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# Factors governing combined fouling by organic and colloidal foulants in cross-flow nanofiltration



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## ABSTRACT

There is some controversy in the literature on the effects of combined fouling of organics and colloids on water flux. There are reports of more flux decline for combined fouling than that of fouling by individual foulants, which has been attributed to synergistic effects. However, some authors observed lower flux decline in combined fouling. In addition, some authors have reported on increased flux declines by calcium addition, whereas others have shown improved fluxes after calcium addition. The discrepancies observed in the literature are systematically investigated in more detail in this manuscript. The organic–colloid concentration ratio was varied in order to explain discrepancies in previously observed effects in combined fouling. Further, the probable contributing factor of colloid type and calcium concentration in combined fouling was also examined. Organic, colloidal and combined fouling was found to be aggravated in the presence of Ca<sup>2+</sup> due to organic–Ca<sup>2+</sup> complexation and the reduction in foulant–membrane repulsive interactions, but this only occurred up to certain Ca<sup>2+</sup>-concentrations. Above these concentrations, the opposite effect was observed. In addition, membrane–foulant as well as foulant–foulant affinity interactions were determined and related to initial and later fouling rates. A novel type of “sequential” fouling experiments were carried out for the first time, and yielded information on how colloids and organics interact when both are present in fouling solutions. It was concluded from these experiments that hindered back diffusion of organics by a layer of colloids was the main factor responsible for flux decline in combined fouling.

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## 1. Introduction

The application of NF/RO membranes in water filtration is challenged by fouling. Organic, colloidal and combined fouling are caused by the accumulation of rejected substances on the membrane surface to form a layer that changes the morphology and physico-chemical properties of the membrane surface, resulting in a significant change in membrane performance not only in terms of water flux, but also in solute rejection [1–6]. Literature on fouling in NF/RO is abundant, but mainly focuses on single foulant studies. Combined fouling is less investigated, and in the literature, there is not always clarity on whether flux decline for combined fouling by organics and colloids

is worse than just the addition of the single foulants. Some studies have observed more flux decline in combined fouling, due to synergistic effects between the foulants [7–10]. On the other hand a study by Lee et al. showed that actual flux decline due to combined fouling was less severe than that predicted from simple addition of the flux decline from the single organic (natural organic matter – NOM) and colloidal fouling, although this largely depended on solution chemistry [11]. Therefore differences in observations and conclusion made on the effects of combined fouling between different studies could be due to variations in the: (1) type (and thus the physico-chemical properties) of the organic and colloidal foulants used; (2) concentration of organics and colloids used; (3) ionic strength and/or concentration of multivalent ions used and (4) operation conditions. In addition to different fouling trends observed, several different fouling mechanisms have been proposed in combined fouling. The flux decline has been proposed to be due to: (i) increased hydraulic resistance imparted by the combined cake layer structure; (ii) hindered back diffusion of organics in the concentration polarisation layer

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(due to the presence of colloids); (iii) modification of the colloid surface properties due to organic adsorption and (iv) a combination of these mechanisms [7,8,11].

In order to shed more light on the different effects of combined fouling observed in the literature, combined fouling is more systematically investigated in this study. Different aspects of combined fouling are investigated for the first time: (1) effect of the type of organic foulants used: in this study, alginate is used as an organic foulant, and combined with two different types of colloids (silica and latex colloids) to foul nanofiltration (NF) membranes; (2) the effect of the concentrations of foulants used: it is also possible that the discrepancies in the literature are due to differences in concentrations of foulants used. This effect is investigated here as well by increasing the concentration of organics from 50 mg/l to 150 mg/l while keeping that of colloids constant at 100 mg/l. Later the concentration of colloids was increased from 50 mg/l to 150 mg/l at constant organic concentration of 100 mg/l; (3) the effect of multivalent ions. Although it has been shown before that  $\text{Ca}^{2+}$  has various effects on single and combined fouling, most studies using  $\text{Ca}^{2+}$  so far have focused on a single calcium concentration, and have compared conclusion with and without calcium. In this study, the effects of divalent cations on fouling are investigated by increasing calcium concentration while keeping foulant concentration constant. This is to elucidate why different observations on the effects of calcium addition have been seen in the literature: some authors have reported a higher degree of flux decline when fouling happens in the presence of  $\text{Ca}^{2+}$  due to formation of calcium bridges between the membrane surface and alginate molecules through binding on carboxylic groups of the membrane and that of alginate [5,12–16]. There are, however, also other studies that have observed less fouling when  $\text{Ca}^{2+}$  is present [17–20]. In those studies, this has been attributed to the formation of larger alginate aggregates when  $\text{Ca}^{2+}$  is present, leading to a lower cake resistance (a more permeable gel layer) and hence less flux decline compared to fouling when only alginate is present [19,20]. (4) Interactions between colloids and organics are further investigated in this study by carrying out a novel type of fouling experiments termed sequential fouling. In these experiments, the membrane is fouled with organics and colloids in alternating sequences. (5) Lastly, it is known that fouling is influenced by foulant–foulant as well as membrane–foulant interactions. For alginate, correlations between these interactions, calculated from contact angles of membranes and filtered lawns of foulants based on the Extended-Derjaguin–Landau–Verwey–Overbeek (XDLVO) approach, and flux declines have been shown before [21–23]. To evaluate if knowledge on these interactions can also be used to better understand combined fouling, these interactions were also determined here and related to initial and later fouling rates to shed some light on fouling trends observed.

