



Gravity-driven microfiltration pretreatment for reverse osmosis (RO) seawater desalination: Microbial community characterization and RO performance

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ABSTRACT

A pilot gravity-driven microfiltration (GDM) reactor was operated on-site for over 250 days to pretreat seawater for reverse osmosis (RO) desalination. The microbial community analysis indicated that the dominant species in the pilot GDM system ($\sim 18.6 \text{ L/m}^2 \text{ h}$) were completely different from those in the other tested GDM systems ($\sim 2.7\text{--}17.2 \text{ L/m}^2 \text{ h}$), operating on the same feed. This was possibly due to the differences in available space for eukaryotic movement, hydraulic retention time (i.e., different organic loadings) or operation time (250 days vs. 25–45 days). *Stichotrichia*, *Copepoda*, and *Pterygota* were predominant eukaryotes at genus level in the pilot GDM. Furthermore, the GDM pretreatment led to a significantly lower RO fouling potential in comparison to the ultrafiltration (UF) system. This was attributed to the fact that GDM filtration produced a permeate with less amount of assimilable organic carbon (AOC) and biopolymers. Accordingly, lower amount of organic foulants (biopolymers and low molecular weight neutrals) and less biofilm formation on the GDM-RO membrane were observed. Although α -proteobacteria were dominant in both RO fouling layers, their bacterial community compositions at genus level were significantly different. *Thalassobius* had higher abundance in the GDM-RO fouling layers, while *Erythrobacter* and *Hyphomonas* were more predominant in the UF-RO fouling layers.

1. Introduction

A dual membrane process for seawater desalination, i.e., a low-pressure microfiltration (MF) or ultrafiltration (UF) membrane followed by a reverse osmosis (RO) process, has been developed at full scale since more than 20 years [1]. Compared to conventional seawater pretreatment (such as coagulation-flocculation and media filtration), the membrane-based pretreatment is able to tolerate unfavourable variations in feed seawater, has high removal efficiencies of colloids and suspended particles, and has lower chemical consumption and sludge production. Consequently, the RO membrane system can be operated at a higher permeate flux and lower frequency of chemical cleaning, thus resulting in a decrease of the overall cost of seawater desalination [2–6].

However, a major disadvantage of the membrane-based pretreatment technology is the fouling of the pretreatment membrane itself,

which causes productivity decline and higher operational costs. In addition, due to the poor rejection of dissolved organic substances by MF/UF, biofouling of RO membrane may not be effectively alleviated [6]. To improve the pretreatment performance, integration of low-pressure membrane processes with other processes has received increasing attention. Coagulation, ion-exchange, and adsorption technologies have been successfully combined with membrane-based pretreatment to mitigate fouling and improve the permeate quality [2,3]. Furthermore, a hybrid process that integrates biological treatment and coagulation/adsorption with a low-pressure membrane (i.e., membrane bioreactor) has also been proposed, in which the dissolved organic substances could be biologically degraded and physically coagulated/adsorbed due to the longer seawater retention time in the bioreactor [7–9].

Alternatively, gravity-driven microfiltration (GDM) has been proven to be effective in pretreating seawater as a chemical free and low

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energy option [10–12], in which the rejected organic substances are biodegraded by the biofilm on the membrane. In our previous study [12], a pilot submerged GDM reactor was successfully operated for > 250 days without any physical and chemical cleaning (the stabilized flux of $\sim 18.6 \text{ L/m}^2 \text{ h}$; the operation of the pilot system is ongoing and almost 450 days at the time of writing this paper). In particular, the biofilm in the pilot GDM reactor facilitated the reduction in assimilable organic carbon (AOC) and biopolymers. It has been reported that the biopolymers would lead to a conditioning layer that initiate the biofilm development as well as organic fouling [13,14], while AOC level (fraction of “labile” or “bio-available” DOC) in RO feed water was found to be positively correlated with the biofouling of RO systems [15–17].

On the other hand, the moving, grazing and sloughing behaviours of eukaryotic organisms within the fouling layer played a dominant role in controlling the morphology of fouling layer, which in turn had great impact on the stabilized flux. Previous studies [18–20] on surface water treatment by GDM systems showed that protists (such as flagellates, ciliates, amoebae, and heliozoans), and metazoa (such as rotifer, nematodes, and oligochaetes) were the major contributors to enhance the formation of heterogeneous fouling layer on the membranes. We proposed that the advantages of the longer residence time of organic substances and the availability of sufficient space for the growth, predation, and movement of the eukaryotes in the pilot GDM reactors were responsible for the higher flux and better degradation of organics in the submerged GDM systems [12].

To further illustrate the microbial behaviors in the GDM system and to evaluate the feasibility of GDM pretreatment for RO seawater desalination, in this study, we aim to (1) identify the dominant prokaryotes and eukaryotes in the pilot GDM reactor, lab GDM reactor, and filtration cell system; (2) compare the performances of RO membranes fed with the pilot GDM permeate and full-scale UF permeate; (3) characterize the organic and microbial community compositions of the fouling layer extracted from the RO membranes. This study allows us to understand better the transportation pathway of organic substances in the GDM-RO system and to provide meaningful insights for further scale-up of the system.

