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Evolution of fouling during crossflow filtration of model EPS solutions

Y. Ye, P. Le Clech, V. Chen*, A.G. Fane

UNESCO Centre for Membrane Science and Technology, University of New South Wales, 2052 Sydney, NSW, Australia

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Abstract

Extracellular polymeric substances (EPS) are a major fouling component in membrane bioreactor (MBR) systems. For a better understanding of the fouling mechanisms of EPS, the evolution of fouling of sodium alginate, a microbial polysaccharide was studied during crossflow ultrafiltration and microfiltration. Incremental flux-stepping experiments and long-term subcritical flux filtration were carried out. A two-stage of transmembrane pressure (TMP) profile reported during long-term filtration of MBR effluents was also observed for alginate solutions. An initial slow and gradual TMP increase was followed by a sudden transition to a rapid TMP rise. Alginate transmission and deposition pattern supported the concept of flux redistribution among open pores as a cause of two steps of fouling profile. A desorption method with sodium hypochlorite was developed to characterise the temporal and spatial distribution of alginate along the membrane surface. The TMP rise was increased, not only due to the increase of alginate loading on the membrane (from 52 to 252 mg m^{-2} in days 1 and 6, respectively), but also because of the temporal increase of the specific cake resistance (from 5.9 to 16.1 $\times 10^{15}$ m kg^{-1} within 6 days). The fouling layer formed in the long-term subcritical flux operation appeared irreversible, while fouling layers formed in the short-term dead-end constant pressure or flux-stepping experiment showed greater reversibility. Membrane autopsy using FESEM technique also confirmed the mostly irreversible nature of the fouling layer during the long-term subcritical flux operation.

Keywords: Extracellular polymeric substance; Critical flux; Specific cake resistance; Membrane fouling; Membrane bioreactor

1. Introduction

The use of membrane bioreactor systems (MBR), the combination of membrane separation and activated sludge process, for wastewater treatment has drawn great interest from researchers and engineers. Despite its many advantages such as smaller footprint and better product quality, the wider application of MBR systems require better control of membrane fouling as foulant removal depends often on aeration with occasional chemical or physical cleaning [1]. Membrane fouling is inevitable due to the complexity of the biological feed, but can be limited by operating the system under appropriate hydraulic conditions.

The key to the identification of the appropriate operating conditions is the so-called "critical flux" (J_c) [2], below which there is no or negligible fouling above which fouling is observed. Several measurement techniques have been proposed to measure J_c such as mass balance calculation (with latex particles) [3] and direct observation through the membrane (DOTM) technique [4]. A more widely and conveniently used method is transmembrane pressure (TMP) measurement in flux-stepping experiments [2,5]. In this method, J_c is defined as the maximum flux for which the permeability remains constant, the permeability may be the same as for solvent ('strong' form of J_c) or less than for solvent ('weak' form). More recently, parameters such as TMP increase between flux step (ΔP), the rate of TMP increase in each flux step (dP/dt) and the permeability (K) were calculated to investigate the critical flux phenomena in more detail. It was found that all the critical parameters indicate roughly the same flux at which fouling starts to become significant [6].

The fouling potential of MBR biomass is high, mainly because of the large number of foulant species, populating the activated sludge. These comprise bacterial flocs, colloids and

* Corresponding author. Tel.: +61 2 9385 4813; fax: +61 2 9385 5966.
E-mail address: v.chen@unsw.edu.au (V. Chen).

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macromolecules (extracellular polymeric substances, EPS). In order to maintain sustainable filtration of MBR systems, the subcritical flux operation is preferred to avoid the rapid fouling indicated by TMP rise. However, extended filtrations under subcritical conditions always feature a relatively long period of slow TMP increase followed by a more severe TMP rise. This phenomenon is reproducible and depends on the imposed flux. Originally, this phenomenon was ascribed to an increase in feed viscosity caused by levels of EPS in the feed [7]. In a later study on a submerged flat sheet MBR, a model was developed assuming that fouling resistance is due to EPS deposition and that the foulant is compressible with an increasing specific cake resistance with increasing TMP [8]. The important role of EPS was confirmed by Cho and Fane using effluent from an anaerobic bioreactor [9]. They proposed an alternative explanation for the rapid TMP rise, suggesting that membrane pore closures and pore blocking by EPS cause an increase of the local flux in remaining open pores and exceed the critical flux of the feed solution resulting in a rapid TMP rise. More recently, Ognier et al. applied a similar local flux concept to model the two-stage of TMP increase. However, in their study, the increase of local flux was hypothesised to be caused by the decrease of pore number (pore blockage by particles) rather than decreasing pore diameters (adsorption or deposition by EPS) [10]. Despite these studies on long-term subcritical filtration, little evidence has been available to confirm and verify these hypotheses due to the complexity of the feeds used. Moreover, the contribution of EPS to the progression in membrane fouling is yet to be fully understood even though polysaccharides seems to play an important role in the evolution of two-stage fouling profile.

