



# Mass transfer of differently sized organic solutes at spacer covered and permeable nanofiltration wall

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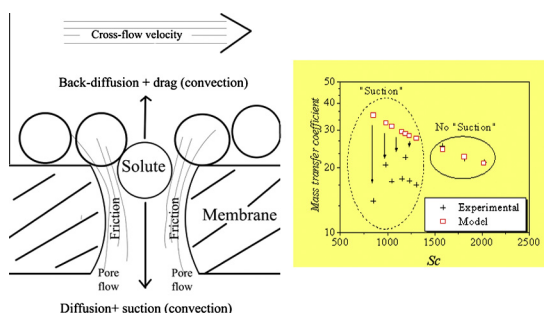
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## HIGHLIGHTS

- Cross-flow velocity significantly affects removal of organics.
- Sherwood relation correctly estimates mass transfer at small solute fluxes.
- At high solute fluxes Schmidt number needs correction.
- Correction correlation is proposed for Schmidt number.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Concentration polarization (CP) phenomena may significantly affect water permeability and removal of organics solutes in cross-flow nanofiltration (NF) making it an important optimization parameter. Most of the models predict CP using mass transfer coefficients that may be estimated using model Sherwood ( $Sh$ ) relations of a general form,  $Sh = a^* Re^b Sc^c$ . In many cases  $Sh$  relations are able to predict mass transfer coefficients remarkably well; however, such relations are in general valid only for non-permeable walls.  $Sh$  relations were experimentally validated using a binary solution of single solute and water where Schmidt numbers ( $Sc$ ) were varied by changing the temperature or density of the solution, at a constant or varied Reynolds number ( $Re$ ). This study evaluated  $Sh$  relations from different angle and used ten organic solutes of different diffusivity at a constant concentration and viscosity of solutions, covering a range of  $Sc$  from 850 to 2022. The aim of this study was to evaluate the  $Sh$  relation in predicting the mass transfer of differently sized organic solutes in rectangular channel, at spacer covered and permeable NF wall of defined porosity. Comparison of experimental  $Sh$ , obtained using the velocity variation method, and model  $Sh$  showed that model  $Sh$  relation correctly predicts mass transfer of organics when particular solute flux through the NF permeable wall is sufficiently low. A correction correlation is proposed for coefficient  $c$  on  $Sc$  in model  $Sh$  relation, where  $c$  approaches the model  $Sh$  value, 0.42, with the increase in size of the solutes. In addition, data presented show that the removal of organic solutes from the water may be significantly improved, up to 280%, by changing the hydrodynamics in the channel.

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## 1. Introduction

Nanofiltration (NF) membranes have been widely applied to drinking water treatment [1], industrial effluent treatment [2],

water softening [3], or concentration of grass juice [4]. NF processes usually remove most of the solutes from water, thus solutes tend to accumulate on the membrane surface developing a concentration polarization (CP) layer. Given that NF processes are pressure and concentration driven processes [5], enhanced concentration at the membrane surface may significantly [6] affect the NF

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performance in terms of flux reduction and removal of solutes [7]. In this context, CP could be considered as one of the important parameters in NF process optimization. Many studies dealt with CP phenomena [8,9] and its effect on removal of salts or mixture of salts [10], but very few of them systematically examined CP effect on removal of organics [7], particularly in nanofiltration. NF processes are also of increasing use in removal of organic solutes [1,11] and it is important to correctly evaluate the impact of hydrodynamic conditions on their removal as well as to properly address this phenomena. Most models which describe CP in cross-flow nanofiltration require a knowledge of mass transfer coefficients [12].

Mass transfer coefficients may be determined experimentally, e.g., using the velocity variation method (VVM) [13] or electrochemical methods [14]. VVM has been known for decades as a simple and efficient method applicable to solutes that are not completely rejected by the membrane [8]. VVM is, however, often criticized because the boundary layer in a RO/NF channel is undeveloped and of a non-uniform thickness. Assuming its uniformity can produce significant errors because local concentration at the membrane wall varies throughout the channel [6,15]. A feed spacer may be used as a promoter of turbulence and local mixing where each mesh on the spacer periodically interrupts the development of boundary layer; in these cases the thickness of the boundary layer could be considered uniform and static. Indeed, a recent numerical simulation of mass transfer in a spacer filled channel [14] has shown that for  $Re > 159$  and  $Sc > 100$ , the boundary layer is becoming closer to uniformity.

Mass transfer coefficients may be determined as well using model  $Sh$  relations, which are valid for channels with non-permeable walls [8,16]. NF membranes are economically viable because they offer high water fluxes. At higher water fluxes, however, a significant “suction” occurs and NF membranes cannot be considered as non-permeable walls. “Suction” may significantly affect the mass transfer coefficient and thus enhance concentration polarization [17] where membrane porosity and pore sizes may play a substantial role. Previous studies showed that membranes, which can be considered as permeable walls at significantly high fluxes, may vary in terms of thickness and swelling [18] as well as heterogeneity [19]. Košutić et al. reported that reverse osmosis membranes have denser and less porous structures compared to NF membranes [20]. Both pore size distributions and number of pores (active porosity) of membranes cannot be simply evaluated since the radii of the pores of NF membranes are below 1 nm [21]. Therefore, an indirect method was used in this paper, similar as reported earlier [22], to evaluate pore size distribution and the number of pores, i.e., active porosity of the membrane representing the permeable wall.