## 2. Materials and methods

### 2.1. Model foulants and foulant characterisation

Sodium alginate (Sigma Aldrich, South Africa), polystyrene carboxylated latex (EOC, Oudenaarde, Belgium) and silica colloids (Nissan Chemicals, United States of America) were selected as model organic and colloidal foulants. Alginate is a good representative of polysaccharides which is ubiquitous in most source waters and has been widely used in previous fouling research [5,21,24]. Latex as a colloid has numerous applications in adhesives, inks, paints, coating, drug-delivery systems, floor polish, films and carpet packing. Silica with a molecular formula of  $\text{SiO}_2$ , is one of the abundant minerals on earth. In industry, silica is applied as a drainage aid in paper making. It is also used as a catalyst, absorbent, surfactant and wine and juice fining agent. Latex and silica detected in water sources is mainly due to its improper disposal by these industries.

The model foulants were characterised for zeta potential and size at a pH of 6.8 and 10 mM KCl using a Zetasizer 2C (Malvern Instruments, United Kingdom). Zeta potential measurements were based on electrophoretic mobility (EM) while foulant sizes were based on dynamic light scattering (DLS) techniques using photon correlation spectroscopy (PCS 100M, Zetasizer 2C, Malvern Instruments, England). The foulant zeta potential relates to the measured EM based on the Helmholtz–Smoluchowski equation (Eq. (1)) [25]:

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

where EM is the measured electrophoretic mobility ( $\mu \text{ m s}^{-1} \text{ cm V}^{-1}$ ),  $\zeta$  is the zeta potential (V),  $f(ka)$  is Henry's function,  $\epsilon$  is the permittivity of water ( $\text{C}^2 \text{ N}^{-1} \text{ m}^{-2}$ ) and  $\mu$  is the electrolyte viscosity (Pa s).

The foulants were also characterised for surface tension components by measuring their contact angles (DSA 10-MK2, Kruss, Germany) with three well characterised probe liquids of known surface tension components (water, glycerol and diiodomethane). The individual foulants (200 mg/l) were therefore deposited on the NF 270 membranes by filtering 200 ml solution in dead-end filtration system. These filtered lawns were then dried in a desiccator overnight before contact angle measurements. The sessile drop method was adopted for the contact angle measurements. A minimum of 15 drops per liquid were placed on the filtered lawns using a microlitre syringe. An average ( $\pm$  standard deviation) from the measured contact angles was calculated. From the measured contact angles, interaction energies were calculated (see Section 2.3).

### 2.2. Membranes and membrane characterisation

Dow-Filmtec NF-270 (Minneapolis, MN) polyamide membranes were used in this study. Upon receipt from the manufacturer the membranes were immediately stored at 4 °C to avoid drying and oxidation.

The zeta potential of the clean membrane was determined by measuring the streaming potential (the potential at the shear plane between the membrane and the solution) using a self-assembled streaming potential analyser at a background electrolyte of 10 mM KCl. The measuring unit is equipped with a 6B11 analog digital converter (Analog Devices GmbH, Germany). The converter has an accuracy of 0.01 mV. The streaming potential cell has the following dimensions: channel length of  $6.42 \times 10^{-2}$  m, channel width of  $2.54 \times 10^{-2}$  m and channel height of  $5 \times 10^{-4}$  m. The tangential mode of analysis was used at a pressure range of  $-1$  to  $-20$  kPa. Briefly, the test solution is allowed to flow through the membrane surface by opening the valve. During the flow mode, a streaming potential (mV) is recorded in the data logger. A second measurement is made in non-flow mode (i.e. when the valve is closed). The difference between the potentials in flow and non-flow modes is used in the calculation of the membrane zeta potential based on Eq. (2):

$$\zeta = \frac{\Delta V \mu \delta}{\Delta P \epsilon} \quad (2)$$

where  $\Delta V$  is the measured streaming potential (V),  $\Delta P$  is the applied pressure (Pa) and  $\delta$  is the electrolyte conductivity ( $\mu\text{S}/\text{cm}$ ). Measured zeta potentials were useful in predicting membrane–foulant charge interactions which were related to membrane fouling.

Before fouling, contact angles of membranes (previously dried in desiccator) were measured using a goniometer (DSA 10-MK2, Kruss, Germany) as described above for the foulants. From the measured contact angles, membrane–foulant free energies of interactions were calculated (see Section 2.3).

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