Forward osmosis membrane performance during simulated wastewater reclamation: Fouling mechanisms and fouling layer properties

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ABSTRACT

A thin-film composite (TFC) FO membrane was used to study the fouling characteristics of effluent organic matter (EfOM) fractions in treated wastewater effluent represented by alginate, bovine serum albumin, humic acid and octanoic acid. These model foulants were chosen to represent carbohydrates, proteins, humic substances and fatty acids. Inter foulant interactions and their influence on membrane flux loss was established by running series of fouling tests with feed solutions containing mixtures of model foulants. The obtained results demonstrated that under our experimental conditions humic acid (HA) and octanoic acid (OA) had no significant role on permeate flux loss during wastewater treatment over short periods. However, alginate and bovine serum albumin (BSA) and their mixtures caused significant total flux loss, through alginate-calcium complexation that led to the formation of a resistant gel layer on the membrane surface. Protein fouling was mainly attributed to multiple layer adsorption onto the polyamide membrane surface. Mixing alginate with BSA saw a further decline in permeate flux, worse than that caused individually by either of the foulants, and gives an indication of the synergistic effect between the two foulants. There were favourable inter-foulant interactions between the carbohydrates and the proteins that promoted the formation of hybrid aggregates that were deposited on the membrane surface and enhanced flux loss. The additional presence of humic acid to the mixture of BSA and alginate further aggravated membrane fouling. Polysaccharides and proteins were found to be the most dominant foulants during wastewater reclamation. The extent of interactions between the organic foulants had an effect on the fouling layer structure and its role in permeate flux loss.

1. Introduction

Wastewater treatment for the purposes of supplementing the continuously decreasing fresh water supplies has caught global attention amongst water authorities and researchers as a potential alternative fresh water source [1–3]. This has prompted several research studies that focus on the viability and efficiency of using such feed streams in membrane-based separation processes [4–6]. The potential benefit of using wastewater as a feed stream in water treatment include its constant availability through-out the year and it could also be a means of limiting contamination of aquatic environments from treated industrial effluent that contains various pollutants [7]. Apart from the inorganic compounds and particulates in wastewater effluent it also contains effluent organic matter (EfOM) which comprises a range of low- to high-molecular-weight organic compounds such as polysaccharides, proteins, humic and fulvic acids, organic acids and lipids [8,9]. Thus,

membrane filtration processes are ideal as advance treatment steps for such feed streams due to their excellent rejection abilities of a wide array of contaminants. However, one key drawback of using wastewater as a feed stream is the intense fouling that is mainly caused by fractions of EfOM [7,8,10].

Membrane fouling is a consequence of the interaction between the membrane and the feed solution, which maybe a complex mixture comprising of colloids, bacteria flocs and dissolved organic macromolecules [11]. The adverse impacts of fouling on membrane performance and the overall water treatment system is well documented in previous studies, and it includes reduced membrane lifespan, poor permeate quality, increased applied pressure and inflated overall treatment costs [5,12]. One strategy of minimising both energy consumption and membrane fouling during wastewater effluent treatment is the possibility of using forward osmosis membrane process as a pre-treatment step. This process is dependent on the solute concentration

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difference between the feed solution and draw solution that drives water across a semi-permeable membrane, therefore; there is no application of external hydraulic pressure and thus fouling is largely reversible [13].

In this work the thin film composite (TFC) membrane was chosen to conduct fouling tests using feed streams simulating secondary treated wastewater effluent. This water characteristically contain effluent organic matter (EfOM) which is known to contribute significantly to organic fouling of nanofiltration (NF) and reverse osmosis (RO) membranes in advanced wastewater reclamation processes [14–19]. The chemical complexity and heterogeneous nature EfOM presents a challenge to developing proper understanding of the role of inter-foulant interactions in permeate flux decline as well as fouling layer formation. Numerous findings have attributed the observed severe flux loss to synergistic effects between co-existing foulants [20]. Ang et al. [21] reported a synergistic fouling effect when RO membranes were fouled by both alginate and bovine serum albumin (BSA). Foulant-foulant adhesion forces were found to increase with calcium concentration. Zhao and co-workers then investigated the interactions between EfOM fractions and RO membranes [22]. It was revealed that hydrophilic neutral fractions, mainly composed of small sized carbohydrates resulted in the highest flux decline and exhibited highest affinity towards the membrane. However, in contrast to previous findings, they found that extracellular polymeric substances (EPS) did not cause significant flux loss.

