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Osmotic effects of biofouling in reverse osmosis (RO) processes: Physical and physiological measurements and mechanisms

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ABSTRACT

Microbial biofilm formation on reverse osmosis (RO) membranes is known to reduce permeate flux and, in most cases, to reduce salt rejection. These effects are consequences of increased overall hydraulic resistance for water permeation through the membrane and a hindered back-diffusion of salts through the biofilm. In return, salt concentration near the membrane is elevated, a phenomenon known as "biofilm enhanced osmotic pressure" (BEOP), resulting in enhanced salt passage. While the effect of elevated hydraulic resistance is clear, the effect of salt passage increase is counterintuitive. In most cases tested, the typical increase of salt passage due to biofouling using commercial high-flux RO membranes cannot be attributed just to RO transport (permeate flux and salt rejection relation), and the typical values of salt passage elevation are too high under biofouling conditions and can only be explained by enhanced concentration polarization effects. Delineating biofouling mechanisms of RO membranes and analyzing the interrelated effects of the biofouling layer on the system performance as well as further changes in the biofouling layer physiology are important for monitoring the extent of biofouling in desalination processes. The BEOP phenomena is enlightened by synthetic biofouling controlled experiments as well as by more realistic studies using tertiary wastewater.

Keywords: Reverse osmosis; Biofouling; Fouling; BEOP; CEOP; Extracellular polymeric substances; EPS; Biofilm

1. Introduction

Water reliable and sufficient supply is essential for human health, modern industry, and agriculture; in fact it takes a part in any aspect of peoples' life. Throughout the world, clean waters, both for drinking and industrial uses are in short supply, more so in arid and semi-arid areas. Treated municipal wastewaters as well as brackish water are potential water resources from which high quality water can be produced by RO filtration. Also, the environmental need for RO filtration is emerging due to its highest treatment efficiency for removal of anthropogenic compounds including endocrine disrupting chemicals, pharmaceuticals, and personal care products [1]. The relatively low ionic strength and the related osmotic pressure of municipal wastewaters reduce the energy costs of filtration through RO membranes. However, fouling and more specifically, biofouling is a major challenge in wastewater reclamation [2,3]. The decrease in performance of RO membranes, due to fouling, in water reuse is a major concern [4–8]. Fouling requires frequent chemical cleaning and ultimately shortens membrane life, thus imposing a large economic burden on RO membrane plant operation. The major types of fouling in RO membranes

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are inorganic salt precipitation, organic, colloidal, and microbiological matter. While organic fouling and scaling increase hydraulic restriction for permeate flux, colloidal fouling and bacterial cells decrease permeate flux due to "cake- and biofilm-enhanced" osmotic pressure [9–12].

There are many efficient ways to reduce both organic and colloidal fouling. For example, conventional pretreatment for reclamation of wastewater with RO, in Fountain Valey, California, Water Factory 21 includes flocculation, lime or alum clarification, re-carbonation, settling, filtration, and activated carbon adsorption. It is reported that 26% of the TOC was removed by the lime clarification and that 30–50% of the TOC was removed by adsorption to GAC [13]. Other pretreatments include microfiltration, ultrafiltration, hybrid process of chemical flocculation, and powdered activated carbon (PAC) followed by MF, UF, and GAC adsorption [14–17]. While the above well established pretreatment processes are appropriate solutions for organic and colloidal fouling, small organic molecules and salts can serve as nutrients for microbial growth in the non-sterile environment of the RO units. Therefore, the biofouling propensity of RO membranes in reclamation of tertiary wastewater or desalination processes is relatively high due to the residual amount of nutrients in the pretreated water used as the RO feed water. Hence, minimizing bacterial biofouling of RO membranes is critical for ensuring a long-term effectiveness and decrease in operation and maintenance costs. This paper describes some of the latest findings on the osmotic processes that take place during biofouling in the RO desalination process. It is clear that in order to develop strategies for biofouling control, a critical knowledge on RO biofouling mechanisms and the related effects on membrane performance are needed to be defined. This knowledge, interdisciplinary in its nature, is obtained by analyzing the physico-chemical properties of the RO membrane surface and the associated fouling layer, RO membrane performance, and RO biofilm physiology.

2. Biofouling of RO membranes: A biofilm phenomena

Biofilm formation is a developmental process that involves mainly bacterial deposition and irreversible adhesion, formation of micro-colonies, maturation, and dispersion of the cells back to their planktonic stage [18]. Recent studies show that biofilm formation is a highly regulated process with different genes being expressed at different stages [19,20]. Also, the dynamics of biofilm formation on RO membranes analyzed recently with laser scanning confocal microscope (LSCM) [21] showed that biofilm structure was changed during its formation. While the dynamic of these changes was faster than other reported biofilms [22], still, distinct steps that include bacterial attachment to the surface, followed by auto-aggregation, microcolony formation, maturation, and cell detachment were detected [21]. In other study, when a mutant of *Pseudomonas aeruginosa* that over-expresses alginate, an important polysaccharide in *P. aeruginosa* extracellular polymeric substances (EPS), was used as a membrane colonizer, the biofouling process was delayed due to a decreased deposition [23]. Consistent with biofilm formation developmental phenomenon, the over-production of EPS, an important matrix in the mature biofilms, is observed not to enhance bacterial adhesion but rather to decelerate bacterial attachment process implying that the timing of EPS production is not coincidental. In fact, induced EPS expression, in most cases, takes place after cell attachment, at later stages of the biofilm formation process [24–26]. Since biofilm formation is a well-established developmental process, the timing of the different steps plays an important role in the biofouling process: An interference with biofilm formation stages on RO membranes should be further investigated for development of other possible biofouling control strategies.

