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Performance of spiral-wound membrane modules in organic solvent nanofiltration – Fluid dynamics and mass transfer characteristics

Binchu Shi [a,b], Patrizia Marchetti [b], Dimitar Peshev [c], Shengfu Zhang [a], Andrew G. Livingston [a,b,*]

[a] Evonik Membrane Extraction Technology Limited, Unit 6 Greenford Park, Ockham Drive, Greenford, London UB6 0AZ, United Kingdom
[b] Department of Chemical Engineering, Imperial College London, Exhibition Road, London SW7 2AZ, United Kingdom
[c] Department of Chemical Engineering, University of Chemical Technology and Metallurgy, 8, Kl. Ohridsky Blvd, Sofia 1756, Bulgaria

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ABSTRACT

During the past few decades organic solvent nanofiltration has received a great deal of attention and a growing number of studies has been reported on development and optimisation of solvent resistant membranes and their transport mechanism. However, most of these studies have used flat sheet membranes. On the other hand, many researchers studied fluid dynamics and mass transfer in spiral-wound membrane modules, almost exclusively in aqueous solutions. This paper reports the performance of four spiral-wound membrane modules tested in 0–20 wt% solutions of sucrose octaacetate in ethyl acetate under various pressures and retentate flowrates. These modules were made of two different types of membranes (a commercial membrane, PuraMem® S600, and a development product, Lab-1, from Evonik Membrane Extraction Technology Limited) and covered three module sizes (1.8″ × 12″, 2.5″ × 40″ and 4.0″ × 40″). All modules had the same feed and permeate spacers. The classical solution diffusion model was applied to describe the transport of solute and solvent through the membrane and regress the unknown model parameters from flat sheet data. Correlations for characterising the fluid dynamics and mass transfer in the spiral-wound membrane modules, as well as the parameters describing the feed and permeate channels, were determined by performing the regression of experimental data of a 1.8″ × 12″ PuraMem® S600 membrane module. The classical solution–diffusion model, combined with the film theory, was then successfully applied to predict the performance of other modules of larger size (such as the 2.5″ × 40″ and 4.0″ × 40″ module sizes) and/or made of a different membrane material (such as Lab-1). The procedure proposed in this paper predicts the performance of a specific module by obtaining a limited number of experimental data for flat sheets and a 1.8″ × 12″ spiral-wound membrane module only (necessary to obtain the fitting parameters characteristic of the membrane and the module). Furthermore, with this procedure, it is not necessary to know a priori the spacer geometry, because the necessary information about the spacer geometry will be also obtained by regression of few experimental data.

1. Introduction

During the past few decades, organic solvent nanofiltration (OSN) has received a great deal of attention in industry, with applications ranging from solute enrichment and solvent recovery to impurity removal and catalyst recycle [1–4]. The development of a membrane process, such as OSN, usually involves several stages, starting from feasibility tests at laboratory scale, passing through pilot plant tests and finishing with large industrial scale processes, and the availability of a reliable simulation model could make the transition between these stages smoother and easier. Three levels can be distinguished within the general process modelling framework: membrane transport at molecular level, fluid dynamics and mass transfer in membrane modules and performance at process level [5]. Many studies on the transport mechanism through OSN membranes have been published; however, most of these studies were made using flat sheet membranes and at the level of molecular transport. Only few studies [1,6,7] described the performance of OSN processes with spiral-wound membrane modules using some simple, non-predictive membrane transport models; however, the effects of the modules at process level were

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Abbreviations: CFD, computational fluid dynamics; EA, ethyl acetate; OSN, organic solvent nanofiltration; SoA, sucrose octaacetate; UNIF-DMD, Dortmund modified UNIFAC method

[*] Corresponding author at: Department of Chemical Engineering, Imperial College London, Exhibition Road, London, SW7 2AZ, United Kingdom.

E-mail address: a.livingston@imperial.ac.uk (A.G. Livingston).

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not explored. To the best of the authors' knowledge, in the literature, there has been only one study which investigated fluid dynamics and mass transfer characteristics in a spiral-wound membrane module for OSN. In that study, Silva et al. [8] reported the experimental and simulated performance of a 2.5″ × 40″ spiral-wound STARMEM™ 122 membrane module in 0–20 wt% solutions of tetraoctylammonium bromide in toluene using a steady-state approach. In their modelling, two solution–diffusion based models were used and the corresponding model parameters were determined from flat sheet data; the former approach was a simple model, which assumed uniform pressure and concentration in both feed and permeate sides, while the latter was a complex model, which considered spatial concentration, velocity and pressure gradients. Although both models showed good agreement with the experimental data for the system under study, the authors pointed out that the complex model is more appropriate when the assumptions of both pressure and mass transfer coefficient constancy are not acceptable. The pressure drop and mass transfer correlations used in their study were adapted from Schock and Miquel's work [9]. Finally, in their study, the effects of membrane type and module size on the overall process performance were not explored.

