Pergamon

Wat. Sci. Tech. Vol. 40, No. 4–5, pp. 269–276, 1999

0273–1223/99 $20.00 + 0.00

PII: S0273-1223(99)00508-9

ASSESSMENT OF THE DESIGN PARAMETERS FOR WASTEWATER TREATMENT BY REVERSE OSMOSIS

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ABSTRACT

A modelling procedure is presented to predict the fluxes and solute concentrations in different flows for reverse osmosis (RO) spiral-wound modules. Important underlying factors for this procedure are the osmotic pressure for various solutions, the hydrodynamic flow profile in the concentrate channel, and the intrinsic separation characteristics of the membrane material. Experiments were carried out using a flat sheet test cell to determine the parameters of the mass transport model. Results of residence time distribution (RTD)-measurements on an industrial spiral-wound module were used to determine macroscopic fluid flow regimes resulting in the definition of dead volume fraction, average residence time and Pe-number. The evaluation of the modelling procedure has been based on experimental data of an industrial membrane plant system. © 1999 IAWQ Published by Elsevier Science Ltd. All rights reserved

KEYWORDS

Hydrodynamics; mass transport; modelling; reverse osmosis; spacer; spiral-wound module.

INTRODUCTION

Reverse osmosis (RO) finds increasing applications as a separation technique in chemical and environmental engineering where desalination, selective separations or wastewater treatment are well established examples. The spiral-wound membrane module is most widely used.

The two most important features are the permeate flux and the separation efficiency with respect to salts. Both parameters depend not only on intrinsic membrane properties, solution characteristics and operating conditions, but also and to a large extent on the geometry of the spiral-wound module and the hydrodynamic flow regime.

The driving force for the water transport across the membrane is the pressure difference between the operating pressure and the osmotic pressure, which is a characteristic of the solution only and not of the nature of the membrane. The salt permeability is primarily a function of the concentration gradient across the membrane. In this case the concentration at the membrane surface needs to be considered, and this concentration frequently exceeds the bulk feed concentration due to polarisation. The polarisation phenomena are a function of the flow and concentration at the concentrate side, and will be less pronounced provided a more turbulent flow is reached, thus also favouring higher permeate fluxes. The presence of the concentrate spacer in the spiral-wound module creates turbulence and thus reduces the effects of polarisation. The operating pressure, i.e. the driving force for permeate flux decreases along the membrane axis as a result of the pressure drop created by the spacer material and of an increased osmotic pressure due to increased solute concentration. The definition of the flow regime is important and obtained through the measurement of the residence time distribution (RTD) which clearly shows the presence of dead volume in the spiral-wound membrane module: only a fraction of the volume is available for the feed flow hence the active surface area and the feed velocity must be corrected.

To predict fluxes and salt rejection, the proposed modelling procedure for RO separation in a spiral-wound module makes use of a discretisation approach. The membrane surface is divided along its length in a series of discrete unit sections. Over a given section all the foregoing aspects are considered, with integration of material balances.

METHODS

Lab scale experiments to define intrinsic membrane properties

Intrinsic properties of membranes were defined by a number of experiments carried out using a 'flat-sheet' membrane cell incorporated in an experimental set up as illustrated in Fig. 1. The positive displacement pump is capable of feeding maximum 2.3 m^3/h. A control valve sets the required pressure. Flow meters and pressure gauges monitor important experimental parameters. Retentate and permeate were recycled to the feed tank so as to keep the feed concentration unchanged. Fluxes and solute concentrations of the different flows were measured as a function of pressure and for different feed concentrations.