3. Are biofilms on RO membranes physiologically unique?

The presence of increased amount of dissolved nutrients close to the membrane surface due to concentration polarization has been shown previously [21]. Using monoculture of *Pseudomonas aeruginosa* in a "proof of concept" study, the growth rate (analyzed *in situ* using a growth-dependent-GFP tagged strain) near the membrane was significantly higher in comparison to the cells that were close to the bulk liquid. Also, gene expression pattern as well as motility and chemotaxis phenotypes of the *P. aeruginosa* sessile culture on an RO membrane was more typical to a faster growing culture comparing to the planktonic cells in the RO unit. In RO desalination processes there is always limited access to dissolved nutrients in the bulk liquid. Hence, due to concentration polarization, faster growing cells will be present in the biofilm layer close to the membrane surface. This aforementioned sessile culture, is so far, the first culture reported that comprised faster growing cells in comparison to an associated planktonic cells and their transcriptomes were defined with DNA microarrays [21]: genes previously reported to be induced in laboratory *P. aeruginosa* biofilms were repressed in the RO biofilms. The repression of genes related to stress, adaptation, chemotaxis, and resistance to antimicrobial agents in the RO biofilm cells [21] raised an important question to the biofilm research community: which physiological changes in biofilm cells relate to the aggregated mode of cell growth (or "biofilm lifestyle"), and which are attributed to changes in environmental conditions (such as concentration polarization of nutrients adjacent to the RO membrane surface)? For example, how come that for *P. aeruginosa* biofilms that grow on RO membrane both phenotypic and genotypic responses showed a reduced chemotaxis [21], while other studies show an increase in chemotaxis response in non-penetrated biofilms that intrinsically face lower nutritional conditions [19,27]? The answer to that question suggests that chemotaxis does not relate to biofilm lifestyle but to the actual nutrients availability to the cells regardless their sessile or planktonic mode of growth.

The faster growth rate of RO biofilms was also shown in other studies: Huertas et al. [28] showed faster growing bacteria in close proximity to nanofiltration (NF) and RO membranes with mono-culture biofilm; Herzberg et al. [29] were using simple live-dead staining of a consortium that was growing on the expense of secondary wastewater and also showed the same differentiation of cell viability in the depth of RO biofilms: Faster and viable growing bacteria close to the surface due to concentration polarization. Fig. 1 shows examples from different biofouling studies in which faster growing cells were analyzed in close proximity to the membrane surface.

In short, the limited nutrient availability prevails in RO biofilms dictates faster growing cells near the membrane surface and higher fraction of dead cells close to the bulk liquid in the outer layers of the biofilm. This simple observation renders biofilms on RO and nanofiltration (NF) membranes as unique cases where the sessile biofilm communities grow faster than their associate planktonic culture, which evidently express a unique transcriptome profile compare to "traditional" non-permeable biofilm where nutrient mass transfer is limited.

4. Role of EPS, cells, and particulate matter in RO biofouling processes

In addition to the apparent changes in biofilm physiology on RO membranes, physical effects of BEOP on membrane performance include interrelated effects of microbial biofilm components on RO membrane performance, permeate flux and salt rejection. While the microorganisms in the biofilm matrix induce elevation of salt concentration near the membrane surface, their secreted biopolymers, the extracellular polymeric substances (EPS), induce elevation of hydraulic resistance to water permeation [9,30]. In the study of Herzberg and Elimelech [9], the deposited bacterial cells on the membrane increased the trans-membrane osmotic pressure, thus, enhanced the concentration polarization of salt near the membrane surface. The so called BEOP phenomena were also observed by Huertas et al. [28] as well as by Chong et al. [31] who showed that the BEOP contribution to permeate flux decline in most cases exceeds 50%. Recently, the contribution of EPS was shown only to elevate the hydraulic resistance of the RO membrane to water transport [30]. Surprisingly, the EPS layer, which its majority is water, was inducing a dramatic increase in the hydraulic resistance of the membrane [30]. This observation, somewhat is similar to observations of fouling of RO membranes by polysaccharides, such as alginate [32].

