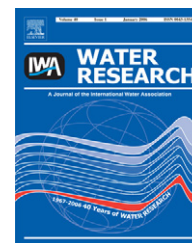


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Evaluating the impacts of membrane type, coating, fouling, chemical properties and water chemistry on reverse osmosis rejection of seven nitrosoalkylamines, including NDMA

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ABSTRACT

Reverse osmosis (RO) treatment has been found to be effective for a wide range of organics but generally small, polar, uncharged molecules such as *N*-nitrosodimethylamine (NDMA) can be poorly rejected. The rejection of seven *N*-nitrosoalkylamines with molecular masses in the range of 78–158 Da, including NDMA, *N*-nitrosodiethylamine (NDEA), *N*-nitrosomethylethylamine (NMEA), *N*-nitrosodipropylamine (NDPA), *N*-nitrosodibutylamine (NDBA), *N*-nitrosopyrrolidine (NPyr), *N*-nitrosopiperidine (NPip) by three commercial brackish-water reverse osmosis membranes was studied in flat-sheet cells under cross-flow conditions. The membranes used were ESPA3 (Hydranautics), LFC3 (Hydranautics) and BW-30 (Dow/Filmtec), commonly used in water reuse applications. The effects of varying ionic strength and pH, dip-coating membranes with PEBAX 1657, a hydrophilic polymer, and artificial fouling with alginate on nitrosamine rejection were quantified. Rejection in deionized (DI) water increased with molecular mass from 56 to 70% for NDMA, to 80–91% for NMEA, 89–97% for NPyr, 92–98% for NDEA, and to beyond the detection limits for NPip, NDPA and NDBA. For the nitrosamines with quantifiable transmission, linear correlations ($r^2 > 0.97$) were found between the number of methyl groups and the log(transmission), with factor 0.35 to 0.55 decreases in transmission per added methyl group. A PEBAX coating lowered the ESPA3 rejection of NDMA by 11% but increased the LFC3 and BW30 rejection by 6% and 15%, respectively. Artificially fouling ESPA3 membrane coupons with 170 g/m² alginate decreased the rejection of NDMA by 18%. A feed concentration of 100 mM NaCl decreased rejection of NDMA by 15% and acidifying the DI water feed to pH = 3 decreased the rejection by 5%, whereas increasing the pH to 10 did not have a significant ($p < 0.05$) effect.

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1. Introduction

In a world where high-quality water resources are becoming increasingly scarce, developing safe new measures for keeping up with soaring water demand is of paramount impor-

tance. One option is to recycle pretreated wastewater by treating it to an acceptable standard using reverse osmosis (RO). While RO removes salts and a broad range of dissolved organics to a very high degree (Reinhard et al., 1986; Schutte, 2003; Bellona et al., 2004), it is known that some harmful trace

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contaminants, such as disinfection byproducts like trihalo-methanes, haloacetic acids and N-nitrosodimethylamine (NDMA) can pass through RO membranes (Bellona et al., 2004). However, the exact removal efficiency of the membrane process with respect to many of these compounds is still unknown, particularly for conditions which one would likely encounter in practice.

In the present study, the removal of a suite of seven N-nitrosamines by three different reverse osmosis membranes under different conditions is investigated. Of this group of probable human carcinogens (U.S. EPA, 2007a), NDMA is known most prominently for its formation during the disinfection of secondary-treated wastewater effluent with chloramines (Mitch et al., 2003; Mitch and Sedlak, 2004). Recently, other nitrosamines have also been found in the effluent from treatment plants (Zhao et al., 2006; Schreiber and Mitch, 2006). Six of the seven nitrosamines studied here are on the list of 25 contaminants to be monitored under the US EPA's Unregulated Contaminant Monitoring Rule 2, taking effect in 2008 (U.S. EPA, 2007b). Monitoring of advanced treatment facilities has shown that NDMA passes through RO membranes at relatively high rates (Mitch et al., 2003; Rodriguez et al., 2004; Plumlee et al., 2007). However, the relationships between compound properties and rejection by RO membranes and the influence of membrane characteristics and water quality factors on nitrosamine rejection are still poorly understood.