Although the literature on spiral-wound membrane modules in OSN is scarce, many researchers studied fluid dynamics and mass transfer through plane, spacer-filled channels, characteristics of spiral-wound membrane modules, in aqueous solutions. To determine the mass transfer coefficient, three main methods have been used in the literature [9–24]: (i) direct measurements, which made use of optical or electrochemical methods; (ii) indirect measurements, which were based on regression of membrane performance data using a combination of film theory and membrane transport models; and (iii) computational fluid dynamics (CFD) simulations, which were based on a priori simulation of the module geometry. The pressure drop characteristics of a module were usually determined either from direct measurements, using accurate pressure gauges, or via CFD simulations [9,12–16,23].

Among the direct measurement studies, Johnson [10] applied an interferometer with a helium-neon laser as a light source to measure concentration polarisation in a reverse osmosis system. However, this method introduced a significant error due to the deflection of the light from a solute even in dilute conditions. Balster et al. [11] studied the effects of various single and multi-layer spacers on mass transfer using the limiting current technique. They concluded that the multi-layer spacer configurations exhibited significant mass transfer enhancement with respect to single-layer ones. However, in their work, the flow was passed along impermeable channel walls, which are obviously different from the semi-permeable membrane walls, presenting in a membrane module. Schock and Miquel [9] measured the pressure drop through various feed and permeate spacer filled channels. A friction coefficient correlation was used to fit their experimental data, in the form of Eq. (1):

$$f = \frac{2\Delta P d_h}{\rho u^2 L} = a\left(\frac{d_h \rho u}{\mu}\right)^b = aRe^b \tag{1}$$

f is the friction coefficient and ΔP is the pressure drop through the channel. d_h is the hydraulic diameter of the channel, L is the length of the channel, ρ is the density of the solution, u is the velocity of the flow along the channel, μ is the dynamic viscosity of the solution and Re is the Reynolds number. a and b are the coefficient and the exponent of Reynolds number in the friction coefficient correlation, respectively. In Schock and Miquel's work, the spacer geometry was measured using a light microscope; however the authors pointed out that this might not be a very accurate technique to obtain the characteristic dimensions of permeate spacers, due to their complicated geometry. They found that the geometry of the feed spacer had little effect on the friction coefficient, while the geometry of the permeate spacer showed more significant effects. Kuroda et al. [12], Da Costa et al. [13] and Schwinge et al. [14] also studied the effects of spacer geometry on the friction coefficient. Various types of spacers were considered in their work and a number of experimentally measured pressure drop data were reported. The significant effects of the spacer geometry on pressure drop performance were observed.

Among the indirect measurement approaches, Schock and Miquel [9] performed regression of flat sheet membrane performance data to determine the mass transfer coefficient in a plane, feed spacer filled channel using the combination of film theory and an empirical membrane transport model. This empirical transport model assumes that the permeate flux is linearly dependant on the difference between applied pressure and osmotic pressure. The authors used a dimensionless correlation to describe the mass transfer coefficient, in the form of Eq. (2):

$$Sh = \frac{kd_h}{D} = \alpha Re^{\beta} Sc^{\lambda} = \alpha\left(\frac{d_h \rho u}{\mu}\right)^{\beta}\left(\frac{\mu}{\rho D}\right)^{\lambda} \tag{2}$$

Sh and Sc are the dimensionless Sherwood and Schmidt numbers, respectively. α, β and λ are the coefficient and the exponents in the Sherwood correlation equation. k is the mass transfer coefficient and D is the diffusivity of a solute in a solvent. Four types of commercial feed spacers were studied in their work; interestingly, no effect of the spacer geometry on the mass transfer coefficient was observed. A similar methodology was used by Da Costa et al. [13] and Schwinge et al. [14], who on the other hand, found that the spacer geometry does affect the mass transfer coefficient in spiral-wound membrane modules. Interestingly, their correlations [13,14] for the same spacers showed good agreement. Although the simple, empirical membrane transport model worked well in these studies carried out in dilute aqueous solutions [9,13,14], it has yet to be established whether the model will describe mass transfer during OSN in spiral-wound membrane modules.

Finally, among the CFD studies on the mass transfer coefficient, Da Costa et al. [15] and Karode and Kumar [16] performed 2 dimensional CFD simulations to visualize the fluid flow structure through various spacer filled flat channels. They found that the flow path was affected by a combination of flow attack angle, filament size, mesh size and angle between crossing filaments. Their friction coefficient correlations for the same spacers showed good agreement. Li et al. [17–19] performed 3 dimensional CFD simulations to study flow characteristics and mass transfer in spacer-filled channels and the results were compared with their experimental data. Good agreement was reported and various correlations in the form of Eq. (2) were presented. Fimbre-Weihs and Wiley [20] presented both 2 and 3 dimensional CFD simulations to study mass transfer in a spacer filled channel, positioned at 45° and 90°, with a single Schmidt number ($Sc = 600$). The authors reported that the exponent of Reynolds number in the Sherwood correlation, represented in the form of Eq. (2), is 0.591. Koutsou et al. [21] even reported a significant amount of mass transfer coefficient data for ten types of spacers and discussed the effect of Schmidt number on the mass transfer coefficient using CFD simulations. Furthermore, various Sherwood correlations were reported based on average mass transfer coefficients and good agreement with Li et al.'s work [19] was observed. Kostoglou and Karabelas [22] developed a comprehensive model which incorporates small scale CFD results on the retentate side and accounts for permeate variables as a step forward to predict the performance of spiral-wound membrane modules in desalination. Karabelas et al. [23] performed a parametric study on the