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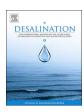
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# Review on strategies for biofouling mitigation in spiral wound membrane systems

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#### ABSTRACT

Because of the uneven distribution of fresh water in time and space, a large number of regions are experiencing water scarcity and stress. Membrane based desalination technologies have the potential to solve the fresh water crisis in coastal areas. However, in many cases membrane performance is restricted by biofouling. The objective of this review is to provide an overview on the state of the art strategies to control biofouling in spiral wound reverse osmosis membrane systems and point to possible future research directions. A critical review on biofouling control strategies such as feed water pre-treatment, membrane surface modification, feed spacer geometry optimization and hydrodynamics in spiral wound membrane systems is presented. In conclusion, biofouling cannot be avoided in the long run, and thus biofouling control strategies should focus on delaying the biofilm formation, reducing its impact on membrane performance and enhancing biofilm removal by advanced cleaning strategies. Therefore, future studies should aim on: (i) biofilm structural characterization; (ii) understanding to what extent biofilm properties affect membrane filtration performance, and (iii) developing methods to engineer biofilm properties such that biofouling would have only a low or delayed impact on the filtration process and accumulated biomass can be easily removed.

#### 1. Introduction

Currently, more than two billion people live in highly water-stressed areas [1,2]. Because of the uneven distribution of fresh water in time and space, the situation is likely to worsen in the future as a large number of regions are expected to experience more extreme climate conditions and rapidly growing demands in water-use sectors: agriculture (crop production, livestock), domestic (municipal), and industry (energy, manufacturing) [2].

Since > 97% of the water in the world is seawater, desalination technologies have the potential to solve the fresh water crisis. Seawater desalination is already used in many countries mainly in water scarce regions such as the Middle East, as well as in countries with adequate freshwater resources.

Desalination technologies can be divided into two major groups: thermal and membrane desalination. While thermal desalination was the main technology in the past, membrane-based desalination technologies gained importance in the last decade, reaching 60% of the

global desalination capacity in 2015 with a continuously increasing trend [3]. This is caused by the improved efficiency and lower energy demand of the membrane-based desalination processes, lowering thus the cost of water production. Reverse osmosis (RO) and nanofiltration (NF) membrane systems currently hold the largest desalination capacity globally [4]. Besides RO and NF there are also alternative emerging membrane-based desalination processes including electrodialysis (ED), membrane distillation (MD) and forward osmosis (FO).

Osmosis is the naturally occurring process where the water from solution passes through a semipermeable membrane to dilute a more concentrated solution. Reverse osmosis (RO) applies hydraulic pressure on the concentrated solution so that the water transport through the membrane is reversed and fresh water can be separated from saline water. RO membranes are able to reject colloidal and dissolved matter from aqueous solutions, resulting in a more concentrated solution called "brine" and fresh water, usually referred as "permeate".

Commercially available membrane modules include spiral-wound, hollow fibre, tubular and modules [91]. Among these, spiral wound

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modules are most commonly applied, due to their high membrane area to volume ratio. The major components of a spiral-wound module are the membrane, the feed and permeate channels, spacers keeping the membrane leaves apart, the permeate tube and the membrane housing [92]. The feed channel spacer may enhance mass transfer near the membrane, but inevitably increases pressure loss along the membrane leaf [93,94]. Membrane sheets with spacers in between are glued together on three sides to form an envelope and multiple envelopes are attached to and rolled up around the permeate tube to create the feed and permeate channels. A pressurized membrane module housing holds the membrane leaves in place. Usually, three or more modules are connected in series in a pressure vessel [5].

The performance of the modules is affected by many factors: (i) spacers geometry, which greatly affects local mixing, mass transfer (concentration polarization) and pressure loss, (ii) fouling propensity and cleaning ability, (iii) plant design and operating conditions, such as feed pre-treatment, feed concentration, feed pressure and permeate recovery.

Four major types of fouling can occur in spiral wound membrane systems: colloidal (suspended particles such as silica), inorganic (salt precipitates such as metal hydroxides and carbonates causing scale formation), organic (natural organic matters such as humic acids), and biological (such as bacteria and fungi). Because the reverse osmosis membranes are nonporous, the formation of a fouling layer on the membrane surface is the dominant fouling mechanism [6]. RO membrane fouling is closely related to the interaction between the membrane surface and the foulant. Previous studies indicated that the physicochemical properties of the RO membrane surface, such as hydrophilicity, roughness, and surface charge, and the feed spacer geometry are major factors influencing membrane fouling [7,8].

Biofouling is considered the major fouling type of the membrane process because microorganisms can multiply over time. Even if 99.9% of microorganisms are removed with pre-treatment of the feed water, there are still enough microbial cells remaining to grow by utilizing biodegradable substances in the membrane installation feed water [9]. Biofouling can be considered as a biotic form of organic fouling while fouling caused by organic matter derived from microbial cellular debris can be considered as an abiotic form of biofouling [10]. Biofouling has been known as a contributing factor to > 45% of all membrane fouling [11] and has been reported as a major problem in nanofiltration (NF) and reverse osmosis (RO) membrane filtration [9,11].