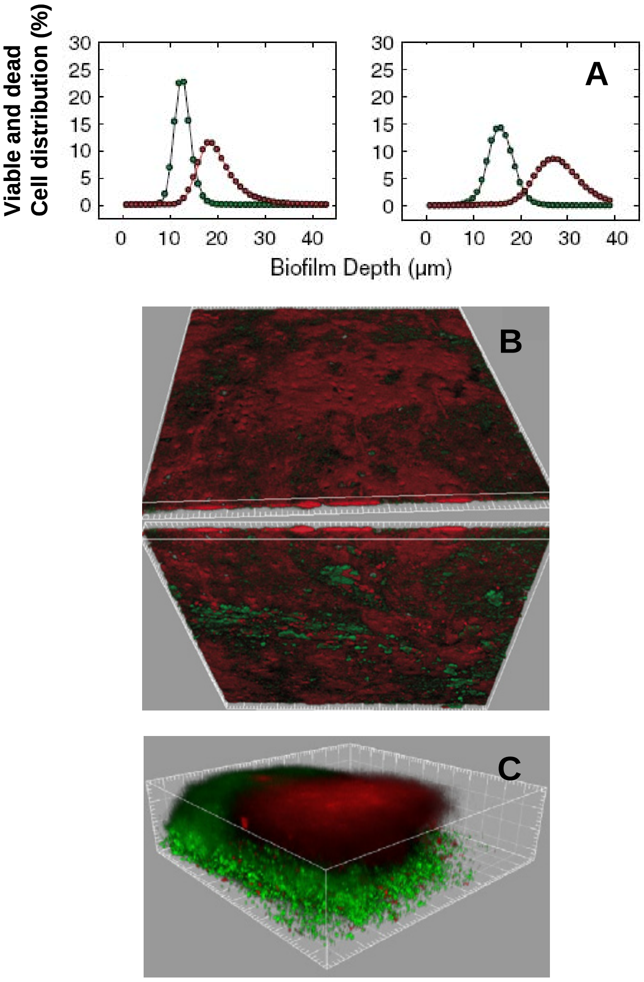

Fig. 1. Different examples of spatial distribution of cell viability in biofilms grown on RO membrane surface during desalination of synthetic and real secondary wastewater: (A) Distribution of dead (indicated with red propidium iodide) and live (indicated with green fluorescent protein controlled by growth-dependent promoter) cells in *P. aeruginosa* PAO1 biofouling layer on RO surface [source: [21]]; (B) Perspective view (750 μm × 750 μm) of a biofouling layer of *P. aeruginosa* chromosomally tagged with short-life green fluorescent protein and counterstained with propidium iodide: the top picture is taken with LSCM from the bulk-biofilm interface and the bottom picture is taken from the biofilm-membrane interface [source: [28]]; (C) Perspective view (128 μm × 128 μm) of a biofouling layer comprised from microbial consortium grown on the expense secondary effluents [source: [29]]. Top side is the bulk liquid side where mainly dead cells (indicated by red propidium iodide) and bottom side is the membrane side where mainly live cells (indicated by SYTO 9).

Nevertheless, in many cases EPS can also counteract the BEOP effect and act as additional "barrier" that reduce salt transport through the fouled membrane. In fact, biofouling layer comprised of a characterized microbial consortium was shown to increase salt rejection only after removal of particles (>0.45 μm) as a pretreatment with a microfiltration (MF) membrane [29]. The additional "barrier" formed by EPS was probably the main reason for the reduced salt mass transport through the fouled membrane as shown also for organic fouling [33]. Interestingly, when particulate mater was not filtered out from the feed solution, simultaneous increase in salt passage from 1 to 3% and faster permeate flux decline were detected. The more evident adverse effects of the combined biofilm and particulate layer than biofilm layer alone highlight the synergistic effects of organic and colloidal fouling, in which membrane performance is extremely reduced [34,35].

Another example of an induced reduction in salt rejection was also observed recently in our lab during RO desalination of brackish water using antiscalants: While on one hand antiscalants prevent membrane scaling, on the other hand, antiscalants that are based on polyacrylates can enhance bacterial and colloidal deposition [36]. We have analyzed these antiscalants effects and corroborating with previous studies, also here, as a result of an increased bacterial deposition after polyacrylate treatment, salt passage was increased from 1 to 3.5% and biofouling process was enhanced [36]. While this reduced salt rejection could not be attributed just to RO transport (permeate flux and salt rejection relation), and these typical values of salt passage elevation are too high under biofouling conditions, these effects can only be explained by biofilm enhanced concentration polarization phenomenon [9,10,30].

5. Implications and concluding remarks

Even small concentration of particles and colloids can enhance concentration polarization: The reduced back-diffusion of salts through biofouling layers is mainly a result of the presence of bacteria, particles, or colloids in the RO feed water. Appropriate pretreatment that should remove nutrients for biofilm growth, i.e., changing the location of the biofilm reactor to an alternative location rather than the membrane surface, has been suggested long ago [37,38]. Obviously, colloidal and particulate matter need to be removed for efficient operation of RO systems [39]. The key points presented here are the inter-related effects of the different types of foulants: In order to reduce BEOP effects, particles, colloids, and microorganisms need to be removed from the feed water. Biofilm growth on the RO membrane cannot be eliminated completely and only needs to be controlled to a point that a lowest interference with the operation of the RO plant will be required. Still, once biofouling occurs, faster cell growth will enhance the decrease in salt rejection due to BEOP effects. The presence of EPS, in most cases will induce a reduction of membrane hydraulic permeability and can also act as a secondary membrane that will lessen the BEOP effects and salt rejection will improve.

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