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The effect of imposed flux on biofouling in reverse osmosis: Role of concentration polarisation and biofilm enhanced osmotic pressure phenomena

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ABSTRACT

This paper describes a systematic study of biofouling in reverse osmosis process using model bacteria of *Pseudomonas fluorescens* and employing a sodium chloride tracer response technique for fouling characterization. It was found that the growth of biofilm at constant flux following initial bacteria colonization of the membrane surface increased with imposed flux. The rationale was that biofilm growth was nutrient dependent, where the nutrient availability at the membrane wall was controlled by the magnitude of concentration polarization, which is driven by flux. The salt tracer response showed that the biofouling comprised a hydraulic resistance and induced an enhanced osmotic pressure phenomenon; known as the biofilm enhanced osmotic pressure (BEOP) effect [M. Herzberg, M. Elimelech, Biofouling of reverse osmosis membranes: role of biofilm-enhanced osmotic pressure, Journal of Membrane Science 295 (2007) 11–20], due to hindered back diffusion of solutes through the tortuous path of the heterogeneous structure of the biofilm. For the conditions studied, the contribution of BEOP to transmembrane pressure increase was greater than the hydraulic resistance.

1. Introduction

Biofouling has long been recognized as one of the most problematic types of fouling in the reverse osmosis (RO) process. Even after a 99.9–99.99% removal of bacteria by the use of microfiltration (MF) as the pretreatment step, biofouling in RO cannot be eliminated as it only requires a few initial colonies on the membrane surface to form a mature biofilm [2,3]. In fact it has been suggested that the majority of the bacterial population involved in biofilm formation are viable but non-culturable (VBNC) and are about 0.2 μm in size, making them difficult to remove [4]. In order to survive, these recalcitrant bacteria adhere to the membrane surface, proliferate and secrete extracellular polymeric substances (EPS), and eventually form a mature biofilm. The situation is aggravated by poor anti-fouling strategies that tend to target only the microorganisms but not the EPS, which is the foundation of a biofilm [3,5]. Since it seems virtually impossible to eradicate the biofouling problem in the RO process, a better understanding of biofouling development is crucial if effective tactics are to be implemented to minimize the impact of biofouling on the membrane performance.

Over the years, many studies have been carried out to offer insight into biofouling in membrane processes (MF/UF/NF/RO). Most of the research work has focused on the initial stage of fouling, which is governed by the transport of bacteria cells to the membrane surface, followed by attachment. Commonly, the micron-sized bacteria are treated as colloidal particles, such that the transport of bacteria is analogous to a simple colloidal transport mechanism, e.g. the shear-induced diffusion model [6]. However, it should be remembered that in reality bacteria cells are semi-solids and have an overall irregular shape due to the flagella, which can assist in the bacteria movement. Nevertheless, the classical Derjaguin-Landau-Verwey-Overbeek (DLVO) theory for colloid stability, which accounts for van der Waals and repulsive electrostatic double layer interaction [7,8] and Lewis acid–base (AB) interaction [9], has been applied to explain bacteria attachment onto a membrane surface [10,11]. The primary role of EPS in the attachment process should not be excluded, as the actual component that comes into contact with the membrane surface is indeed the EPS. Notwithstanding the complexity in the nature of bacterial transport and attachment, this step could be described by the concept of critical flux [12]. However, it may be unrealistic to have a critical flux value for biofouling under prolonged operation of membrane processes. Instead, a sustainable flux concept [13] seems more appropriate, where the growth of the biofilm is considered acceptable; and no frequent cleaning of membrane is required.

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Apart from its influence on the transport and attachment step, solute concentration polarization (CP) in RO is also a function of flux, J_v, as well as the mass transfer coefficient, k_m, via the film model:

$$\mathrm{CP} = \frac{C_w - C_p}{C_b - C_p} = \exp\left(\frac{J_v}{k_m}\right) \quad (1)$$

Thus, by varying J_v and k_m, a different magnitude of CP is obtained, which determines the concentration of solutes and nutrients at the membrane surface. This is important as the attached bacteria multiply at the expense of local nutrient. So biofilm growth could be significantly accelerated at high CP level. However, the relationship between biofilm development and flux-mass transfer (or CP) in a RO system is still poorly understood.

The build up of biofilm on the membrane surface induces a hydraulic resistance (R_f) to the permeation. In addition, since the biofilm is comprised of micron-sized bacteria cells embedded in a matrix of EPS [14], the retained solutes now have to diffuse through the tortuous path of the deposit in order to back transport to the bulk fluid. A reduction in the effective diffusion means an increase in the CP and hence an enhanced osmotic pressure. This situation is similar to that of colloidal fouling [15–18]. In this case, the enhanced osmotic pressure phenomenon is known as biofilm enhanced osmotic pressure (BEOP), as reported in a recent work [1]. The consequence of the BEOP effect is an additional membrane performance loss on top of the loss caused by the biofilm hydraulic resistance. More importantly, the enhanced CP could induce a higher concentration of nutrient at the membrane surface and this would further exacerbate the biofilm growth.