Biofouling of the RO membrane results in a decline in permeate water flux and a decrease of salt rejection. The decline in membrane performance it is due to the increase in the hydraulic resistance and the trans-membrane osmotic pressure of the fouled membrane. The increase in the trans-membrane osmotic pressure it is the result of bacterial cells deposition, which enhance the concentration polarization of salt near the membrane surface [12,13]. The greatest effect of biofilms on membrane systems may be attributed to the physical properties of the extracellular polymeric substance (EPS) matrix produced by the embedded microorganisms by increasing the hydraulic resistance and thus reducing permeate production.

Several fouling control strategies have been developed and tested in full-scale membrane installations. Colloidal, inorganic and organic fouling can generally be controlled by pre-treatment or by dosage of chemicals (e.g. antiscalants). However, biofouling can only be restricted and delayed by pre-treatment, but not eliminated [14]. Direct dosage of oxidizing biocides such as free chlorine is not possible due to damage of the membrane structure causing reduced membrane performance. Several non-oxidizing biocides would be used as nutrient by the microorganisms, thus enhancing biofilm growth [15,16]. Current research is focused on membrane surface modification, non-oxidizing biocides application and modification of the feed channel geometry and operating condition in order to reduce the biofouling in spiral wound membrane systems [17–23]. Quorum sensing is another approach for biofouling control, aiming at biofilm dispersal in response to certain

biochemical compounds; however, no application has been implemented in practice [24]. Despite the efforts on controlling fouling in spiral wound membrane systems, biofouling remains the major problem in membrane filtration processes, causing increased energy demand and unreliable water production. It is therefore crucial to gain more fundamental understanding of the biofilm formation in spiral wound membrane systems, in order to develop strategies to control and keep biofouling at an acceptable level. If biofouling cannot be avoided in the long run, biofouling control strategies should focus on delaying biofilm formation, reducing its impact on membrane performance and removing biofilms by advanced cleaning strategies.

The objective of this review is to provide an overview on the state of the art strategies to control biofouling in spiral wound reverse osmosis membrane systems and point to possible future research directions.

#### 2. Biofouling control strategies

Biofouling is considered the major fouling problem in membrane systems for water treatment. In spite of extensive research to prevent and eliminate biofouling, no successful control strategy has been developed yet. Most common biofouling control strategies are: (i) feed water pre-treatment, (ii) membrane surface modification, (iii) feed spacer design and (iv) chemical/mechanical membrane cleaning.

#### 2.1. Pre-treatment by water filtration and bacterial inactivation

For a constant and reliable operation of reverse osmosis membrane systems good quality feed water is essential. Good feed water quality is defined by the membrane manufacturers as water with a turbidity lower than one Nephelometric Turbidity Unit, NTU, silt density index SDI <3 (or SDI <4), oil and grease  $<1\,\mathrm{mg}\cdot\mathrm{L}^{-1}$  [25]. When the source water does not meet these criteria, the feed water has to be pretreated before entering the reverse osmosis membrane system. Most commonly applied pre-treatment technologies are based on water filtration (e.g., filtration over granular media and low pressure membrane filtration) and disinfection.

Filtration over granular materials can be categorized into single, dual and mixed media filtration [26], meaning that one or more materials can form the filter bed. The most important filtration mechanism here is deposition of the suspended particles to filter media grains, as the raw water passes through the filter bed. The most commonly used filter media are sand and anthracite [27]. Furthermore, to protect the reverse osmosis systems from particle fouling, cartridge filters with a pore size range between 1 and 20  $\mu m$  are applied after the media filter [25].

Low pressure membrane filtration such as microfiltration (MF) and ultrafiltration (UF) has gained importance in the past years as pretreatment for reverse osmosis systems. Although MF and UF pre-treatment removes very well the bacterial cells from the feed water, biodegradable nutrients can pass UF membranes enabling eventual microbial growth in the subsequent RO installation.

In some cases, activated carbon or biofiltration is used to remove dissolved organic matter from the feed water. Activated carbon adsorption, either in granular or powder form, has also been considered as a feasible mean for reducing membrane fouling, either alone or in combination with other pre-treatment processes [28–30]. Chinu et al. [31] delayed fouling development in a lab scale setup by using biofiltration as pre-treatment using real seawater.

To protect the reverse osmosis membranes from biological fouling the raw water is usually disinfected by addition strong oxidant, such as chlorine, monochloramine, hypochlorite, chlorine dioxide, ozone, or UV irradiation.

In case of addition of chemical oxidants such as chlorine a further step is required. Since the reverse osmosis membranes are not resistant to oxidants, residual chlorine has to be removed from the raw water prior to entering the RO membrane system, commonly achieved by

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