The separation process for RO is most often explained via the solution-diffusion process model (Wijmans and Baker, 1995), which states that both water and solute passing through the membrane must first dissolve into the membrane matrix, then diffuse through it to reach the permeate side (Lonsdale et al., 1965). The “rejection” of contaminants is thus based on a number of processes, as outlined in Fig. 1. Many compounds, such as those carrying charge, will be repelled by the membrane surface and be immediately rejected (Kimura et al., 2003). Uncharged solutes can approach the membrane surface, where they presumably follow the process outlined in the solution-diffusion model. The extent of compound rejection is therefore controlled by the relative rates of dissolution into and diffusion within the membrane matrix between water and solute. Additionally, surface adsorption and internal absorption have been shown to play a role in the initial rejection of some compounds, where the rejection decreases over time until all sorption sites are filled (Ng and Elimelech, 2004; Nghiem et al., 2004). Many factors may influence these processes, amongst them charge, size, hydrophobicity, hydrogen bonding capacity and dipole moment (Bellona et al., 2004; Van der Bruggen et al., 1999; Nghiem et al., 2004; Kim et al., 2005).

In general, a coating layer on a membrane (produced intentionally, or as the result of foulant accumulation) can act in several ways with respect to rejection: In the simplest case, it acts as an additional barrier to the solutes of interest without retarding the solvent, thus increasing rejection. But even if the bulk material of the coating layer does not in itself significantly affect the transmission of either solute or water, it can also affect the overall membrane performance. For instance, an additional layer (coating or foulant) on the surface can prevent mixing, thus exacerbating the effects of

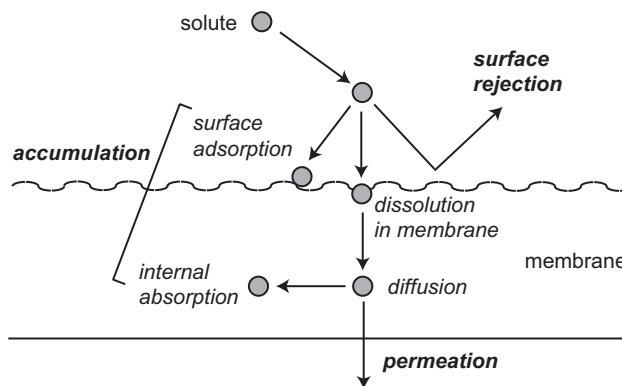


Fig. 1 – Schematic of processes controlling the permeation of small organics through an RO membrane via solution-diffusion.

concentration polarization and leading to decreased rejection (Lee et al., 2004; Ng and Elimelech, 2004). Indeed, a layer of foulant can have a significant impact on the rejection of trace contaminants (Schäfer et al., 2000; Xu et al., 2006). Yet other effects have been postulated, specifically for the PEBAX coating used in this study (Louie et al., 2006): interaction between the membrane surface and the coating, or some reaction during the coating process might create a third “interfacial” layer between “bulk” membrane material and bulk coating material that has properties different from either bulk phase. This could lead to increase or decrease in the rejection of solutes depending on the interfacial layer’s transmissivity toward those solutes relative to that of water.

The objective of this work was to quantify the rejection of seven N-nitrosamines, including NDMA, by three different RO membranes, in the interest of improving membrane selection and conditions for water reuse applications where these compounds may be of concern. To this end, the effects of membrane coating, surface fouling with alginate and feed solution chemistry (ionic strength and pH) on the rejection of these compounds were investigated.

2. Materials and methods

2.1. Chemicals

Unless otherwise specified, all chemicals used were of analytical grade. Sodium chloride and calcium chloride were obtained from Mallinckrodt-Baker (Phillipsburg, NJ, USA), sodium hydroxide (1N) and hydrochloric acid (1N) from Fisher Scientific (Santa Clara, CA, USA), phenol from Alfa Caesar (Ward Hill, MA, USA) and sodium hypochlorite (6%) from VWR Scientific (Pittsburgh, PA, USA). Sodium alginate from brown algae was obtained from Sigma-Aldrich (St. Louis, MO, USA) and PEBAX 1657 from Arkema Inc. (Philadelphia, PA, USA). All but one of the nitrosamine compounds were purchased from Supelco (Bellefonte, PA, USA): N-nitrosodimethylamine (NDMA), N-nitrosodiethylamine (NDEA), N-nitrosodipropylamine (NDPA), N-nitrosobutylamine (NDBA), N-nitrosopyrrolidine (NPyr), N-nitrosopiperidine (NPip). The seventh nitrosamine, N-nitrosomethylethylamine (NMEA), was purchased from ULTRA

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