However, only one study of the BEOP phenomenon has been reported in the literature [1]. In that work, a high cell density of late exponential culture of *Pseudomonas aeruginosa* PA01 was inoculated into the RO system with a supply of synthetic wastewater medium (~77 ppm TOC) to accelerate biofouling under low crossflow and high initial flux conditions. There was 80% decline in flux in a 24 h interval. This accelerated biofouling process occurred under conditions that did not mirror the actual biofilms found in practical RO operation as pointed out by the authors, where biofilms are formed through a series of sequential events such as transport, attachment, cell multiplication, secretion of EPS, biofilm maturation and detachment. Also, in this study, it was possible that the build up of the biofilm was mainly contributed by the accumulation (filtration) of suspended bacteria cells rather than from the steady growth of attached cells on the membrane surface.

In addition, the above reported work did not give a quantitative measurement of the actual CP values. The mathematical model developed for enhanced osmotic pressure estimation in the author's previous work [15–17], which was based on uniform colloidal cake properties, could not be applied here. The BEOP phenomenon in biofouling was deduced from the flux decline data as well as scanning electron microscopy (SEM) image analysis. It was further concluded that the cells (without EPS) contributed to BEOP but the contribution of EPS was mainly through an increase in the hydraulic resistance. It should be noted that this statement is not in agreement with our previous work [18] where a fouling layer of alginate (used as surrogate for EPS) was found to cause an enhanced osmotic pressure effect. Therefore more work is required to clarify the contribution of resistance and the BEOP effect in biofouling of RO membranes.

In this study, a systematic approach was developed to study biofouling in RO. It is difficult to use real wastewater as a feed to study biofouling in the laboratory due to great variations in culture concentrations and compositions over time. This is non-controlled, and non-controllable, which complicates the development of biofouling. To simplify the study, a representative species and a laboratory culture media were used. *Pseudomonas fluorescens* was selected as the model bacteria since *Pseudomonas* was the most common species isolated from real world biofilms on RO membranes [19,20]. A steady and continuous low density of culture from a chemostat was injected into the RO system with background ions of sodium chloride as well as Nutrient Broth (NB) as the carbon source to allow the deposition of bacteria cells and subsequent growth of biofilm on the membrane surface. The system was subjected to different permeate fluxes and crossflow conditions. Here, biofilms were formed in long-term (150 h) tests.

The experimental work comprised two parts. Firstly, the bacteria growth kinetics was studied in batch tests to determine the growth constants at different nutrient concentrations and salinities. This was important as nutrient and salinity variations can occur at the membrane wall due to the dynamics of concentration polarization effects. The second part was the preparation of a chemostat for delivering a continuous and steady culture supply to the RO unit in the biofouling studies to investigate the effect of CP on biofilm growth, biofilm resistance and the BEOP phenomenon. The sodium chloride tracer response technique, developed to elucidate the enhanced CP effects in a colloidal cake layer [18], was applied here to probe the BEOP phenomenon in a biofilm.

2. Theory

Details of the sodium chloride tracer response technique were presented in our previous publication [18]. The technique makes use of the unique property of an RO membrane to reject sodium chloride salts. When a pulse of sodium chloride is injected into the system, this causes an increase in the osmotic pressure. In order to maintain constant flux operation, the trans-membrane pressure of the system has to be raised. The transient CP estimation is based on the classical osmotic pressure model for filtration, as such for constant flux operation,

$$\mathrm{CP} = \frac{\mathrm{TMP_s} - \mathrm{TMP}}{\Delta\Pi_{bs} - \Delta\Pi_b} \quad (2)$$

where $\mathrm{TMP_s}$ and TMP are the trans-membrane pressure of the system during and before the pulse, respectively; $\Delta\Pi_{bs}$ and $\Delta\Pi_b$ and are the osmotic pressure difference between the feed and permeate solutions during and before the pulse, respectively. All the parameters in Eq. (2) can be easily measured or determined assuming a linear function of osmotic pressure and concentration. It is assumed that the hydraulic resistance, R_f, remains constant during the duration (<30 min) of the pulse test.

3. Materials and methods

3.1. Bacteria batch growth tests

All preparation work was performed in a Class II Bio-safety Cabinet (Esco Micro, model Esco Smart Control) to minimise contamination. Glassware and solutions used in the tests were autoclaved in an autoclave (Hirayama, model HV-110) at 121 °C and 1 bar for 20 min.

The bacteria strain, *P. fluorescens* (American Type Culture Collection, ATCC 700830) that used in previous biofilm studies [21], was supplied by Dr. Zhuang Weiqin from the Environmental Engineering Research Centre at Nanyang Technological University in agar plate (solid medium) form. *Pseudomonas* was transferred and subcultured in a newly prepared Nutrient Broth (NB) agar plate made from 1 wt% Agar (Becton Dickinson, Bacto) and 8 g L^{-1} of Nutrient Broth (Becton Dickinson, Difco) to ensure healthy and uncontaminated cells. This was done on a weekly basis throughout the whole experimental period.