



Rejection of pharmaceuticals by nanofiltration (NF) membranes: Effect of fouling on rejection behaviour



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ABSTRACT

The aim of this study was to investigate the effects of membrane fouling by sodium alginate, latex and a combination of alginate + latex on the rejection behaviour of salts and organics. Sodium chloride and caffeine were selected to represent salts and organics, respectively. The effects of the presence of calcium chloride on the fouling behaviour and rejection of solutes were investigated. The results revealed that the salt rejection by virgin membranes was 47% while that of caffeine was 85%. Fouling by alginate, latex and combined alginate–latex resulted in flux decline of 25%, 37% and 17%, respectively. The addition of Ca^{2+} aggravated fouling and resulted in further flux decline to 37%. Fouling decreased salt rejection, an observation that was further aggravated by the addition on Ca^{2+} . However, it was also observed that fouling with alginate and calcium and with latex and calcium minimised salt rejection by 30% and 31%, respectively. This reduction in salt rejection was attributed to the decrease in permeate flux (since rejection is a function of flux). There was a slight increase in caffeine rejection when the membrane was fouled with latex particles. Moreover, the presence of foulants on the membrane resulted in a decrease in the surface charge of the membrane. The results of this study have shown that the NF 270 membrane can be used to treat water samples contaminated with caffeine and other organic compounds that have physicochemical properties similar to those of caffeine.

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1. Introduction

There have been numerous reports on the occurrence of organic pollutants in source waters, a trend that has alarmed consumers, and highlighted the need to design water purification technologies that can remove such contaminants from water sources and provide safe drinking water. This is due to concerns in terms of adverse health effects on humans as well as animals (Fernández et al., 2010). The presence of a number of pharmaceuticals has been reported in wastewater treatment plant effluents (Fernández et al., 2010). Examples of the reported pharmaceuticals include analgesics, central nervous system stimulants, anti-epileptics, metabolites, β -blockers and antibiotics (Van der Ven et al., 2006). There is therefore a need to devise purification technologies that can address the problems of the presence of pollutants in water so as to ensure compliance with effluent quality standards as set by the authorities. Membrane filtration with

respect to nanofiltration and reverse osmosis has been applied as a promising technology for the treatment of organic micro-pollutants that occur in contaminated water (Eriksson, 1988; Raman et al., 1994; Nystrom et al., 1995). A membrane acts as a semi-permeable barrier between polluted water and treated water. In pressure-driven membrane filtration processes (such as nanofiltration and reverse osmosis), pressure is applied to drive the contaminated water (also referred to as feed water) through the membrane to the permeate side. Due to selective properties of the membranes and other properties such as small pore size, large solutes are rejected and the treated water (the permeate) will thus have lower concentrations of the target solute than the feed. The rejection of trace organics by virgin membranes has also been investigated by other researchers (Kiso et al., 2001; Nghiem et al., 2004; Schäfer et al., 2003; Kimura et al., 2003; Bellona et al., 2004). Several factors have been found to play major roles in controlling the rejection behaviour. These factors include steric hindrance, Donnan exclusion, hydrophobic interactions as well as solute/membrane properties (Bellona et al., 2004; Nghiem et al., 2005). However, a combination of

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rejection mechanisms is possible for the removal of a single compound (Van der Bruggen et al., 2006). This makes it difficult to fully understand the dominant mechanism, leading to misinterpretation of observed results. Although membranes may prove to be a promising technology, they are highly prone to fouling by organic and colloidal foulants, as well as a combination of organic and colloidal foulants. Fouling has been reported to result in poor permeate quality, reduced permeate flux and shortened membrane life (Lee et al., 2004a,b; Li and Elimelech, 2004; Speth et al., 1998; Beverly et al., 2000). This is due to the change in the surface morphology and chemistry of the membrane which may influence the rejection of organics (Xu et al., 2006). Depending on the foulant type, the membrane becomes more hydrophobic and the surface charge changes (Roundman and DiGiano, 2000; Cho et al., 2000). Hydrophobic membranes may poorly reject hydrophobic organic compounds (due to membrane–solute hydrophobic interactions). Hydrophobic solutes are believed to adsorb onto hydrophobic membrane surfaces, and are then able to partition into the membrane as permeate when saturation is reached (Kimura et al., 2003; Verliefde et al., 2009).