2. Materials and methods

2.1. Seawater feed

Seawater was collected from the R&D site next to a full scale desalination plant in Singapore. As seawater was chlorinated at the intake before it was delivered to the collection tank, de-chlorination was performed before use. A certain amount of sodium bisulphite (Acros Organics, USA) was added into the feed tank to ensure the total chlorine concentration was zero (measured by a colorimeter, Thermo Fisher scientific, USA).

2.2. GDM setup

A pilot-scale GDM reactor (effective volume of 720 L, operation for 250 days, hydraulic retention time (HRT) of 21.6 h) was set up at the R&D site next to a full scale desalination plant in Singapore. A flat sheet microfiltration (MF) membrane (PVDF, $0.08 \mu\text{m}$) module was submerged into the reactor and the module was located 40 cm below the water level of the overflow line (i.e., a hydrostatic pressure of 0.04 bar). The room temperature was at $26 \pm 1^\circ\text{C}$. A lab-scale GDM reactor (effective volume of 8.4 L, operation for 45 days, HRT of 13 h) and a GDM filtration cell system (feed side volume of 0.0046 L, operation for 45 days, HRT of 0.74 h) were operated at the same hydrostatic pressure at 0.04 bar using the same flat sheet membrane (PVDF, $0.08 \mu\text{m}$). The room temperature was $21 \pm 1^\circ\text{C}$. The details of each setup were described previously [12] and are summarized in Table S1.

The permeate flux ($\text{L/m}^2 \text{ h}$) was obtained by dividing the volume of permeate collected during a given period of filtration by the membrane area. The permeate flux was normalized to a temperature of 27°C (yearly mean atmosphere temperature in Singapore) as described previously [12].

2.3. RO setup

Two parallel stainless steel RO cells with commercial RO membrane (DOW FilmTec, model BW-30) were set up at the pilot plant. The pilot GDM permeate and the UF (PES, 120 kDa) permeate from the co-located full scale desalination plant were collected as the RO feed respectively. In each RO unit, a stirrer (IKA, Germany) and chiller were installed inside of the feed tank (10 L) in order to maintain a constant temperature of $26 \pm 1^\circ\text{C}$. A high-pressure pump (HydraCell, USA) was employed to deliver the feed water to the RO cell at a constant crossflow velocity at 0.17 m/s. The two RO systems were operated at the same permeate flux of $20 \text{ L/m}^2 \text{ h}$ and their respective permeate flowrate was regulated by a mass flow controller (Brooks Instrument, USA). The conductivities of feed and permeate were measured by conductivity meters (Thermo Scientific, USA). In this study, the RO concentrate and RO permeate were recirculated to feed tank and the test solution in the feed tanks was replenished daily. A computer equipped with data logging system (LabVIEW, National Instruments, USA) was used to continuously record the pressure, flowrate, and conductivity in both RO systems [21,22]. The transmembrane pressure (TMP) was calculated based on the difference between the feed and permeate pressure. The ratio of TMP/TMP_0 was used to describe the RO membrane fouling development, in which TMP_0 represents the initial TMP.

2.4. Water quality parameters

The dissolved organic carbon (DOC) of water sample was monitored using a TOC analyzer (Shimadzu, Japan) after being filtered through a $0.45 \mu\text{m}$ hydrophilic filter. The turbidity of the seawater was examined using a turbidity meter (Hach, US). The bacterial amount in the permeate was measured by a flow cytometry (BD Biosciences, US).

2.5. Transparent exopolymer particles (TEP) measurement

The TEP in the permeate was examined after the permeate sample was filtered through a polycarbonate filter (Millipore, USA; a pore size of $0.1 \mu\text{m}$). Three mL of 0.02% aqueous solution of alcian blue in 0.06% acetic acid (Aldrich-Sigma, USA) was used to stain the retained TEP on the filter. The excess dye was then removed by distilled water. The filter with stained TEP was put into a beaker with 6 mL of 80% H_2SO_4 solution (Honeywell, Korea) and the solution was collected after 2 h. The absorption of the solution was measured at a wavelength of 787 nm using a spectrometer (Hach, USA) and the concentration of TEP was calculated based on a calibration curve with gum xanthan (Sigma-Aldrich, USA) as a TEP standard [23,24].

2.6. Liquid chromatography-organic carbon detection-organic nitrogen detection (LC-OCD-OND)

To quantify the soluble organic fractions ($< 0.45 \mu\text{m}$, e.g., biopolymers, humics, building blocks, low molecular weight neutrals and acids) in the feed, permeate, and soluble foulants, size-exclusion chromatography integrated with organic carbon detection and organic nitrogen detection was used. The mobile phase (1.1 mL/min) was delivered by an HPLC pump (Knauer, Germany) to an autosampler (MLE, Germany) and the chromatographic column (Toso, Japan). A UV-detector (at a wavelength of 254 nm, Knauer, Germany) was employed to analyze organic carbon. A variable wavelength UV-detector (220 nm, 254 nm, 280 nm and 350 nm, Knauer, Germany) and a Deuterium lamp

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