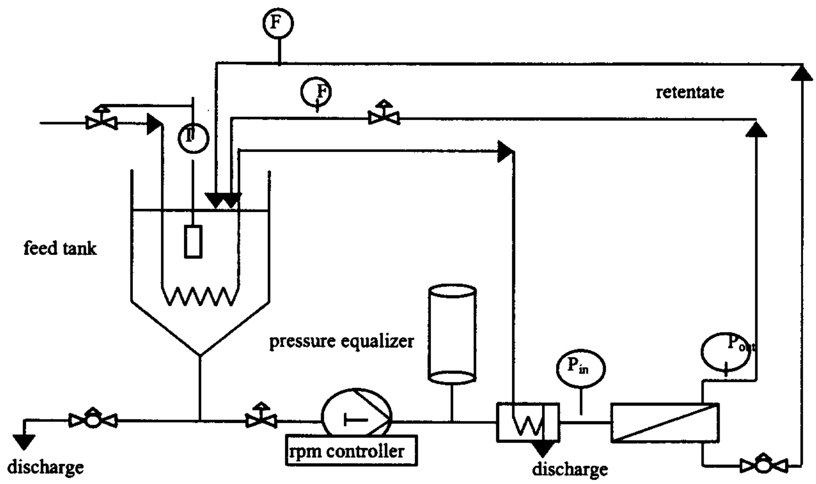

Figure 1. Flow sheet of the experimental set-up.

Lab scale experiments to define the macroscopic fluid flow in the concentrate channel

The experimental set up is shown in Fig. 2. The membrane module is of spiral-wound asymmetric nature (DESAL 4040 11AD). Because no pressure is applied to the feed solution, no permeation will occur through the membrane. The solution will only flow at the known flowrate through the 'concentrate channel' of the spiral-wound module. The method of this investigation is that of a step injection of the tracer solution after establishing a flow regime using demineralized water. To avoid preferential flow patterns in the concentrate spiral, the test cell is provided with an inlet zone. To eliminate the effect of this zone on the RTD of the membrane module, the principle of convolution needs to be applied.

The flows of demineralized water and tracer solution were metered by a rotameter located downstream of the membrane module. Tracer concentrations are measured from electrical conductivity of the solution: a linear relationship between the concentration and the conductivity is valid at low concentrations. Details of the techniques are described elsewhere (Van Gauwbergen and Baeyens, 1997).

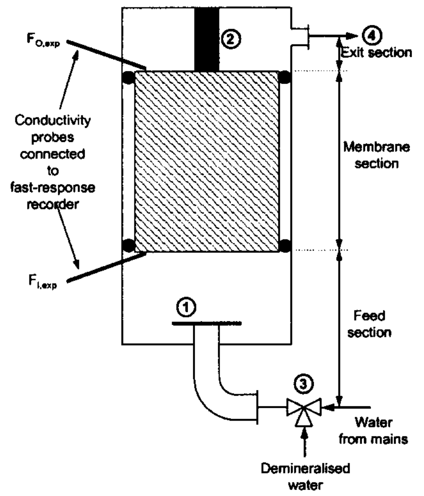

Figure 2. Details of the RTD experimental layout. 1: Flow equalizer; 2: Membrane positioning; 3: Three-way valve; 4: To flowmeter.

Industrial scale experiments

To evaluate the usefulness of the modelling procedure for spiral-wound modules, experiments were carried out at an industrial RO-plant, designed to treat wastewater issued from producing microelectronic chemicals. A schematic flow diagram of the 3-staged RO-plant is shown in Fig. 3. During the validation testing of the unit, a batch of synthetic wastewater containing NaCl, Na_2SO_4, KNO_3 or Na_3PO_4 was fed to the RO1-unit. Due to the limited batch-volume, only RO2 could be operated in a steady state for longer periods. Since stationary experimental results are mandatory for the evaluation of the modelling procedure, only the measured data of the RO2-unit could be used.