Detailed studies of EPS filtration under controlled flux can provide a better understanding of the fouling process in MBR operation as well as in other biotechnological areas. Experimentally, two forms of EPS solution: bonded (or extractable) EPS solution and soluble EPS (or soluble microbial products, SMP) have been identified. The chemical compositions of EPS are usually reported to be very heterogenous, consisting of mainly polysaccharides, protein and nucleic acids [11]. The amount of EPS present in the biomass solution depends on a large number of factors, including the physiology of the microorganisms, nutritional status and imposed environment created by the MBR system operation.

Constant flux filtration of protein, one of the main components of EPS, has been extensively studied, while research based on polysaccharide filtration remains relatively rare. The role of polysaccharides in membrane fouling is significant not only because of its relatively large amount in the EPS, but also because of its higher and wider molecular weight distributions than those of single component protein solutions. Some previous studies already showed the important relationship between fouling in MBR and the polysaccharide fraction present in supernatant [12–14]. In addition, high molecular weight polysaccharides presenting good thickening and gelling properties play a strong role in forming sticky hydrogels on membrane surface [15,16].

In order to achieve a better understanding of the fouling phenomena of MBR subcritical flux operation, alginate was used as the model foulant to exclude the influence of suspended solids (SS) and bacterial flocs and to present experimental data to verify previous hypothesis about membrane fouling in long-term MBR filtration. Alginate, a linear copolymer composed of 1–4 linked β-D-mannuronic acid, C-5 epimer and α-L-guluronic acid in varying proportions, is one of the main microbial polysaccharides [17]. *P. aeruginosa* alginates were used by Strathmann to investigate the function of EPS in biofilms [18]. In this paper, the fouling mechanisms of polysaccharides under the crossflow constant flux filtration are studied in detail by using alginate as the model solution. The critical flux of alginate was evaluated using varying membrane morphologies and operation conditions. The influence of bovine serum albumin (BSA) on alginate fouling and transmission behaviour was investigated. The specific cake resistance, cake compressibility the temporal and spatial distributions of alginate fouling are explored during the long-term subcritical flux experiments.

2. Methods and materials

2.1. Experimental

All experiments were performed in a typical crossflow filtration module with a 1 mm high, 25 mm wide and 210 mm long flow channel. A peristaltic pump in the permeate line was used to control the permeate flux. A pressure transducer connected to both feed and permeate sides of membrane was used to measure the TMP, while a flow sensor and balance were used to monitor the crossflow velocity (0.33 m/s unless otherwise indicated) and the permeate flux, respectively. Both the balance and pressure transducer were all connected to the computer for data recording.

Three different membranes were used in this study, being 30 kDa polyethersulfone (PES) membrane (Synder, US), 100 kDa PES membrane (Synder) and 0.22 μm hydrophilic polyvinylidenefluoride (PVDF) membrane (Millipore, Australia). Virgin membranes were used for each experiment.

Sodium alginate (Ajax Chemicals) was used as a model solution in this study. NaN_3 (0.02%) was added to the solution to prevent the growth of bacterial, particularly during the long-term subcritical filtrations. Alginate concentration were measured from the feed and the permeate solutions in terms of carbohydrate with a colorimetric method [19]. An accurate molecular weight distribution of alginate is difficult to analyze. But based on the filtration of alginate with 100 kDa membrane, no alginate was detected in the permeate, thus it indicates that the molecular weight of alginate used in this paper is higher than 100 kDa. The viscosity of alginate used in this study was found to be higher than the medium viscosity alginate (Sigma) and lower than the high viscosity alginate (Sigma), whose molecular