, 53.96% and 16.61% higher than that for R1, respectively, indicating active

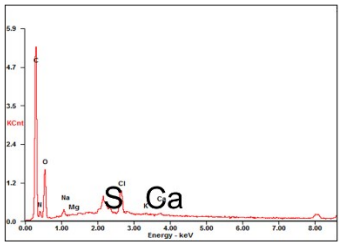
biofilm growth and severe biofouling.

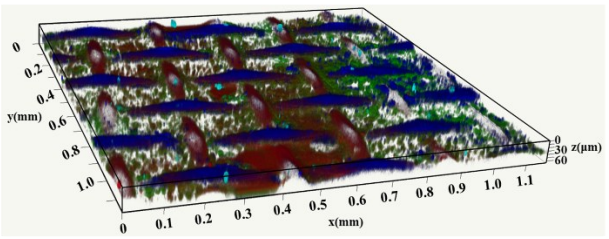
Such unfavorable biofouling in closed-loop R2 AnOMBR was mainly driven by the accumulation of nutrients and low molecule-weight organic substance in the draw solution [10, 11, 14]. Bacteria would utilize the accumulated nutrients in the draw solution, thereby excreting metabolites such as, extracellular polymeric substances and varying proteins would facilitate biofilm growth within membrane support layer [24], resulting in severe biofouling.

From the perspective of inorganic scaling, ion profile of draw solution for both AnOMBR raised concern for inorganic scaling, particularly for mineral precipitation of $CaCO_3$ and $MgCO_3$ within the membrane porous support layer. Indeed, concentrations of calcium and magnesium ions in the draw solution for AnOMBR-RO hybrid system (R2) were an order of magnitude higher than those for AnOMBR (R1) (Table S1). Given that the solubility product constant (K_{sp}) of $CaCO_3$ and $MgCO_3$ were $10^{-8.48}$ and $10^{-5.17}$, respectively [25, 26], it is highly likely that $CaCO_3$ precipitation could occur and could subsequently deposit within the porous support layer, thereby deteriorating internal concentration polarization and decreasing FO permeate water flux [27]. As a result, it was necessary to inhibit the formation of calcium carbonate via either pH adjustment or adding scale inhibitors.

