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Comparison Studies of Applied Pressure and Concentration Gradient Driving Forces in Ceramic Nano-Filtration Membrane for the Production of Intravenous Salt Solution

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Abstract: In this study, the boundary-resistance layer model and solution-diffusion model were used to investigate the applied driving pressure force technique and the concentration driving force technique, respectively, for the production of intravenous drip solution. A 5 kD monolithic membrane coated with Al_2O_3 and TiO and NaCl aqueous solution as the feed solution was used. The results show that the boundary-resistance layer model diffusivity coefficient, $D = 1.8\times10^{-9}$ m^2 sec^{-1} and the mass transfer coefficient, $k = 1.19^{-}10^{-4}$ m sec^{-1} which were both slightly higher than the solution-diffusion model. Applying Fick's law for the solution diffusion model, the calculated operating pressure inside the lumen was 15 kPa. Clearly, the findings suggested that the boundary-resistance layer model should be chosen for the production of pure and sterile intravenous salt solution as it provided higher diffusivity and mass transfer coefficient than the solution-diffusion model.

Key words: Intravenous solution, nanofiltration, boundary-resistance layer model, solution-diffusion model

INTRODUCTION

During the past 40 years, membrane filtration processes have found wide application in the pharmaceutical industry in the preparation and processing of parenteral solutions. Though the health significance of particulate matter introduced into parenterals is at best controversial, injectable solution should not contain particulates that could be introduced into the blood stream. Basically, intravenous solutions that are labelled as being sterile are processed by one of the two methods called terminal sterilisation and aseptic processing. Although the sterilisation is accomplished through the use of steam and autoclave, membrane filters are used to reduce bioburden and for pyrogen management (Wesley, 2000; Goldman and Maki, 1973). Mass transfer is the phrase commonly used in engineering for physical processes that involve molecular and convective transport of atoms and molecules within physical systems.

The driving force for mass transfer is a difference in concentration; the random motion of molecules causes a net transfer of mass from an area of high concentration to an area of low concentration. The amount of mass transfer can be quantified through the calculation and application of mass transfer coefficients. In case of membrane process, the findings of studies examining the various parameters and variables such as volume or mass flux of permeate, transmembrane pressure (TMP), feeding concentration, temperature and rejection of particulate show significant improvements. Earlier researchers (Paris *et al*., 2002, Ahmad *et al*., 2006 and Tung and Jen 2001) have examined the flux pattern of Dextran T500 solutions using various models such as boundary-resistance layer model, modified boundary-resistance layer model and osmotic pressure model. They have described experimentally and theoretically the differences between the various models. The results show that the modified boundary-resistance layer model has a good agreement and is best fitted with the experimental data.Despite that, very little research has been done for production of intravenous solution using membrane processes. The purpose of this study is to investigate in detail the application of crossflow filtration under different transmembrane pressures (TMP) and osmotic pressures, as well as the different concentrations in nanofiltration membrane for producing intravenous solution.

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THEORY

Applied pressure driving force: Applied pressure driving force technique states that the transport of solvent is directly proportional to the applied pressure. The feed solution that passes through the membrane will be separated into two parts; permeate and retentate. The appropriate models that can be used for a dilute solution where both the solvent and solute can easily pass through the membrane are the boundary-resistance layer and osmotic pressure model.

Recently, a method for predicting fluxes in laminar ultrafiltration of macromolecular solutions was proposed by Tung and Jen (2001). The method is a modified osmotic pressure model where the mass transfer coefficient is estimated from the Leveque solution and the diffusion coefficient is evaluated from the mean concentration of the concentration polarisarition layer. Kargol (2001) has developed a mechanistic model of transport processes in porous membranes where a simple molecular model of membrane transport processes generated by the hydrostatic pressure (ΔP) and the osmotic pressure ($\Delta\pi$) was presented.