In another study by Ang et al. [23] RO membranes were fouled by a mixture of organic foulants simulating wastewater effluent using alginate, BSA, Suwannee river natural organic matter (NOM) and octanoic acid. They found that membrane fouling was governed by alginate in the presence of Ca^{2+} which was in turn influenced by alginate aggregate size and conformation in the bulk solution. Li et al. [24] investigated a different influential factor where the role of RO membrane surface properties in membrane fouling by BSA and sodium alginate. Experimental results revealed that membrane surface roughness had the greatest effect on fouling by the biopolymers. A considerable synergistic effect was observed when both BSA and alginate co-existed in the same feed solution which was attributed to the large foulant particle sizes resulting from the formation of BSA-alginate aggregates. Another importance of membrane surfaces was reported by Wang and Tang [25], who observed that membranes with smoother and more hydrophilic surface turn to possess favourable electrostatic repulsion experienced less initial flux membrane fouling. The influence of membrane surface was also highlighted by Mo et al. [26], who found that protein-membrane interactions influenced the fouling behaviour of polyethersulfone ultrafiltration membranes.

The above mentioned studies offers insight onto the influence of both feed composition and membrane surfaces on membrane fouling using pressure driven processes (RO). However, the few studies conducted in forward osmosis offer little insight into membrane fouling mechanisms but rather a performance evaluation of the process in direct wastewater reclamation [27]. Linares et al. [28] assessed the efficiency of a submerged FO system to treat wastewater. Permeate flux loss was attributed formation of fouling layer on the membrane surface which was mainly composed of biopolymer-like substances. Another attempt to treat wastewater was made by Achilli et al. [29] where they utilized a submerged FO membrane inside a bioreactor and found that fouling was mild and permeate flux was almost completely restored using osmotic backwash.

This literature review has availed valuable information into membrane fouling behaviour and the characteristic fouling potential of wastewater effluent components. Proper understanding of the chemical composition of wastewater is important since it can help predict possible of reactions and interactions amongst organic foulants and inorganics [30–33]. However, the systematic understanding of the interactions between the different EfOM fractions is still poorly understood. The labile nature and complex structure of biological

foulants such as proteins brings additional challenges in fouling characterization. Thus, in this work we aim at investigating the exact inter foulant interactions that influences permeate flux decline and the fouling layer formation when the EfOM fractions co-exist in feed streams. Fouling tests were performed using the high performance polyamide modified TFC FO membrane with hydrophilic surfaces. Particular emphasis was made to identifying EfOM fractions with a superior role to permeate flux loss during wastewater effluent treatment by performing sequential and combined foulant experiments.

2. Experimental

2.1. Model foulants

Several parameters can be used to describe typical wastewater, and composition is one of the universally used parameter. Typical domestic wastewater effluent organic matter (EfOM) can be generally characterized by varying concentrations of proteins, carbohydrates, aquatic humic substances and oil/fats [2], thus for the purposes of this work we chose bovine serum albumin (BSA), sodium alginate (ALG), humic acid and octanoic acid to represent the previously mentioned EfOM fractions. These model organic foulants were supplied by Sigma-Aldrich, St Louis MO. Bovine serum albumin, sodium alginate and humic acid were received in powder form and stock solutions (1 g/L BSA; 2 g/L alginate; 2 g/L humic acid) were prepared by dissolving each foulant in deionised water. The stock solutions were continuously stirred over 24 h to ensure complete dissolution of each foulant and then kept at 4 °C. A stock solution of octanoic acid was also prepared in DI water (1 g/L) and its pH was adjusted to around 6.7 using 0.1 M NaOH prior to addition to the feed solution.

2.2. Forward osmosis membrane

The flat sheet thin-film composite membrane (OsMem™ thin-film composite (TFC-ES)) was the principal membrane used in all fouling tests and was supplied by Hydration Technologies Inc. (HTI), Albany, OR, USA. The measured intrinsic membrane separation parameters were as follows: salt rejection (*R*) was 98.2%, pure water permeability (*A*) was 1.17 L/m²h bar, salt permeability coefficient (*B*) was 0.2×10^{-7} m/s, and a membrane structural factor (*S*) of 1227 μL [34–36].

2.3. Foulant and gel layer characterization

2.3.1. Dynamic light scattering

Dynamic light scattering experiments were performed to measure the average hydrodynamic diameters of all four foulants using a Malvern Nanosizer (Malvern Instruments, UK). The same foulant concentration and ionic composition as those used for fouling experiments were used during size determination analysis. Macro-particle surface charge in the feed solution was monitored using the same instrument by measuring electrophoretic mobility which is then converted to zeta potential.

2.3.2. Microscopic characterization

The topology, structure and roughness of the TFC membrane and the fouling layers were characterized using an atomic force microscope; AFM, Alpha30 (WITec focus innovations, Ulm, Germany). Scanning electron microscopy (SEM) was also carried out to investigate the morphology of the fouling layers using a Jeol IT300 microscope (Jeol Ltd, Tokyo, Japan).

2.3.3. Streaming potential measurements

Streaming potential measurements were carried out with a SurPASS Electrokinetic Analyzer, (Anton PaarGmbH, Graz, Austria) to determine membrane surface charge before and after filtration experiments.