



The effects of feed solution temperature on pore size and trace organic contaminant rejection by the nanofiltration membrane NF270



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ABSTRACT

This study investigated the effect of feed temperature on membrane pore size and the rejection of trace organic contaminants (TrOCs) by the nanofiltration (NF) membrane NF270. Filtration experiments were conducted using a cross flow membrane system at 20, 30 and 40 °C. The membrane pore radius was estimated using the pore hindrance transport model at each temperature and the rejection data of three reference organic solutes (i.e. erythritol, xylose and glucose) experimentally obtained in this study. The results suggest that the pore size of an NF membrane is dependent on the feed solution temperature. An increase in the feed temperature from 20 to 40 °C led to an increase in the effective pore radius from 0.39 to 0.44 nm. Consequently, the increase in the feed temperature also caused a considerable drop in the rejection of all TrOCs investigated in this study. The decrease in rejection observed here could be attributed to not only the increase in the solute diffusivity but also the enlargement of the membrane pore size. As the feed temperature increased, the decrease in rejection of neutral TrOCs was more severe than that of negatively charged compounds. This is because in addition to size exclusion (or steric hindrance) the rejection of negatively charged TrOCs is also governed by electrostatic interaction given that the membrane surface is also negatively charged.

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1. Introduction

Over the last few decades, there has been much research on the rejection of trace organic contaminants (TrOCs) such as endocrine disrupting chemicals, pharmaceuticals and pesticides by nanofiltration (NF) membranes [1–7]. However, despite the fact that temperature is an important factor governing mass transfer in membrane separation processes, most previous studies were performed at an arbitrary temperature and the impact on temperature on TrOC rejection has not been systematically investigated. However, previous work has demonstrated that the temperature has a significant impact on NF membrane performance. According to Goosen et al. [8], polymeric membrane is sensitive to changes in the feed temperature. They reported an increase of up to 60% in the permeate flux when the feed temperature was increased from 20 to 40 °C. A linear relation between temperature and water flux by NF performances has been reported in the temperature range from 10 to 30 °C [9] and 20 to 70 °C [10]. It was explained that the flux increases with temperature may be attributed to the thermal expansion of the membrane material [9,10]. In a study on the

effect of temperature on the permeation characteristics of NF membranes, Sharma et al. [11] suggested that with increasing temperature, the average pore size increased and the pore density decreased because of the thermal expansion of the polymer constituting the active layer of thin-film composite membranes. These could be the cause of the reduction in rejection of organic solutes by NF membranes with increasing temperature. Therefore, a comprehensive understanding of the rejection mechanisms and mathematical models of TrOCs by NF processes under different temperature conditions is very important for the prediction of solute rejection, optimisation of treatment processes, and development and application of new methodologies.

The membrane pore size and molecular dimensions of organic solutes are important factors which can affect their rejection during NF filtration [3,12]. Several mathematical models, applied so far to estimate the membrane pore size, have been based on the uncharged solute transport across the membranes under different permeate flux conditions. Wang et al. [13] applied the steric-hindrance pore (SHP) model to estimate the pore radius of NF membrane (G-20) from the permeation experiments of different uncharged solutes (such as glucose, saccharose, raffinose and α -cyclodextrin) at 25 °C and demonstrated that the rejection was influenced strongly by molecular weight, Stokes' radius and

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diffusion coefficient of these solutes. In another study, Bowen and Welfoot [14] estimated the NF membrane pore radius (Desal-DK) based on the Donnan-steric pore model (DSPM) using two uncharged solutes including glycerol and glucose at 25 °C. Their findings showed a difference in the pore radius for both tested solutes, owing to their different physicochemical properties (such as Stokes' radius and diffusion coefficient). López-Muñoz et al. [15] successfully applied the SHP model in order to calculate the pore radius of NF membranes (NF270 and NF90) using three uncharged solutes (ribose, glucose and sucrose) at the same temperature. They also elucidated the relationship between the membrane pore size characterisations and rejection efficiency of phenolic TrOCs and concluded that rejection of these compounds by the NF90 membrane was higher than for the NF270 membrane, because of its smaller pore size [15]. Despite a large volume of research on the effects of the membrane pore size on the rejection of trace organic solutes by NF membranes, to date, there have been no studies on the effect of temperature on membrane pore size and rejection of TrOCs by NF membranes. Additionally, the combined influence of the membrane pore size and diffusion of TrOCs across the membrane on their rejection efficiency is still poorly understood.

The objectives of the current work were to study and assess the effects of the feed temperature on pore radius and TrOC rejection by the NF process. The pore-hindrance transport model was used to determine the average pore radius of the membrane at 20, 30, and 40 °C using reference neutral organic solutes. Subsequently, the influence of the membrane pore size and diffusion behaviour of the target TrOCs on their rejection at different temperatures were examined and discussed. The results enable the evaluation of the role of feed temperature on TrOC rejection by the NF process.

