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## Long-term effect on membrane fouling in a new membrane bioreactor as a pretreatment to seawater desalination

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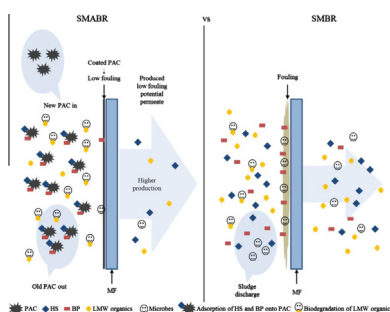
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### HIGHLIGHTS

- The semi-pilot scale SMABRs were operated on-site for a long-term.
- LMW organics responsible for biofouling of RO membrane were biodegraded.
- Biofouling reduction was validated with a rapid bioluminescence AOC measurement.
- Linear correlation between LMW organics and AOC concentration was observed.
- PAC helped to reduce fouling by coating on membrane without any damage.

### GRAPHICAL ABSTRACT

Comparison of SMABR and SMBR.



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### ABSTRACT

Submerged membrane adsorption bio-reactors (SMABR) were investigated as a new pretreatment for seawater reverse osmosis (SWRO) desalination. They were tested with different doses of powder activated carbon (PAC) on-site for a long-term. The biofouling on the membrane was assessed in terms of DNA (cells) and polysaccharide distribution. MBR without PAC addition resulted in severe fouling on membrane. When PAC is added in the MBR, PAC could reduce the organic fouling. Hence the biofilm formation on membrane was reduced without any membrane damage. PAC also helped to remove low molecular weight (LMW) organics responsible for biofouling of RO membrane. A linear correlation between assimilable organic carbon (AOC) and LMW organics was observed. A small amount of PAC (2.4–8.0 g of PAC/m<sup>3</sup> of seawater) was sufficient to reduce biofouling. It indicated that SMABR is an environmentally-friendly biological pretreatment to reduce biofouling for SWRO.

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**Abbreviations:** AOC, assimilable organic carbon; ATP, adenosine tri-phosphate; BB, building blocks; BP, biopolymers; CLSM, confocal laser scanning microscopy; ConA, Concanavalin A; DAPI, 4'6-diamidino-2-phenylindole; DO, dissolved oxygen; DOC, dissolved organic carbon; EPSS, extracellular polysaccharides; HMW, high molecular weight; HPI-DOC, hydrophilic DOC; HRT, Hydraulic retention time; HS, humic substances; LC-OCD, liquid chromatography with organic carbon detection; LMW, low molecular weight; MBR, membrane bio-reactor; MF, microfiltration; PAC, powder activated carbon; PS, polysaccharide; RO, reverse osmosis; RSW, raw seawater; SEM, scanning electron microscope; SMABR, submerged membrane adsorption bio-reactor; SMBR, submerged membrane bio-reactor; SRT, sludge retention time; SW, seawater; SWRO, seawater reverse osmosis; TMP, trans-membrane pressure; UF, ultrafiltration.

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## 1. Introduction

The membrane bioreactor (MBR) process was introduced in the late 1960s and its use has been increasing with the development of commercial ultrafiltration (UF) and microfiltration (MF) membranes and with the development of submerged MBR. Recent innovations in membrane technology and significant membrane cost reduction have enabled MBR to become an established process option to treat wastewater (Bouhabila et al., 2001; Sonune and Ghate, 2004). As a result, the MBR process has now become an attractive option for the treatment and reuse of industrial and municipal wastewaters, as evidenced by their constantly rising numbers and capacities (Rosenberger et al., 2005; Santos et al., 2011).

Recently, seawater has been attracted as a promising alternative source for drinking water. Reverse osmosis (RO) is considered as the most efficient technique for seawater desalination. However, RO application in desalination is not free from membrane fouling that adversely affects the performance efficiency in RO plants. Especially, biofouling will cause a decrease in membrane flux which in turn will result in an increase in operational pressure, and an increase in the frequency of membrane cleanings. This incurs a higher energy demand (Guo et al., 2012; Khan et al., 2011). Biofouling is caused by the attachment and growth of bacteria and accumulation of the bacterial metabolic products such as extracellular polysaccharides (EPSs), proteins, and lipids on the membrane surface. Once the bacterial cells deposit onto the surface, minimal amounts of nutrients are sufficient for cells to produce biofilm (Chen et al., 2013; Vrouwenvelder et al., 2011). Based on the specific bacterial characteristics and stages in biofilm growth, biofilm distribution on the surface ranges from uneven, discontinuous colonies to bulky, continuous films.

The main goal of the pretreatment process is to protect the RO membranes by reducing the fouling propensity of seawater (Cheng et al., 2013; Valavala et al., 2011). Conventional pretreatment uses deep bed filtration and cartridge filtration to remove particulate matter prior to the entry of seawater to the RO unit. MF membrane used as a pretreatment reduces turbidity and removes suspended solids and bacteria. However, such pretreatment methods cannot control biofouling completely (Kumar et al., 2006).