In this context, this study aims at checking the validity of model  $Sh$  relation in predicting the mass transfer coefficients in rectangular channels with one highly permeable and porous wall. Experimental average mass transfer coefficients of organic solutes, which differ in molecular mass and diffusivity, were estimated using VVM and afterwards compared with model  $Sh$  relations, which well described hydrodynamic conditions in this work [14]. The organic solutes were chosen to cover a range of  $Sc$  which are of particular interest for NF, ranging from 850 to 2022. Highly permeable wall considered herein was commercial high flux NF membrane, NF270, while mass transfer coefficients were studied at a permeation velocity of  $40 \mu\text{m s}^{-1}$ , when significant “suction” occurs [23].

## 2. Theoretical background

### 2.1. Sherwood relation

The  $Sh$  relation that fully describes hydrodynamic conditions in the system [24] is described using general expression below.

$$Sh = \frac{kd_H}{D} = aRe^bSc^c \left(\frac{d_H}{L}\right)^d \quad (1)$$

Eq. (1) is used to estimate mass transfer coefficients while the empirical coefficients  $a$ ,  $b$ ,  $c$  and  $d$  may be found in literature and depend on hydrodynamic conditions and feed spacer characteristics [8,14,16]. The  $Re$  in Eq. (1) is calculated as  $Re = (u d_H)/\nu$ , and  $Sc$  as  $Sc = \nu/D$ , where  $u$  is cross-flow velocity in the channel,  $d_H$  hydraulic diameter,  $D$  is diffusivity, and  $\nu$  is kinematic viscosity. The height and the length of the channel are constant and Eq. (1) may be simplified taking  $a^* = a (d_H/L)^d$ :

$$Sh = a^* Re^b Sc^c \quad (2)$$

Hydraulic diameter,  $d_H$ , for a rectangular channel with the spacer in the channel was calculated as proposed by Shock and Miquel [25].

$$d_H = \frac{4P}{2/h + (1-P)(S_{SP}/V_{SP})} \quad (3)$$

In Eq. (3)  $P$  is the porosity of the spacer,  $P = 1 - V_{SP}/V_{TOT(SP)}$ ,  $S_{SP}$  is the surface of the spacer,  $V_{SP}$  is the volume of the spacer, and  $V_{TOT(SP)}$  total volume of the spacer. Average velocity of fluid can be calculated as proposed below.

$$u[\text{m s}^{-1}] = \frac{Q[\text{m}^3 \text{s}^{-1}]}{P_{FC}A[\text{m}^2]} \quad (4)$$

where  $Q$  is feed flow rate and  $A$  is cross-sectional area of the channel and  $P_{FC}$  porosity of the channel expressed as  $P_{FC} = V_{SP}/V_{FC}$ , where  $V_{FC}$  is the volume of the channel.

### 2.2. Mass transfer coefficients – VVM and model $Sh$ relation

VVM [13] is used in this study to assess experimental mass transfer coefficients.

$$\ln\left(\frac{1-f}{f}\right) = \ln\left(\frac{1-f_m}{f_m}\right) + \frac{J_v}{k} \quad (5)$$

In Eq. (5), mass transfer coefficient,  $k$ , can be approximated [6] as  $k \approx K \cdot u^B$  where  $u$  is average cross-flow fluid velocity. Eq. (5) is used in a simple experimental procedure where  $u$  is varied at constant  $J_v$  and constant temperature while rejection,  $f$ , and cross-flow velocity,  $u$ , are measured. A fit to the experimental data gives the constant  $K$  which is used to approximate average mass transfer coefficient in the channel as well as the true membrane rejection,  $f_m$ . The value of coefficient  $B$  has to be set before fitting and this study takes  $B$  as proposed below. By combining Eqs. (1) and (2), the mass transfer coefficient of one solute can be calculated as presented below.

$$k = a^* \frac{D}{d_H} Re^b Sc^c \quad (6)$$

In Eq. (6),  $Re$  is the only variable while other parameters are constant because only one solute is considered in a diluted feed solution at a constant temperature and in constant geometry of the channel. Eq. (6) can be rewritten as proposed below, where terms in elongated brackets represent the constant.

$$k = \left\{ a^* \frac{D}{d_H} Sc^c \left(\frac{d_H}{\nu}\right)^b \right\} u^b \quad (7)$$

The analogy may be noticed between  $k = K u^B$  in Eqs. (5) and (7). Parameter  $B$  in Eq. (5) can be taken as coefficient  $b$  on  $Re$  number in  $Sh$  relation, which is usually available in literature. The parameters  $b$ ,  $c$  and  $B$ , which correspond to geometry of the feed spacer used ( $l_M/d_f = 12$  and  $\beta = 90^\circ$ ), and hydrodynamic conditions in the channel are taken from the work of Koutsou et al. [14]. Theoretical mass transfer coefficient,  $k$ , in our system can then be calculated as presented below.

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