2. Experimental

2.1. Nanofiltration membrane

The NF270 membrane (Dow-Filmtec, Minneapolis, MN) was selected for this study. According to the manufacturer, it is a thin-film composite polyamide membrane that is widely used for water and wastewater treatment application. This is a loose NF membrane with a relatively high permeability (of approximately 11 L/bar m² h). At pH 4 and above, this membrane is negatively charged [16]. The flat sheet membrane samples were stored dry before use.

2.2. Theoretical background and calculation method

In order to determine the pore size of the membrane and the effect of temperature, three uncharged reference solutes were used. In the pore-hindrance model, transport of uncharged solutes through a membrane are governed by two important mechanisms of solute transport through the NF membrane, namely diffusion, as a result of concentration gradients and convection caused by the pressure difference across the membrane [13,17]. Their transport equation is thus given as the sum of the diffusive and the convective terms as follows [12,18]:

$$J_s = K_c C_j v - D_p \frac{dC}{dx} \quad (1)$$

$$J_s = J_v C_p \quad (2)$$

where J_s and J_v are the solute and solution fluxes, respectively, K_c is the convective hindrance coefficient, C is the solute concentration in the membrane pore, D_p is the hindered solute diffusivity in the membrane pores, $D_p = K_d D$ where D is the bulk diffusivity and K_d is the diffusive hindrance coefficient, x is the axial position within

the membrane pore and C_p is the solute concentration of the bulk permeate.

The hindrance factors (K_c and K_d), are functions of the ratio of the solute radius to the pore radius (λ):

$$\lambda = \frac{r_s}{r_p} \quad (3)$$

In the range of $0 < \lambda < 0.8$, these factors can be calculated by the following equations [12,18]:

$$K_d = 1 - 2.3\lambda + 1.154\lambda^2 + 0.224\lambda^3 \quad (4)$$

$$K_c = (2 - \Phi)(1 + 0.054\lambda - 0.988\lambda^2 + 0.441\lambda^3) \quad (5)$$

The steric partition factor (Φ) is defined as the ratio of the solute-accessible area to the pore area and is expressed as follows:

$$\Phi = (1 - \lambda)^2 \quad (6)$$

Because the diffusion coefficient (D) is associated with solute radius and the mass transfer coefficient, it is an important parameter that influences solute transport across the membrane. Diffusion coefficient of TrOCs in water can be calculated by Wilke and Chang equation [19]. This equation can give satisfactory predictions for the diffusivity of trace organic solutes in water.

$$D = \frac{1.173 \times 10^{-13} (\phi M)^{0.5} T}{\mu V_m^{0.6}} \quad (7)$$

where ϕ is an association factor for the solvent ($\phi = 2.6$ for water), M is molecular mass of solvent ($M = 18$ g/mole for water), μ is the viscosity of solvent, T is the absolute temperature and V_m is molar volume of the solute at its boiling point. V_m can be estimated from the group contributions of structural contributions to molar volumes [19].

The solute radius (r_s) used in Eq. (3) is the effective solute radius in the pore, commonly estimated by Stokes–Einstein equation [3,20].

$$r_s = \frac{kT}{6\pi\mu D} \quad (8)$$

where k is Boltzman's constant, T is the absolute temperature, and μ is the viscosity of the solvent which is water in this case. As can be seen from Eqs. (7) and (8), the solute size is independent of the solution temperature. Thus, at a given permeate flux, changes in the solution temperature are related only to the membrane pore size and the diffusion coefficient of the solute.

The previous assumption also enables the introduction of the Hagen–Poiseuille equation definition of pore solvent velocity in the Peclet number definition (Pe), thus it can be considered a model parameter for the rejection of the solutes by the membrane [21]:

$$Pe = \frac{K_c J_v \Delta x}{D_p} \quad (9)$$

where Δx is the length of the pore.

The concentrations of the solute at the inlet and outlet sides of the pore (C_i and C_o , respectively) are expressed in terms of the steric partition factor and the permeate and membrane surface solute concentrations (C_p and C_m) as follows [18,22]:

$$C_i = \Phi C_m \quad (10)$$

$$C_o = \Phi C_p \quad (11)$$

In order to obtain an expression for rejection of the solute, Eq. (1) is integrated across the membrane with the solute concentrations in the membrane at the upper ($x = 0$) and lower ($x = \Delta x$) surfaces expressed in terms of C_m and C_p using the equilibrium partition coefficient, Φ . $C_{x=0} = \Phi C_m$ and $C_{x=\Delta x} = \Phi C_p$. In terms of real

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