

Performance of nanofiltration membrane in rejecting trace organic compounds: Experiment and model prediction



Xiao-mao Wang^{a,c,*}, Bing Li^b, Tong Zhang^c, Xiao-yan Li^c

^a State Key Joint Laboratory of Environmental Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing 100084, China

^b Graduate School at Shenzhen, Tsinghua University, Guangdong 518055, China

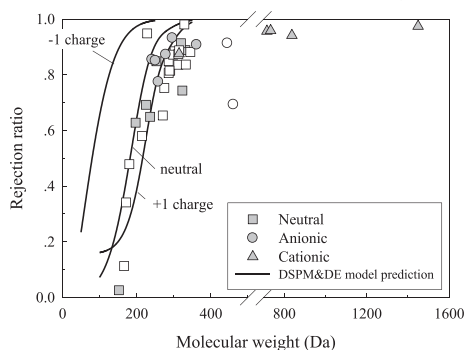
^c Department of Civil Engineering, The University of Hong Kong, Pokfulam Road, Hong Kong, China

HIGHLIGHTS

- Real rejection ratios for TOrcs by Desal HL were determined.
- The NF membrane rejected most TOrcs effectively except for a few.
- Steric effect was the primary mechanism for NF rejection of TOrcs.
- DSPM&DE model generally over-predicted the rejection ratios.

GRAPHICAL ABSTRACT

All the studied 40 TOrcs other than acyclovir, caffeine, carbamazepine, chloramphenicol, metronidazole, nandrolone, oxytetracycline, sodium nifurstyrenate and trenbolone were effectively rejected by the Desal HL membrane. The DSPM&DE model over-predicted the rejection ratios for most TOrcs, except for acyclovir, caffeine and ranitidine which have low molecular weight and hydrophobicity.



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ABSTRACT

This study aimed to investigate the rejection ratios for 40 trace organic compounds (TOrcs) by the Desal HL nanofiltration (NF) membrane and to assess the applicability of the DSPM&DE model in predicting the rejection performance. Steady state rejection ratios were considered. A higher water flux normally led to a higher rejection ratio. At a filtration flux of 6.59×10^{-6} m/s, the NF membrane could effectively reject most of the TOrcs (>80%), except for a few that included acyclovir, caffeine, carbamazepine, chloramphenicol, metronidazole, nandrolone, oxytetracycline, sodium nifurstyrenate and trenbolone. Most poorly rejected TOrcs had a molecular weight lower than 275 Da. Steric hindrance effect was the primary mechanism that contributed to the rejection of TOrcs by the NF membrane. The rejection ratio for methylparaben decreased when the water flux increased. The DSPM&DE model was successful in predicting the rejection ratios for acyclovir, caffeine and ranitidine, which all have relatively low molecular weight and are hydrophilic. The model generally over-predicted the rejection ratios for the remaining TOrcs. The over-prediction could not be explained by high hydrophobicity only. Lack of consideration of the TOrc-membrane interactions on the partitioning of TOrcs to the membrane material was probably the primary reason for over-prediction by the model.

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* Corresponding author at: State Key Joint Laboratory of Environmental Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing 100084, China.
E-mail address: wangxiaomao@tsinghua.edu.cn (X. Wang).

1. Introduction

One of the many emerging challenges facing drinking water treatment is the occurrence of a large number of trace organic compounds (TOrcs) in water sources. For example, the median concentrations of detectable TOrcs in streams in the United States are generally in the ng/L to µg/L range [1]. These emerging contaminants may include pharmaceuticals and personal care products (PPCPs), hormones, products of oil use and combustion, other extensively used chemicals, and disinfection by-products. Many of the TOrcs have endocrine disrupting activity and/or are potentially carcinogenic and mutagenic. Existing conventional drinking water treatment plants were not designed for the removal of these TOrcs. Previous studies have revealed that conventional drinking water treatment processes have low removal of most TOrcs [2–4]. The removal efficiency can be enhanced by activated carbon adsorption for less polar compounds, by ozonation for some polar and aromatic compounds, and by some other recently developed processes. Nevertheless, some TOrcs could still remain in the water after advanced treatment [2]. Although adverse health effects cannot be attributed with certainty to low levels of TOrcs, drinking water should be relatively free of these compounds.

Nanofiltration (NF) is a promising technology for the removal of TOrcs since the majority of the TOrcs have a molecular weight within 150–500 Da [5] and the molecular weight cut-off (MWCO) for most commercial NF membranes ranges from about 200 to 2000 Da. TOrcs are expected to be largely removed by physical sieving if their molecular weights are larger than the membrane MWCO. TOrcs of small molecular weight would also be partly removed by the steric hindrance effect and, if both the TOrc and membrane surface were negatively charged, by the electrostatic effect [6]. Compared with reverse osmosis, NF can be operated at lower applied pressure (demanding less energy) and rejects fewer mineral ions (which means that the permeate needs less post-treatment).

