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Influence of organic, colloidal and combined fouling on NF rejection of NaCl and carbamazepine: Role of solute–foulant–membrane interactions and cake-enhanced concentration polarisation



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ABSTRACT

The effects of single and combined fouling by sodium alginate, latex and Al₂O₃ on membrane performance in terms of permeate flux, salt rejection and rejection of the pharmaceutical carbamazepine were evaluated in a laboratory-scale cross-flow filtration unit. Fouling resulted in different extents in permeate flux, salt rejection and carbamazepine rejection over time. Combined fouling resulted in larger flux declines, and the influence of Ca²⁺ addition on flux was pronounced. In contrast, the influence of combined fouling and Ca²⁺ addition on salt and carbamazepine rejection was limited. Several hypotheses were tested to assess the influence of fouling on carbamazepine rejection. Relating observed rejection results to the solution-diffusion model revealed insight in whether the effect of fouling on rejection was mainly flux-driven or not. For most foulants, rejection declined less than what was predicted as a function of flux by the solution-diffusion model, indicating that CECP is not the main mechanism responsible for the decline in rejection upon fouling, especially not for latex fouling layers (regardless of the presence of Ca²⁺). This was corroborated by CECP modelling data. Solute–membrane interaction energies revealed that carbamazepine had higher affinity for the foulants than for the clean membrane surface. As such, it could be hypothesised that for most fouling layers, the relatively small decrease in carbamazepine rejection was mainly due to the decline in flux and the formation of a dense fouling layer on the membrane surface, which had some rejection properties.

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1. Introduction

Nanofiltration (NF) and reverse osmosis (RO) are key processes widely used for the removal of ions and organics from contaminated feed water in process water, drinking water and wastewater treatment [1]. Although NF/RO membranes are effective contaminant removal barriers in the provision of high purity water, their main drawback is fouling, mostly by organic macromolecules or colloids which may not be removed during pre-treatment [2,3]. The organic and colloidal foulants get deposited on the membrane surface to form cake layers [4]. In addition to colloidal and organic fouling, bacterial colonies may also accumulate on and colonise the membrane surface and spacers, resulting in bio-fouling [4]. Some studies carried out on organic fouling of membranes

reported on the use of sodium alginate as a model polysaccharide and a typical model compound to mimic extracellular polymeric substances (EPS) excreted by biofilms [3,5,6]. In addition to sodium alginate, natural organic matter (NOM) isolated from specific water types (e.g., Suwannee River natural organic matter) and humic acid have also been used to represent organics in feed water that may potentially foul membranes [7,8]. The effect of the presence of divalent cations such as Ca²⁺ on membrane fouling propensity has also been intensively investigated. It has been shown that addition of Ca²⁺ in a feed solution containing organic foulants can result in severe decreases in permeate flux. For example, alginate can form a well organised structure in the presence of calcium ions due to the preferential binding of Ca²⁺ on the carboxylic functional groups of the alginate to form an “egg-box” structure. This results in a tight layer that significantly decreases permeate flux [9]. Also the influence of monovalent ions on fouling (e.g., by alginate) has been tested. Sodium ions have been reported to show slightly enhanced binding onto alginate compared to potassium ions [10] but the effect on flux decline is

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almost insignificant. Colloidal fouling of NF and RO membranes has also been extensively investigated [2,11–13]. Silica, latex, ferric hydroxide and aluminium oxide have been used as model colloidal foulants for fouling studies of both NF and RO membranes. These foulants result in a change in membrane performance in terms of decrease in permeate flux. Hydrophobic membranes were found to be fouled more by small colloids such as silica and aluminium in comparison to hydrophilic membranes [12]. Also, the rougher the membrane, the more prone it is found to be to colloidal fouling [2,13–15]. Physicochemical interactions between suspended colloids as well as between colloids and the membrane are fundamental in influencing membrane fouling as they determine attachment of colloids on the membrane [16]. The stability of colloids (which is related to foulant charge) has an overall influence on their potential in fouling membranes propensity. Stability of colloids is related to particle charge. The dividing line between stable and unstable suspension is generally put between +30 mV and –30 mV (with the range between the two indicating colloid suspension instability) [16]. Generally, stable colloidal suspensions foul less. Other foulants include cellulose [17], natural organic matter [18], humic acid [19–21], bovine serum albumin [19,22], fatty acids [19] and surfactants [20].

Some researchers have reported the decline in permeate flux and salt rejection to be due to cake enhanced concentration polarisation (CECP) more than the colloidal cake layer resistance [15,23,24]. The colloidal fouling layer is believed to prevent back diffusion of salt from the vicinity of the membrane surface resulting in increased salt concentration. The salts end up finding their way through the membrane hence lower rejection values.