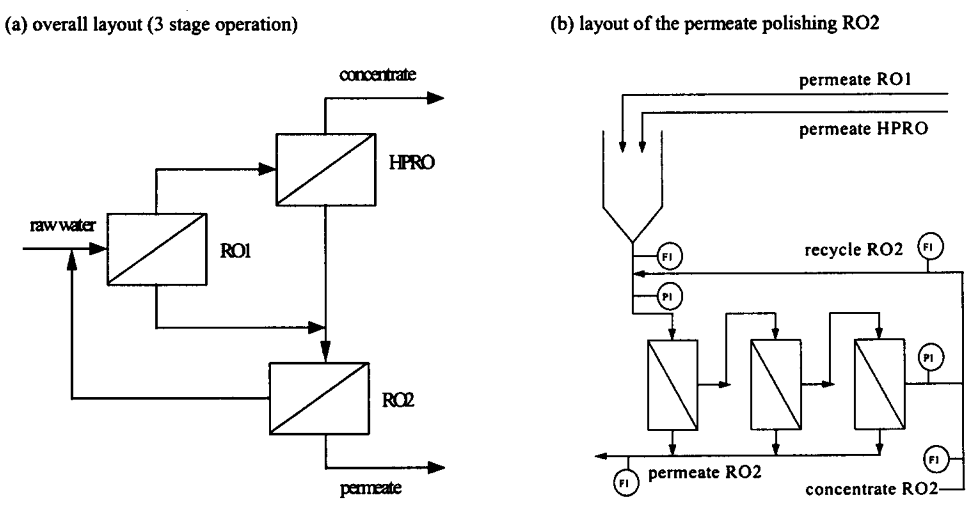

Figure 3. Schematic flow diagram of the RO industrial plant.

RESULTS AND DISCUSSION

Osmotic pressure

The osmotic pressure for electrolyte solutions can be predicted using the Van't Hoff equation for low concentrations. It has been found however (Van Gauwbergen *et al.*, 1997) that Van't Hoff overrates the osmotic pressure at higher concentrations and that the Pitzer equation predicts this solution characteristic with greater confidence.

Hydrodynamic flow regime

Dead zone volume. The normal feed rate for the DESAL 4040 spiral-wound module in industrial applications varies between 700 and 3600 l/h. RTD experiments are carried out for this range of feed flow rate. According to Westerterp et al. (1984), the experimental average residence time in the concentrate channel can be defined as follows :

$$\tau_{channel,exp,av} = \text{average}\left\{\left[\int_0^1 t \cdot dF(t)\right]_{out,exp} - \left[\int_0^1 t \cdot dF(t)\right]_{in,exp}\right\} \quad (1)$$

The experimental volume of the concentrate channel available for flow is determined from the slope of the $1/\tau_{channel,exp,av}$ vs flow curve (see Fig. 4). Results are summarized in Table 1.

Table 1. Calculation of the dead zone volume from RTD-experiments

experimental channel volume						
Flow rate [l/h]	548	1065	1581	2097	2613	
$\tau_{channel,exp,av}$ [sec]	35.8 ± 0.3	18.1 ± 0.6	10.3 ± 0.3	7.7 ± 0.1	5.8 ± 0.2	$V_{channel,exp}$ = **4.1 litre**
semi-theoretical channel volume						
immersion liquid	surface tension [dynes/cm]		$V_{channel,theo}$ [litre]			
water	73.05		4.58			
isopropanol	24.6		4.55			$V_{channel,theo}$ = **4.57 litre**

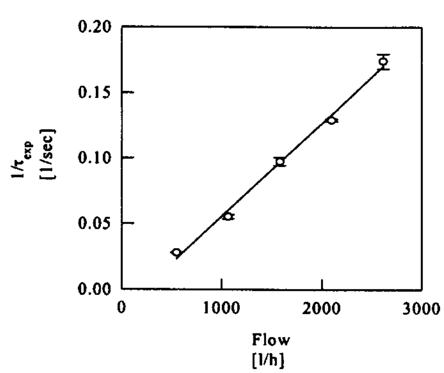

Figure 4. Determination of the experimental channel volume.

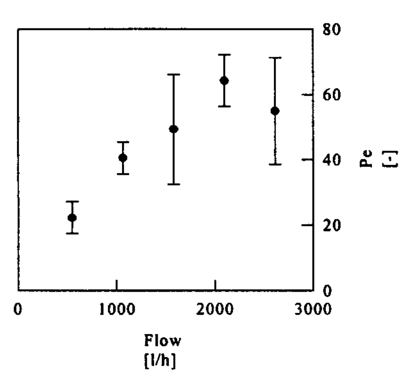

Figure 5. Pe-number for the concentrate flow regime as a function of flow rate.