The effects of fouling on the rejection of solutes have been previously investigated (Ng and Elimelech, 2004; Xu et al., 2006; Agenson and Urase, 2007; Nghiem and Hawkes, 2007; Verliefde et al., 2009; Yangali-Quintanilla et al., 2009; Vogel et al., 2010; Hajibabania et al., 2011; Fujioka et al., 2013; Mahlangu et al., 2014). Depending on the physicochemical properties of the solute, its rejection may be improved, remain unchanged or decline as a result of fouling. For example, the rejection of high-molecular-weight non-ionic hydrophilic compounds (which is governed by size exclusion mechanisms) is not altered due to fouling because size exclusion mechanisms are mostly independent of fouling (Hajibabania et al., 2011). A decrease in the rejection of small and hydrophilic neutral compounds as a result of fouling has been reported (Ng and Elimelech, 2004; Yangali-Quintanilla et al., 2009). Rejection of high-molecular-weight organic compounds such as caffeine, chloroform, bromoform and trichloroethylene has been found to increase as a result of fouling (Xu et al., 2006; Yangali-Quintanilla et al., 2009). Solute rejection has also been found to relate to solution pH and membrane type. For example, the rejection of sulphamethoxazole by a fouled NF 270 membrane was found to be increased at pH 6 and 8 while a decrease in rejection at pH 6 was measured for fouling of NF 90 membrane. There was no change in rejection at pH 8 (Nghiem and Hawkes, 2007). On the other hand, the rejection of ibuprofen and carbamazepine for fouling of NF 270 membranes declined (regardless of solution pH) while rejection by NF 90 membranes remained relatively unchanged. The rejection trends were found to be related to membrane pore sizes as well as membrane–solute charge interactions which are pH-dependent. The presence of a fouling layer on the membrane surface is believed to prevent back-diffusion of solutes of small molecular weight from the membrane surface to the feed (a concept known as cake-enhanced concentration polarisation, Lee et al., 2005). This is due to the creation of a tortuous path for diffusion. The solutes accumulate on the membrane surface leading to a concentration gradient across the membrane interface. To maintain a concentration equilibrium, the solute diffuses through the membrane to the permeate side leading to poor rejection. Evidence of these effects on the rejection of carbamazepine, bisphenol and sulphamethoxazole have been reported by Vogel and co-workers for fouling of polyamide NF 270 membrane with humic acid (Vogel et al., 2010). Recently Fujioka and co-workers investigated the effects of fouling of three membrane types (NF 90, ESPA2 and ESPAB) on the rejection of N-nitrosamine of molecular weights between 70 g mol^{-1} and 160 g mol^{-1} (Fujioka et al., 2013). The authors reported an increase in rejection upon fouling due to narrowed passage for the small solutes as a result of cake-

layer formation and membrane compaction. Mahlangu and co-authors found that although carbamazepine rejection by membranes fouled with colloidal latex declined over time, its rejection was higher than that of a virgin membrane when rejection was compared at similar fluxes (Mahlangu et al., 2014). This shows that rejection may also decline due to a reduction in permeate flux (as a result of fouling) and may be more pronounced than the CECF effects.

The aim of this study was to investigate the effects of organic, colloidal and combined organic/colloidal fouling of a nanofiltration membrane on the rejection of caffeine (a neutral organic compound). Sodium alginate and latex were used as representatives of organic and colloidal foulants, respectively. Most studies have only used one type of foulant to investigate the effect of fouling on rejection of organics (Xu et al., 2006; Nghiem and Hawkes, 2007; Agenson and Urase, 2007). The authors did not look at the effect of fouling in the presence of divalent cations on rejection of trace organics. The presence of divalent cations has been shown to influence organic fouling due to organic–calcium complexation. Therefore rejection of organics by membranes fouled with the addition of calcium may be different to that of fouling without the addition of the divalent cations. In actual water treatment, the feed water contains a cocktail of organic and colloidal foulants (in the presence or absence of calcium ions) in addition to other foulant types. It is therefore important to investigate combined organic/colloidal fouling effects in the presence of calcium ions also. Caffeine is a central nervous system (CNS) stimulant. It is the main component of coffee and it has been described as one of the most widespread pharmaceuticals in the environment (Nikolau et al., 2007). Excessive intake of caffeine may result in caffeine intoxication. The primary metabolite of caffeine is paraxanthine (Guerreiro et al., 2008). A high concentration of paraxanthine in the blood is believed to result in miscarriages in pregnant women (Klebanoff et al., 1999). It is therefore necessary to investigate the rejection of caffeine by a membrane before and after fouling to establish the effect of fouling.

It is important to gain an understanding of the effect of fouling on the performance of membranes since their application in water treatment for the production of safe drinking water has increased. The challenge is to understand fouling behaviour and identify key membrane parameters that change as a result of fouling. Once these features are identified, membranes with anti-fouling properties may be synthesised and applied in the treatment of water containing a wide range of pollutants and foulants.

2. Materials and methods

2.1. Model organic and colloidal foulants

Sodium alginate was selected as the model organic foulant and was obtained from Sigma–Aldrich (Johannesburg, South Africa). This organic foulant (dosed at 20 mg L^{-1}) has been reported to have a negative charge in solution due to the dissociation of the sodium ions exposing the negative carboxylic functional groups. The sodium alginate was dissolved in Milli-Q water (18.4 M-ohms) and the solution was allowed to stand for at least 24 h (or until complete dissolution) before commencement of the fouling experiments. Polystyrene carboxylated latex was used as the model colloidal foulant and was obtained from EOC, Oudenaarde, Belgium. The polystyrene carboxylated latex was dosed at concentrations of 30 mg L^{-1} . Latex is widely used in many applications such as adhesives, inks, paints, coating, drug delivery systems, floor polish, films and carpet packing. This foulant may end up in water sources due to improper disposal.

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