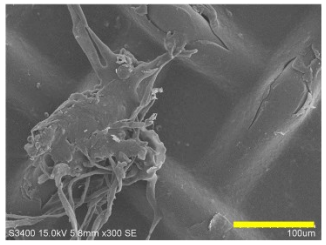
AnOMBR, **R1**

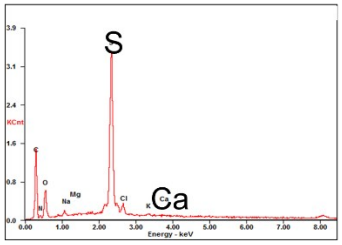


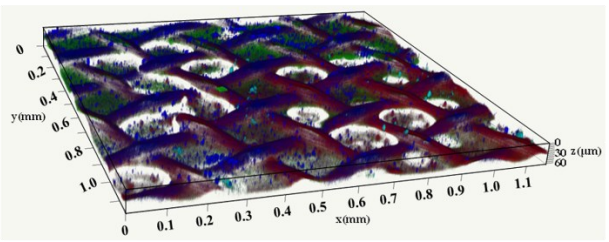




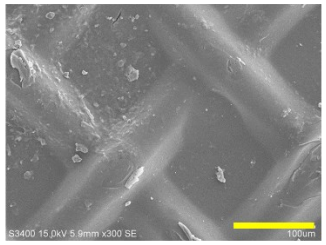
AnOMBR with RO unit, **R2**

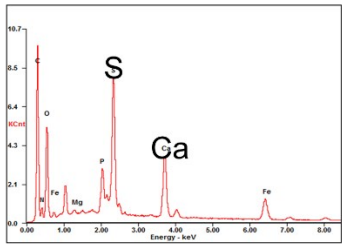


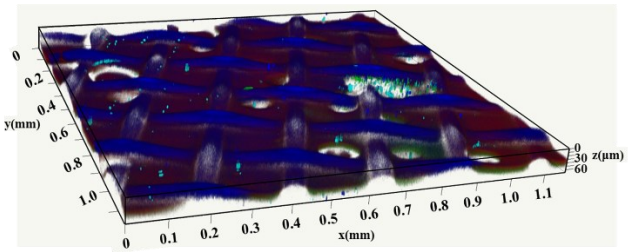




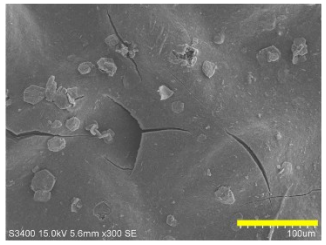
AnOMBR with RO unit; EDTA draw, **R3**

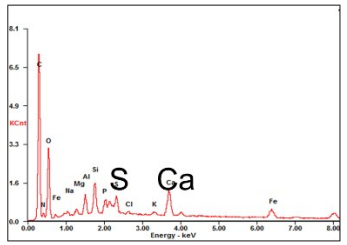






AnOMBR with RO unit; NaClO cleaning, **R4**





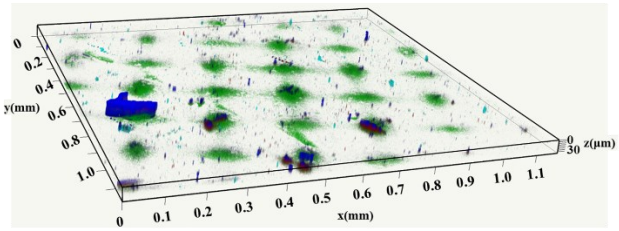


Figure 2: Characterisation of membrane fouling of membrane porous support layer in four different AnOMBRs (A-C for AnOMBR, R1; D-F for AnOMBR hybrid with the RO unit, R2; G-I for AnOMBR hybrid with the RO unit, and EDTA was added to the draw solution, R3; and J-L for AnOMBR hybrid with the RO unit, and periodically NaClO cleaning was performed every 5 days, R4). SEM and EDX element analysis of fouled porous membrane support layer for (A and B) R1, (D and E) R2, (G and H) R3 and (J and K) R4. Scale bar in the scanning electron microscope is 100 μm. CLSM 3D images of fouled porous membrane support layer for (C) R1, (F) R2, (I) R3 and (L) R4.

3.3 Fouling mitigation in membrane support layer of AnOMBR

3.3.1 Introducing EDTA into draw solution

Introducing chelate reagent into draw solution can be an effective approach to mitigate inorganic scaling, particularly $CaCO_3$. In R3, EDTA was introduced into the draw solution to minimize $CaCO_3$ scaling within membrane porous support layer. EDTA, with a relatively large molecular weight, could be well rejected by the FO and RO membrane, which corroborated by similar TOC concentration in the final product water from R2 (6.75 mg/L) and R3 (6.61 mg/L) (Figure S2).

Unexpectedly, FO water flux profiles in R2 and R3 were similar after introducing EDTA into draw solution (Figure 1). Such counter-intuitive phenomena raised concern why EDTA had an adverse effect on flux performance while it could work effectively as scale inhibitor. It was hypothesized that addition of EDTA into draw solution could introduce carbon and nitrogen source that were favorable for microorganism activities. Indeed, EDTA may be used as a carbon source and a nitrogen source in bacterial growth [28]. In addition, EDTA is also an effective ligand to most cations, thereby being helpful for the uptake of essential elements to facilitate biofilm development. Strong peaks of nitrogen and sulfur were detected on the membrane support layer from AnOMBR R3 (Figure 2H), which indicated active biofilm development.

280 More alarming, the CLSM images of membrane support layer from AnOMBR R3 demonstrated
281 a thick and compact biofouling layer (Figure 2I). In addition, detailed analysis of biovolume
282 composition of biofouling in AnOMBR R3 (Table 1) demonstrated that biovolume of β-D-
283 glucopyranose polysaccharides, proteins and total cells in R3 were 37%, 35 % and 43% higher
284 than those in R2.