The rejection of organics by NF membranes has also been reported in the literature [24–26]. Membrane pore size and charge, solute molecular weight, charge and polarity, feed solution chemistry [25] and solute–membrane interactions such as steric, electrostatic and hydrophobic effects have been reported to govern rejection [26]. The effect of fouling on rejection of organics has been reported. Depending on the foulant type, a decrease or increase in rejection may be observed [25,26]. Generally, increase in rejection after fouling was subjected to the formation of a more dense layer which serves as an additional fouling layer [26]. Discrepancies in solute rejection by fouled membranes compared to clean ones are subjected to cake enhanced concentration polarisation (CECP) [25,24]. However, we hypothesise that CECP might not always be the single factor, as there might be other parameters which may determine solute rejection. The factors may include changes in membrane surface properties due to fouling. What many authors fail to assess well is the simple operational consequence of a decrease in permeate flux due to fouling on rejection. They also fail to acknowledge the simple transport phenomenon that fouling results in reduced flux (when fouling is carried out at constant pressure), and following the simple solution-diffusion model, it is clear that a reduction in flux will result in reduced rejection. This study will show that CECP is not substantial for some foulants and it is not the sole component that explains poor membrane performance in terms of solute rejection upon fouling.

Despite tremendous research efforts on understanding membrane fouling and its effects on rejection of organics, the fundamental mechanisms in NF/RO membrane processes remain unknown due to their complexities. It is important to study the effects of combined organic and colloidal fouling in terms of rejection of neutral organics. Investigation of this effect is important because ideally, organic and colloidal foulants co-exist in the same feed water. Sodium alginate (SA), polystyrene carboxylated latex and Al_2O_3 were selected as surrogate foulants to get more insight into the effects of fouling on rejection. Alginate was chosen because it is a good representative of polysaccharides which are

ubiquitous in most source waters and it has been used by other researchers before. Latex is widely used in many applications such as adhesives, inks, paints, coating, drug delivery systems, floor polish, films and carpet packing. It was selected because it is likely to be detected in water sources if it is improperly disposed, and as a negatively charged colloid, is representative for other inorganic colloids in surface water. Al_2O_3 was used because of its similar particle hydrodynamic radius as latex but different charges (positively charged), in order to investigate the effect of colloid charge on fouling. Carbamazepine was used as a model trace organic, because it is a widely used as an anti-epileptic drug and has been frequently detected at trace level in raw and secondary treated sewage [8]. Its neutral charge gives an advantage of eliminating charge effects in investigating factors playing a role in its rejection, as those effects are not the subject of this study.

The thrust of the study lies in investigating the effects of combined organic and colloidal fouling on membrane filtration performance in terms of solute (salt and carbamazepine) rejection. The interest was in investigating whether the decline in solute rejection was due to CECP, a decline in permeate flux or a combination of both. The effect of flux decline on solute rejection was filtered out in order to validate if CECP was significant for the fouling experiments. The CECP model was used to predict solute rejections, and this was compared to observed rejections.

2. Materials and methods

2.1. Foulants and characterisation

Sodium Alginate (Sigma Aldrich, United Kingdom), polystyrene carboxylated latex (EOC, Oudenaarde, Belgium) and aluminium oxide (Evonik Degussa GmbH, Hanau-Wolfgang, Germany) were used as model foulants. Latex and Al_2O_3 were used as colloid model compounds with different charges but similar particle sizes of 154 nm and 186 nm respectively (see Table 1S in Supporting information).

All foulants were characterised for charge and size at neutral pH in a background electrolyte of 10 mM KCl. A Zetasizer 2C (Malvern Instruments, United Kingdom) was used to measure the electrophoretic mobility of the foulants. The electrophoretic mobility was then correlated to zeta-potential based on the Helmholtz–Smoluchowski equation

$$EM = \frac{(2\epsilon\zeta f(ka))}{3\eta} \quad (1)$$

where EM is the measured electrophoretic mobility, ζ is the zeta potential, $f(ka)$ is Henry's function, ϵ is the permittivity of water ($\text{C}^2 \text{N}^{-1} \text{m}^{-2}$) and η is the electrolyte viscosity (Pa s). Particle sizes were determined by means of dynamic light scattering (DLS) using photon correlation spectroscopy (PCS 100M, Zetasizer 2C, Malvern Instruments, England).

2.2. Model trace organic and analysis

Carbamazepine is an anticonvulsant and mood stabilizing drug commonly used in the treatment of epilepsy and bipolar disorders. Physico-chemical properties of the organic are listed in Table 1. Carbamazepine was analysed as total organic carbon (TOC) using a TOC analyser (Shimadzu TOC-VCSH, Shimadzu Scientific Instruments, USA) with a limit of detection of 0.1 mg/l. Carbamazepine was dosed in concentrations of 10 mg/l to be able to measure 98% rejection of carbamazepine. Influence of incomplete rejection of alginate and/or latex did not influence TOC measurements in the permeate since rejection of the foulants was complete (results not shown).

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