The performance of NF in removing TOrcs has been extensively investigated in laboratory and onsite studies using both simulated and natural water [7–16]. Many brands of NF membranes have been tested for a large number of TOrcs. The general conclusion has been that NF is not always effective at removing all TOrcs. For example, by using an NF membrane with a nominal MWCO of 200 Da, the rejection ratios for 52 endocrine disrupting compounds and PPCPs in modeled and real surface water were found to vary from 0% to over 90% [9]. Radjenović et al. [13] found that the NF membrane with a nominal MWCO of 200 Da installed in a full-scale drinking water treatment plant had excellent rejection of 9 out of 12 pharmaceuticals but poor rejection of acetaminophen, gemfibrozil and mefenamic acid, of which only about 50% was rejected. Generally, poor rejection was associated with NF membranes with relatively large MWCO, and uncharged TOrcs with low molecular weight. An apparent time-dependency of the rejection ratio by NF was also observed for some uncharged TOrcs [17,18], which strongly suggests membrane adsorption affecting the rejection performance. Less polar (indicated by low dipole moment) and more hydrophobic (indicated by high octanol–water partition coefficient) compounds can usually be preferentially adsorbed [12]. Adsorption usually impairs the rejection performance after the NF membrane adsorption capacity is exhausted.

A useful rejection diagram was proposed by Bellona et al. [6], which can be used for rough estimations of the rejection ratios. For better design of the NF system and optimization of the operational conditions, it is highly desirable if the ability of given NF membranes to reject TOrcs of interest can be accurately predicted. Among the many mathematical models available so far, the DSPM&DE (Donnan Steric Pore Model & Dielectric Effect) developed by Bowen and Welfoot [19] and refined by Bandini and Vezzani [20] has a promising potential. The DSPM&DE model is relatively simple to use and involves only three adjustable parameters, which are average pore size, effective thickness and surface charge density. The model was found successful in predicting the ability

of NF to reject inorganic salts. The model was also applied to predict the NF performance in rejecting organic compounds such as hormones [21] and a selection of PPCPs [22]. However, results showed that the model generally over-predicted the rejection ratios. The lower-than-expected rejection ratio was attributed to the under-estimated partitioning of the organic compounds to the membrane (i.e. the solid phase). Many hydrophobic organic compounds had strong affinity to the membrane due to hydrophobic interactions. (In this context, affinity exclusively refers to the interaction caused by hydrophobic effect and does not include other specific interactions such as hydrogen bonding.) A later refinement of the DSPM&DE model took into account the affinity effect, and the prediction accuracy was greatly improved [23]. The applicability of the DSPM&DE model to predict the rejection of TOrcs (present at trace levels) by NF is as yet unknown. In this case, the TOrc concentrations are far less than that of the background electrolytes. It was hypothesized that if a TOrc were hydrophilic and had no specific interaction with the membrane material, the DSPM&DE model would be able to predict the rejection ratio well.

In this study, the removal of a total of 40 hormones and PPCPs in a lab-scale NF system was investigated under different applied filtration pressures. Bearing the time-dependency of rejection in mind, we considered the “real” rejection performance by taking into account the TOrc concentration decrease in the feed water. To apply the DSPM&DE model for the prediction of the rejection ratios, it was assumed that the NF membrane surface charge was dictated by the background electrolytes but not by the TOrcs. The applicability of the DSPM&DE model was assessed. The TOrcs tested had very diverse physicochemical properties, and the assessment would be valuable for the future improvement of the DSPM&DE model.

2. Materials and methods

2.1. Nanofiltration setup and operation

The lab-scale NF setup consisted of three parallel cross-flow filtration cells, a feed tank (20 L effective volume), a high-pressure diaphragm pump (Hydro-Cell, Wanner Engineering, USA), a number of pressure sensors (Type A-10, WIKA, Germany) and flow-rate sensors (CH-2563, Digmesa, Switzerland), and other accessories. The filtration cells were made of Acetal, while the feed tank, the tubing and the valves were all made of stainless steel. The effective filtration area and filtration channel height of each filtration cell were $120 \times 50 \text{ mm}^2$ and 2 mm, respectively. During the filtration, all permeates from filtration cells were returned to the feed tank, except when sampled for chemical analysis. A commercial NF membrane (Desal HL, GE, USA) was used in this study. It has a nominal MWCO of 150–300 [24].

The feed water was different depending on the purpose. De-ionized water was filtered to determine the permeability (A) of the NF membrane, i.e. the dependence of water flux (J_v) on applied filtration pressure (ΔP), according to

$$A = J_v / \Delta P. \quad (1)$$

Feed water containing either 10 mg/L glucose or 10 mmol/L NaCl was filtered to obtain the raw data for the determination of the NF membrane characteristic parameters (see Appendix A). A mixed solution of TOrcs was filtered for the determination of the respective rejection ratio for each TOrc. All 40 TOrcs were dissolved in a 10 mmol/L NaCl solution to achieve a concentration of 20 µg/L for each. The NaCl acted as the background electrolyte in this solution. The pH of the TOrc-containing feed was measured at about 6.5, which was slightly lower than the pH used during evaluation of the TOrc charge (shown in Table 1). It was assumed that this small pH deviation would not affect the TOrc charge estimation. During each filtration test, at least two parallel filtration cells were used as replicates. One fresh membrane coupon was used for each cell and each filtration. Both the

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