285 **Table 1**: Comparison of biofilm characteristics in the membrane support layers in different AnOMBRs [a]

286

AnOMBR	Thickness (μm)	α-D-glucopyranose ($\mu m^3/\mu m^2$)	β-D-glucopyranose ($\mu m^3/\mu m^2$)	Proteins ($\mu m^3/\mu m^2$)	Total cells ($\mu m^3/\mu m^2$)
R1	61.68 ± 1.92	4.06 ± 0.23	4.44 ± 0.64	5.30 ± 1.14	6.44 ± 0.04
R2	57.33 ± 0.38	4.04 ± 0.01	9.37 ± 1.41	8.16 ± 0.58	7.51 ± 0.47
R3	63.15 ± 0.64	4.65 ± 2.13	12.81 ± 0.91	11.04 ± 1.16	10.71 ± 3.90
R4	32.71 ± 0.26	4.42 ± 0.29	6.43 ± 0.35	9.90 ± 1.17	5.89 ± 1.36

287 [a] Values are presented as average values ± standard deviation (number of measurements: n = 3 from two random samples).

288 3.3.2 Periodical, chemical cleaning of membrane support layer

289 Another perspective to control membrane fouling within support layer in AnOMBR was
290 periodical chemical cleaning using NaClO. NaClO solution can oxidize microorganisms as well
291 as part of biofoulants, which is extensively used in MBR cleaning [29]. Indeed, 1% NaClO
292 cleaning was used by Linares et al. [30] for CTA FO membrane in a sequential batch reactor-
293 FO system, and achieved satisfying water flux recovery.

294 Periodical, NaClO chemical cleaning was performed every five days in AnOMBR R4.
295 Such NaClO chemical cleaning was effective in membrane biofouling control and water flux
296 recovery. Biofilm establishment and growth were strongly inhibited with NaClO cleaning. Only
297 patchy biofilm could be visualized on the membrane support layer using CLSM (Figure 2I).
298 Coincidentally, biovolume of β-D-glucopyranose polysaccharides and total cells were
299 substantially reduced in R4 AnOMBR, being only 68% and 78% of those in AnOMBR R2
300 (Table 1).

301 Such chemical cleaning not only effective mitigate membrane biofouling, but also
302 enhanced FO water flux recovery. R4 AnOMBR demonstrated higher stable water flux in
303 comparison to both R2 and R3 AnOMBRs. Distinctively, FO water flux was promptly restored
304 after NaClO chemical cleaning at day 10 (Figure 1). However, efficacy of NaClO chemical
305 cleaning attenuated at days 16 and 21, achieving only 4.2% and 2.2% water flux recovery,
306 respectively. This diminishing cleaning performance was due to the accumulation of inorganic
307 scalants that were trapped within membrane support layer and could not be washed by NaClO
308 chemical cleaning. Indeed, SEM imaging and element analysis confirmed that inorganic
309 crystals, particularly calcium-based scalants, filled in the membrane porous support layer

310 (Figure 2K).

311

312 **4. Conclusion**

313 Results reported here showed that closed-loop AnOMBR-RO system experienced more
314 severe fouling than single AnOMBR. This discrepancy in AnOMBR performance cannot be
315 explained by similar membrane biofouling on the membrane active layer. Severe fouling
316 occurred within the membrane support layer in the AnOMBR-RO hybrid system, featuring
317 inorganic fouling and biofouling. A set of two approaches – introducing EDTA into draw
318 solution and NaClO chemical cleaning – were carried out to mitigate the membrane fouling
319 within membrane support layer in the AnOMBR. Results showed that EDTA could work as
320 inhibitor to cope with inorganic scalant precipitation; but it could provide nutrients for microbes,
321 which would deteriorate biofouling conversely. On the other hand, periodical NaClO chemical
322 cleaning could effectively control biofouling, but while such efficacy attenuated after several
323 cleaning cycles due to calcium-based inorganic crystals accumulated within the porous support
324 layer.

325 Implications gleaned from this study shed light on membrane fouling mechanisms and
326 cleaning approaches for AnOMBR application. Such counter-intuitive findings shifted focus to
327 membrane fouling within porous membrane support layer in the closed-loop bioreactors. In
328 addition, there was a trade-off in controlling biofouling and inorganic scaling in the AnOMBR
329 operation, which demands a holistic approach to mitigate membrane fouling for sustainable
330 AnOMBR operation.

331

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6. References

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