MBR is a suitable and environmentally friendly option for effective control of biofouling (Meng et al., 2009). This is because that MBR can remove the biodegradable organic matter and at the same time inactivate the microbes. Alternative MBR technology is also being tested in seawater or brackish treatment as a pretreatment system to solve this significant problem of biofouling in desalination process (Achilli et al., 2009; Kitade et al., 2013). Our group investigated a lab-scale MBR with powder activated carbon (PAC) addition (referred to as SMABR) as a pretreatment to RO (Jeong et al., 2013a). In MBR, PAC was used for organic adsorption and microbial development to degrade the organics adsorbed onto PAC. Only 2.14 g of PAC was required to treat 1 m<sup>3</sup> of seawater. SMABR removed a majority of organics up to 72% with only a marginal increase of trans-membrane pressure (TMP). Further it provided a stable bioactivity. This pretreatment also helped to increase the initial permeate flux of RO by 20% (by 6.2 L/m<sup>2</sup> h, LMH) compared to RO operation without pretreatment (Jeong et al., 2012a). The permeate flux decline in RO operation was only 34% during 45 h run. Apart from this, less foulant was found on the RO membrane (consisting of less biopolymer). The positive results obtained from these preliminary studies demonstrated the necessity for a detailed and a long-term study using a semi pilot scale-MBR as a sustainable pre-treatment to reduce biofouling. Also, so far this technology has not yet been applied as a pretreatment in SWRO desalination and only few papers can be found in the literature.

In MBR system, biofilms on partially or totally submerged membrane surfaces can promote or discourage the settlement of

microorganisms (Lee et al., 2001; Hwang et al., 2012). Further, a wide range of microbial products from low molecular weight (LMW) metabolites to high molecular weight (HMW) exopolymers (or biopolymers) including EPSs are generated. HMW organics can be retained by the membrane, leading to an accumulation of dissolved organic matter (DOM) in the reactor. The physical and chemical nature of biopolymers can also play an important role in the subsequent attachment of microorganisms. A previous paper observed EPSs from marine environment at early stages of biofouling (Ding et al., 2013). Thus, the major obstacle to the wide application of MBR technology is membrane fouling. Polysaccharide (PS) is assumed to be one of the major membrane foulants (Drews, 2010). Moreover, there is a linear relationship between fouling rate and PS concentration in the supernatant of MBR (Rosenberger et al., 2006). Therefore, understanding of PS fouling is critical for successful and widespread application of microfiltration (MF) membrane in MBR.

PS fouling is a complicated process due to the complex interaction between polysaccharide and membrane and between PS molecules. The fouling mechanisms of MF membrane have been attributed to membrane pore blocking and/or pore constriction during initial fouling, followed by cake formation on the membrane surface during long-term operation (Jeong et al., 2012b).

Thus, this paper focuses on a detailed investigation of long-term effect on membrane fouling in MBR. The reactors were operated in parallel for 56 days (8 weeks) on-site with continuous feeding of real seawater (Submerged membrane bioreactor, SMBR without PAC addition; SMABRs with 1.5 g/L and 5.0 g/L of PAC addition). During the operation, membrane performance was evaluated in terms of TMP development and organic removal. Biological activity was monitored in terms of adenosine tri-phosphate (ATP). Recently developed bioluminescence-assimilable organic carbon (AOC) method was used to detect the organic fraction which is utilized by microbes. The bio-available organic carbon results in the bio-fouling. In this study, LMW organic matter was related to AOC in effluent. Different positions of fouled membrane underwent a detailed investigation to study the role of PAC in MBR system for sustainable operation. DNA amount on MF membrane was quantified and PS distribution was also observed over time. Membrane surface was also observed using scanning electron microscope (SEM) technique.

## 2. Methods

### 2.1. Experimental design

The braid-reinforced hollow fibre MF (Cleanfil®-S, Kolon, Republic of Korea) used in the study comprised of three materials (Polysulfone, Polyethersulfone, Polyvinylidene Fluoride) as a coating layer. In the MBR system, a hollow fibre MF membrane module with 0.34 m<sup>2</sup> of an effective surface area and 0.9% of a packing density was used. The membranes were oriented in horizontal direction. The pore size of hollow fibre MF was 0.1 μm. The length of each fibre is 30 cm with an outer fibre diameter of 2.0 mm and an inner fibre diameter of 0.8 mm (total number of fibres in the module is 240). Membrane module was immersed in a tank with a working volume of 12 L (Fig. 1). Filtration was carried out in outside-to-inside direction. Aeration was made through a diffuser, at 5 L/min (0.9 m<sup>3</sup>/m<sup>2</sup> h), to help biodegradation and to keep the PAC with biomass in suspension. The permeate was withdrawn using a peristaltic pump. Backwashing was operated with the permeate at same permeate flux for 2 min once a day. The TMP (kPa) was measured using a pressure sensor.

As shown in Fig. 1, submerged membrane bioreactor (SMBR) was operated at 10 LMH without PAC addition. A 10 LMH was

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