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Trace organic solutes in closed-loop forward osmosis applications: Influence of membrane fouling and modeling of solute build-up

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ABSTRACT

In this study, trace organics transport in closed-loop forward osmosis (FO) systems was assessed. The FO systems considered, consisted of an FO unit and a nanofiltration (NF) or reverse osmosis (RO) unit, with the draw solution circulating between both units. The rejection of trace organics by FO, NF and RO was tested. It was found that the rejection rates of FO were generally comparable with NF and lower than RO rejection rates. To assess the influence of fouling in FO on trace organics rejection, FO membranes were fouled with sodium alginate, bovine serum albumin or by biofilm growth, after which trace organics rejection was tested. A negative influence of fouling on FO rejection was found which was limited in most cases, while it was significant for some compounds such as paracetamol and naproxen, indicating specific compound-foulant interactions. The transport mechanism of trace organics in FO was tested, in order to differentiate between diffusive and convective transport. The concentration of trace organics in the final product water and the build-up of trace organics in the draw solution were modeled assuming the draw solution was reconcentrated by NF/RO and taking into account different transport mechanisms for the FO membrane and different rejection rates by NF/RO. Modeling results showed that if the FO rejection rate is lower than the RO rejection rate (as is the case for most compounds tested), the added value of the FO-RO cycle compared to RO only at steady-state was small for diffusively and negative for convectively transported trace organics. Modeling also showed that trace organics accumulate in the draw solution.

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1. Introduction

Forward Osmosis (FO) has gained attention in recent years as a water treatment technology capable of handling heavily impaired water sources and capable of concentrating solutions with a high fouling potential (Cath et al., 2006; Zhao et al., 2012). The main benefits of FO in this regard are: the production of high quality permeate because of a high rejection of different pollutants and operation under osmotic driving force without the need for a hydraulic pressure difference (Zhao et al., 2012). FO has been used to reclaim water from (un)treated domestic waste water (Alturki et al., 2012; Cath et al., 2010; Cornelissen et al., 2008; Hancock et al., 2011; Zhang et al., 2011), anaerobic digestion concentrate (Holloway et al., 2007), and to concentrate liquid foods (Sant'Anna et al., 2012).

Impaired water sources, such as waste water treatment plant (WWTP) effluents, are often contaminated with trace organic compounds (TOrCs) (Daughton and Ternes, 1999; Ternes et al., 2005). TOrCs are anthropogenic organic compounds present in waste water at concentrations in the ng/L to μg/L range. This group of compounds consists of, among others, pharmaceuticals, personal care products, flame retardants and pesticides. One potential consequence of chronic exposure to TOrCs is endocrine disruption (Markey et al., 2002). Although some controversy remains whether endocrine disruption has significant effects on humans (Giwercman, 2011; Sharpe, 2003), the effects of endocrine disruption caused by estrogenic compounds in aquatic vertebrates has been reported (Sumpter and Johnson, 2005). Conventional waste water treatment systems, such as coagulation, trickling filters, sand filters and activated sludge remove TOrCs to varying degree, but often unsatisfactory when waste water is to be reclaimed. Svenson et al. (2003) found an increased removal rate of estrogenic compounds in treatment media with an increased bioactivity. Ternes (1998) found a TOrCs removal rate varying between 10 and almost 100% for pharmaceuticals in German WWTPs, which is the same conclusion reached by Van De Steene et al. (2010) who also found removal rates varying from 0 to almost 100%.

For the practical implementation of FO, maintaining a high rejection of TOrCs under different operating conditions is a key challenge, especially if FO is used to produce reclaimed water from WWTP in- or effluent. Alturki et al. (2012) noted that rejection of TOrCs by FO in an osmotic MBR (OMBR), of which the sludge was conditioned to the tested TOrCs, was consistently high for the solutes with a molecular weight above 266 g/mole, while the rejection of smaller solutes appeared to relate to their biodegradation susceptibility. This could indicate that the actual rejection rates of the smaller solutes were quite low. Hancock et al. (2011) have tested TOrC-rejection in both bench- and pilot-scale installations, using MBR-treated domestic waste water as a feed. The rejection of hydrophobic nonionic compounds by FO appeared to improve with increasing TSS concentration in the feed, which could indicate sorption of these compounds onto the TSS and subsequent filtration of the TSS. The rejection of mainly negatively charged organic solutes was consistently high, regardless of the TSS concentration.

Although previous studies have investigated TOrCs rejection by FO membranes, there is still relatively limited knowledge on the actual transport mechanisms of these solutes in FO, which contrasts the transport of TOrCs in pressure-driven processes such as NF and RO (Bowen et al., 1997; Kim et al., 2007; Ramon et al., 2012; Verliefde et al., 2009a). In addition, although the influence of fouling by several water matrices on rejection of TOrCs has been investigated in practice, most studies have not tried to identify underlying mechanisms of influence of fouling on rejection. In contrast to studies on NF/RO (Botton et al., 2012), the influence of different model foulants and of biofilm formation on rejection of TOrCs in FO has not been systematically investigated.

When FO is used to reclaim water from impaired sources, the effect of draw solution regeneration in a closed-loop system on the TOrCs concentration in the final product water has not been investigated yet. Hancock et al. reported a build-up of TOrCs in the draw solution (Hancock et al., 2011) when using RO to regenerate the FO draw solution (consisting of NaCl) in a closed-loop configuration. Cath et al. (2010) made a similar observation in a closed loop FO-RO configuration. Although both groups reported a total rejection of TOrCs by the combined FO-RO system in the order of 99%, the statement of FO-RO being a double barrier against micropollutants has not been thoroughly assessed in closed-loop systems. Both groups reported that the build-up of TOrCs in the draw solution was caused by a higher rejection of TOrCs by RO than by FO (Cath et al., 2010; Hancock et al., 2011). A TOrCs build up in the draw solution might negatively impact the TOrCs concentration in the final permeate. It is therefore imperative to investigate the fate of TOrCs when FO is used in a closed loop system.

In this study, different model foulants were used to foul FO membranes and effects on FO rejection of 20 pharmaceuticals was studied. In addition, long-term biofouling experiments were carried out, in which the biofouled membrane was extensively characterized and again the effect on the FO rejection of pharmaceuticals was investigated. The build-up of TOrCs in the draw solution is systematically studied and modeled in closed-loop FO-RO/NF applications, and the potential implications for potable water production are discussed.

2. Materials and methods

2.1. FO setup and filtration protocols

2.1.1. FO setup

The FO membranes used in this study, were commercial cellulose tri-acetate (CTA) membranes produced by Hydration Technology Innovations (HTI) (Albany, Oregon, USA). Membrane properties are shown in supplementary information. The membrane orientation in this study was in FO mode (active layer facing the feed solution). The membrane cell was a transparent polycarbonate cell, with a flow channel length of 250 mm, a width of 50 mm and a membrane surface area of 124 cm^2. The membrane cell was oriented horizontally, with the feed channel on top. Feed and draw solution were delivered to the membrane module in counter-current mode, both