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Performances of dead-end ultrafiltration of seawater: From the filtration and backwash efficiencies to the membrane fouling mechanisms

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ABSTRACT

The present work investigates the fouling mechanisms of PVDF hollow fiber membrane (0.03 μm) during the dead end ultrafiltration at a fixed permeate flux (outside to inside configuration) of complex synthetic seawater composed by humic acids, alginic acids, inorganic particles and numerous salts at high concentrations. Short term ultrafiltration experiments at $100 \text{ L h}^{-1} \text{ m}^{-2}$ show that the optimal specific filtered volume seems to be equal to 50 L m^{-2} . A residual fouling resistance equal to $2 \cdot 10^{10} \text{ m}^{-1}$ is added after each cycle of filtration during 8 h of ultrafiltration at $100 \text{ L h}^{-1} \text{ m}^{-2}$ and 50 L m^{-2} . Most of the fouling is reversible (80%). Organics are barely (15% of humic acids) retained by the membrane. Backwash efficiency drops during operation which induces less organics into backwash waters. Humic acids could preferentially accumulate on the membrane early in the ultrafiltration and alginic acids after the build-up of a fouling pre-layer. Colloids and particulates could accumulate inside a heterogeneous fouling layer and/or the concentrate compartment of the membrane module before being more largely recovered inside backwash waters.

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1. Introduction

Ultra- and microfiltration membranes are widely used for seawater treatment instead of or after granular filters located at aquaculture or desalination plants [1–6]. Porous membrane units are often more compact than conventional sand filtration ones [7,8] and allow to limit the (bio)-fouling of downstream units such as Reverse Osmosis (RO) membranes, and therefore the volume of chemicals used [9]. Moreover, Ultra- and microfiltration enable a constant and good quality of the produced water [8,10,11]. Pretreatment by ultrafiltration can represent 10–40% of the total energy consumption of RO desalination plants [9]. The total cost of a RO plant can be 2–7% lower in the case of an UF pretreatment comparatively to a sand filter one [9]. Nevertheless, the mitigation of UF membrane fouling remains a key issue for sustainable operation of desalination plants.

Two modes of dead-end membrane ultrafiltration are widely used for the pretreatment of seawater: filtration under pressure from the lumen of fibers to the exterior or in the opposite direction and filtration by suction with submerged membranes [9]. Polyvinylidene fluoride, polysulfone or polyethersulfone mem-

branes are often used [12,13]. Permeate and backwash fluxes range from 20 to $150 \text{ L h}^{-1} \text{ m}^{-2}$ and from 60 to $350 \text{ L h}^{-1} \text{ m}^{-2}$ respectively and are usually encountered for seawater pretreatment by porous membranes [14–16]. Backwash and filtration cycles can last from a few seconds and up to several minutes or hours respectively [17,18]. Therefore, the present work deals with the fouling mechanisms of membranes during seawater ultrafiltration in dead-end mode (from the outside to the inside of the PVDF hollow fiber membrane). The mass balances were carried out by considering the backwash waters and permeates. Thus, it is possible to identify at any time of the filtration, the compounds that participate to the membrane fouling. Hence, the changes of fouling behavior could be monitored.

Characteristics of natural seawater vary according to season, weather, tide, location, etc. Seawater is mostly composed of water (96%). The remaining 4% are dissolved and particulate organics or minerals such as polysaccharides, proteins, humics, phytoplankton, and salts. On average, the salinity ranges from 25 to 45 g L^{-1} . The marine organic carbon is mainly (>92%) present in dissolved form (DOC). The particulate organic carbon represents 2–8% of the total organic carbon (TOC); the living part of the particulate organic carbon (POC) is very small except during algal blooms. On average, the concentrations of particulate organic carbon and nitrogen in coastal waters or surface waters are between 0.01 and 1 mg L^{-1} ;

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much higher concentrations are frequently found in marine phytoplankton blooms. DOC concentration ranges from 0.5 to 20 mg L⁻¹ for coastal seawater can be found [19–21]. Colloidal and soluble organics can be released by phytoplankton and/or result from cell lysis, solubilization of particles, etc. Humic substances constitute a part of this matter (60–80% of DOC) [22], which are themselves divided into fulvic acids (80%) and humic acids (20%). Sugars represent around 10% of DOC meaning a few hundred micrograms per liter of seawater. They come mainly from phytoplankton [23] and include sugars such as glucose, fructose, galactose, mannose and xylose [20,24]. Finally, the lipids are found in trace amounts. As a matter of fact, due to this high variability of natural seawater composition, it is difficult to repeat similar filtration experiments unless several plants operate in parallel. In fact, an infinite change in the process control or seawater composition can induce huge changes on fouling. As a consequence, membrane fouling behavior cannot be attributed to a defined parameter. For the present study, the ultrafiltration experiments of the synthetic seawater are replicated. Based on the average composition of natural seawater, a synthetic and reproducible saline suspension is prepared. Special attention is paid to the reconstitution of seawater's organic matter and more particularly the polysaccharides and humic substances due to their significant fouling potential [25–31]. Moreover, the fouling potential of organic matter changes in the presence of salts due to the change of interactions between the compounds and the membrane [27,29–35]. Thus, divalent ions of seawater could significantly increase the fouling by the organic matter [36]. Therefore, several types of ions are added to the synthetic seawater.