The semi-theoretical channel volume can be determined by immersion experiments. The spiral-wound module is inserted in a glass tube with known dimensions and the tube is filled with a precise volume of liquid. After a long period of time the liquid reaches a stationary level and the volume of liquid inside the module can be calculated from the height of this level, the dimensions of tube and module, and the porosity of the membrane and the permeate collector tube. As surface tension might affect the liquid penetration in the channels, immersion experiments are carried out using both water and isopropanol. Results are given in Table 1. Since the experimental channel volume is smaller than the semi-theoretical one, an approximately 10% dead volume is present in the spiral-wound module, independent of the feed flow rate. This dead zone reduces the effective and active surface of the membrane but proportionally increases the feed velocity in the active concentrate channel.

Flow regime in the concentrate channel. As previously used for small scale spiral-wound modules (Van Gauwbergen and Baeyens, 1997), it is assumed that the flow regime in the industrial concentrate channel can also be described by the plug flow with disperion model (PFDM) [eq. (2)] (Levenspiel, 1979) : a limited dispersion is superimposed upon a regime of plug flow, and is characterized by the dimensionless Péclet number. A large value of Pe indicates that deviations from plug flow are minor whereas the PFDM-model approaches perfect mixing at low Pe numbers.

$$E_{m,theo}(t) = \frac{1}{\sqrt{4\pi/Pe}} \cdot \frac{1}{\tau} \cdot \exp\left\{ \frac{-(1-t/\tau)^2}{4/Pe} \right\} \tag{2}$$

When combining the convolution principle and the PFDM-model for the membrane section, the optimum value for the parameter Pe can be determined by minimizing the sum of squares of the function deviations between the calculated and the experimental data points of the F-curve.

$$F^* = F_{I,exp} \; * \; E_{m,theo} \tag{3}$$

$$DSQ = \sum \left(F^*(t) - F_{O,exp}(t) \right)^2 \cdot \Delta t \tag{4}$$

The results of the calculations applied to the DESAL 4040 spiral-wound module are summarized in Fig. 5. At these values of Pe, plug flow with limited dispersion appears to be representative of the flow regime in the concentrate channel.

<u>Mass transport through the membrane</u>

Flat sheet experiments, using the same DESAL 11AD membrane, show that the mass transport through the membrane can be described by the solution-diffusion model (Wijmans and Baker, 1995).

	$= A(\Delta P - \Delta\pi)$	[m/sec]	(5)
J_s	$= B(\Delta C)$	[mole/m^2.sec]	(6)
	with A = water permeability	[m/sec.Pa]	
	B = salt permeability	[m/sec]	

Rearranging the equation yields

$$\frac{1}{R} = 1 + \frac{B}{J_v} \tag{7}$$

This dependency is verified and validated by the results of Fig.6, proving the applicability of the solution-diffusion approach. The membrane constants A and B have been measured using the same electrolyte solutions as in the industrial scale experiments (Daloze, 1998). From the results, summarized in Table 3, it can be concluded that the water permeability A is a function of the feed concentration of the solute : A decreases with increasing feed concentration. The solute permeability B depends on the nature of the species to be rejected by the membrane, and the DESAL 11AD membrane shows the highest selectivity towards Na_2SO_4.

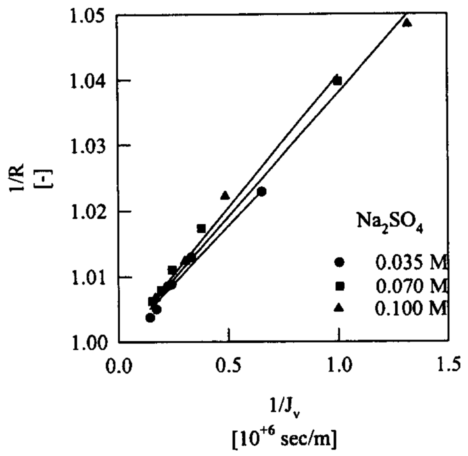

Figure 6. Validation of the solution-diffusion model.