For the present work, the fouling mechanisms of an ultrafiltration membrane are studied for a (i) high salinity mixture with organic and inorganic multi components rather than only one or two components, (ii) hollow fiber dead-end filtration with fluxes as well as duration of filtration and backwash cycles in the range of values to those found for the industrial plants and (iii) long term ultrafiltrations respectively for a lab scale. Critical conditions of synthetic seawater ultrafiltration are determined thanks to short term experiments. These critical conditions are chosen for longer experiments of ultrafiltration (8 h) in order to study the fouling behavior.

2. Material and methods

2.1. Synthetic seawater preparation

Natural seawater contains the same families of components all over the world. However, the contents of the compounds vary according to tides, seasons, temperatures, regions, weather, etc. (Table 1).

Therefore, synthetic seawater, as much as possible reproducible, is prepared in order to carry out several experiments with similar conditions and define an average behavior of the UF (Table 1). Synthetic seawater should take into account both mineral and especially organic compounds (from particulate to dissolved compounds) since the latter pose big problems of fouling on reverse osmosis pre-treatment [22,41]. For each class of compounds, mineral and organic, one compound was selected as a model.

2.1.1. Mineral matters – dissolved fraction

Most of the research on membrane fouling is performed with lightly salted solutions or with only one or two salts [27,29,31,42]. For the present study, Instant ocean™ salts are dissolved in osmosed water in order to obtain a concentration of 35 g_{salts} L⁻¹. These salts are regularly used for aquariums in order to obtain a reproducible ionic composition with several salts.

Table 1

Characteristics of the prepared seawater and natural seawaters sampled at different worldwide locations [17–22,35–40].

	Prepared seawater (average on 4 synthetic seawaters)	Natural seawater
Turbidity (NTU)	3.1 ± 0.5	0.1–20
pH	7–8	7.4–8.5
Suspended solids (mg L ⁻¹)	8.7 ± 1.7	–
Humic acids (mg _C L ⁻¹)	0.8	–
Alginic acids (mg _C L ⁻¹)	0.2	–
SDI ₁₅ (%/min)	>6.6	>6.6
TOC (mg L ⁻¹)	1.8 ± 0.4	0.8–4.0
Conductivity (20 °C) (mS cm ⁻¹)	42 ± 3	49–62
[Salts] (mg L ⁻¹)	34,684 ± 3	
Na ⁺	10,074 ± 13	10,945–15,000
K ⁺	332 ± 1	383–690
Mg ²⁺	1274 ± 56	1334–1660
Ca ²⁺	414 ± 6	405–780
Cl ⁻	20,173 ± 38	19,080–26,500
Br ⁻	73 ± 1	45–67
NO ₃ ⁻	158 ± 1	0.2–25
SO ₄ ²⁻	2185 ± 15	2400–2965

2.1.2. Mineral matters – particulate fraction

Bentonite from Riedel de Haën has been selected as model for particulate mineral material. It is already present in natural water and has a size inferior to 200 µm. Kieselguhr, a silica-based mineral, is also added to the synthetic seawater. This diatom is largely present in natural seawater. Both bentonite and kieselguhr are maintained in an oven at 100 °C before the preparation of synthetic seawater and 5 mg L⁻¹ of each are used for seawater reconstitution.

2.1.3. Organic matters – dissolved/colloidal fraction

Dissolved and colloidal organic matters (DOM) are mainly composed of humic substances and polysaccharides. They are strongly involved in the fouling of reverse osmosis and ultrafiltration membranes of seawater desalination plants [43,44]. Humic and alginic acids are selected as model compounds.

A concentrated solution of humic acid is prepared. One gram of humic acid (HA) (supplied by Acros Organics) is added to 500 mL of osmosed water. Then, 62.4 mL of NaOH solution (2 N) are added before completing up to one liter with osmosed water and stirring for 24 h in the dark. One liter of this concentrated suspension is added to 1 L of Instant ocean™ salted water at 70 g_{salts}/L. Finally, the pH is adjusted to 7–8 by addition of HCl (2 N) and the solution is filtered through GF/F Whatman filters (0.7 µm) in order to retain the complexes (particles) formed between humic acids and salts. The filtrate is recovered and represents the acid humic stock solution which is stored in the dark at 4 °C during less than 24 h before the preparation of synthetic seawater.

An alginic acid stock solution is also prepared. Thirty five grams of Instant ocean™ salts are dissolved in 500 mL of osmosed water. Then, 0.2 g of alginic acid (supplied by Sigma Aldrich) is introduced before completing up to 1 L with osmosed water. Finally, the suspension is filtered with a 0.7 µm GF/F Whatman filter and stored at 4 °C. It represents the alginic acid stock solution.

The organic carbon concentration of humic and alginic acid stock solutions is determined the day before each synthetic seawater preparation in order to determine volumes to be added in order to obtain the suitable concentrations (0.8 mg_C L⁻¹ for humic acids and 0.2 mg_C L⁻¹ for alginic acids). Humic and then alginic acids

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