Table 3. Intrinsic characteristics ('solution-diffusion' model) of the DESAL 11AD membrane

solute	feed concentration [mole/l]	A [10^{-7} m/sec.bar]	B [10^{-6} m/sec]
KNO_3	0.0495	7.72	1.198
	0.0990	7.33	1.086
	0.1980	6.78	1.783
Na_2SO_4	0.0350	7.68	0.1516
	0.0700	7.21	0.1566
	0.1000	6.97	0.1323
Na_3PO_4	0.0613	7.67	0.3230
	0.1230	7.66	0.4368
	0.1840	7.56	0.4976

Modelling procedure

The basic equations outlined in the foregoing paragraphs are integrated together with the mass balances in an overall modelling procedure for spiral-wound membrane modules. Schock and Miquel (Schock and Miquel, 1987) have shown that the bent envelope does not significantly influence the flow behaviour in the concentrate channel. The unwound module is therefore considered and a discretization approach is used. (see Fig. 7). The membrane surface is divided along its length in a series of separate unit sections and flat sheet intrinsic characteristics are used. The osmotic pressure in each section is calculated using Pitzer's equation.

Permeate fluxes and permeate concentrations so calculated are in reasonable agreement with the experimental findings for the industrial scale set up as illustrated in Fig. 8. Additional effects of possibly enhanced polarisation in the final sections of the series needs however to be accounted for.

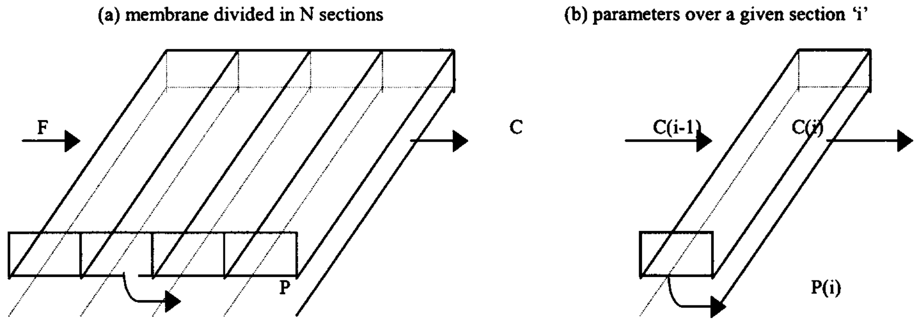

Figure 7. Discretisation approach to spiral-wound membrane modules.

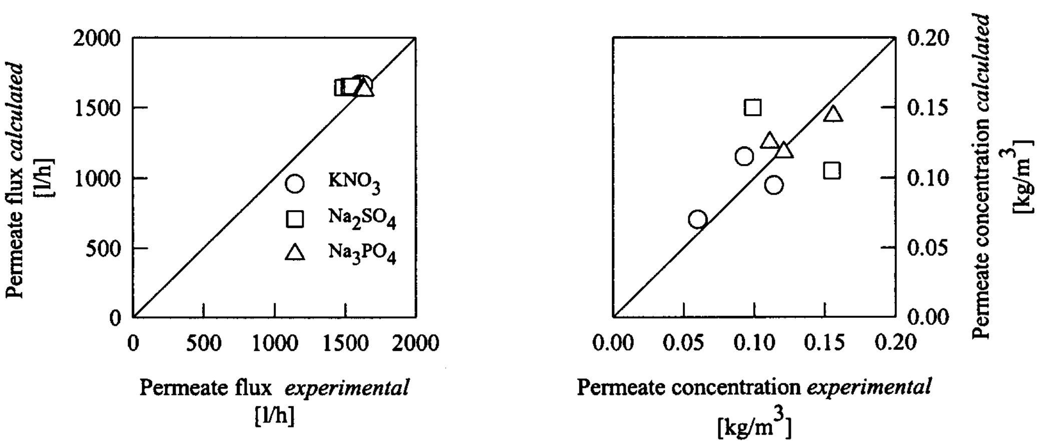

Figure 8. Results of the modelling procedure.

CONCLUSIONS

Intrinsic membrane characteristics were determined from experimental results obtained in a flat-sheet test unit resulting in water and salt permeability coefficients of the solution-diffusion transport model.

The residence time distribution of flow elements in the concentrate channel of an industrial spiral-wound membrane defines average residence times, fractional dead volume and dimensionless dispersion number (Péclet) as a function of the flow rate in the concentrate channel.

The modelling of an industrial spiral-wound membrane by discretisation of the membrane surface is based upon the experimental flat sheet and RTD-results, together with calculated values of the osmotic pressure. Comparison of predicted and calculated permeate flux and concentration are in reasonable agreement although further work on e.g. polarisation along the membrane sections is required.

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