Boundary-resistance layer and osmotic pressure models: The general equation of the boundary-resistance layer model which describes the permeate flux in terms of the applied pressure gradient and resistance to transport or flow across the concentration polarisation (CP) boundary layer and the membrane can be expressed in two equivalent forms based on Darcy's law:

$$\text{Flux}, J = \frac{\Delta P - \Delta\pi}{\mu(R_m + R_{cP})} \quad (1)$$

where, R_m is the resistance of the membrane, R_{CP} the polarised layer resistance and $\Delta P = P_m - P_p$. By plotting graph of flux versus TMP, the value of R_{CP} can be calculated from the slope of the graph as:

$$\text{Slope} = \frac{1}{\mu(R_m + R_{CP})} \quad (2)$$

The mass transfer coefficient k of the concentration polarisation layer is determined from the Leveque equation for a flow in laminar regime, as presented below:

$$\text{Slope} = \frac{1}{\mu(R_m + R_{CP})} \quad (3)$$

where D is the solute diffusivity in the solvent.

Concentration driving force: Concentration driving force technique is the movement of molecules from high concentration to low concentration through a semi-permeable membrane by diffusion process. Only molecules that are small enough to fit through the membrane pores are able to pass through the membrane until the entire volume of the solution in the system reaches equilibrium. Once equilibrium is reached, there is no further net movement of the substance across the membrane. By contrast, large molecules that cannot pass through the membrane pores will remain at the same side of the membrane. The movement of the molecule can be described by the solution-diffusion model, Wijmans and Baker (1995).

Solution diffusion model: The solution-diffusion theory which uses Fick's first law is a phenomological model that describes the potential or flux relationship for diffusive mass transfer in binary system. It states that the diffusive flux is proportional to the concentration gradient, mole fraction or the chemical potential at constant pressure. The solution diffusion model can be described by the following general form of Fick's law:

$$N_i = -D_i C \frac{d(\mu_i / RT)}{dx} \quad (4)$$

where:

N_i : The mass of component i transported, kmol m^{-2} sec^{-1}

D_i : The diffusivity of component i, m^2 sec^{-1};

C : IS The solute concentration, kmol m^{-3};

μ_i : The chemical potential of the diffusing substance, J $kmol^{-1}$.K

x : The distance, m.

In most cases, activity coefficients are close to one; hence the Fick's first law can be written as:

$$N_i = -D_i \frac{dC_i}{dx} \quad (5)$$

Assuming D_i is a constant, C_i is in equilibrium and with the same membrane thickness the Fick's law may be written as:

$$N_i = D_i \frac{(C_f - C_p)}{l} = D_i \frac{\Delta C}{l} \quad (6)$$

where:

l: The thickness of the membrane active layer

C_f and C_p: The concentration of the feed and the concentration of the permeate, respectively.

By plotting the flux versus concentration gradient, ΔC (g L^{-1}), the diffusivity of the solution, D, can be calculated from the slope of the graph as follows:

$$\text{Slope} = \frac{D}{\Delta l} \tag{7}$$

Wijmans and Baker (1995) have derived the phenomenological equations using the solution-diffusion model, where the flux is proportional to the gradient in the chemical potential. The model assumes that the pressure within the membrane is uniform.

Therefore, the chemical potential gradient across the membrane is expressed as the concentration gradient only. For a single solute, the Nernst-Haskell equation can be used to estimate the diffusion coefficient Geankoplis (2003), as presented below:

$$D = 8.928 \times 10^{-10} T \frac{\left(\frac{1}{n_+} + \frac{1}{n_-}\right)}{\left(\frac{1}{\lambda_+} + \frac{1}{\lambda_-}\right)} \tag{8}$$

where:

n_+ = the valence of the cation (Na^+)

n_- = the valence of the anion (Cl^-)

λ_+ and λ_- = the limiting ionic conductance in dilute solutions (A/cm^2) (V/cm) (q equiv./cm^2).

MATERIALS AND METHOD

Materials: All reagents used are of analytical grade. Sodium chloride (NaCl) powder with purity of 99% was purchased from Kinabalu Scientific supplier. A 5 kD monolithic membrane module coated with Al_2O_3 and TiO was supplied by Orelis Company, France. It has 19 channels (lumens), where each lumen has a diameter of 3.5 mm; a length of 35.0 cm and a total membrane area of 816.0 cm^2.

Water permeability test: The water permeability test was run by using the same experimental set-up used for the applied pressure driving force technique study as shown in Fig. 1. Distilled water from the feed tank was pumped into the membrane across the control valve for 30 min. The TMP was monitored by using two pressure gauges at both ends of the membrane. During the filtration process the permeate flux was monitored by measuring the volume of permeate in a measuring cylinder for 1 minute. The permeate flux rate was calculated by dividing the volume of permeate collected in 1 min to the membrane area.

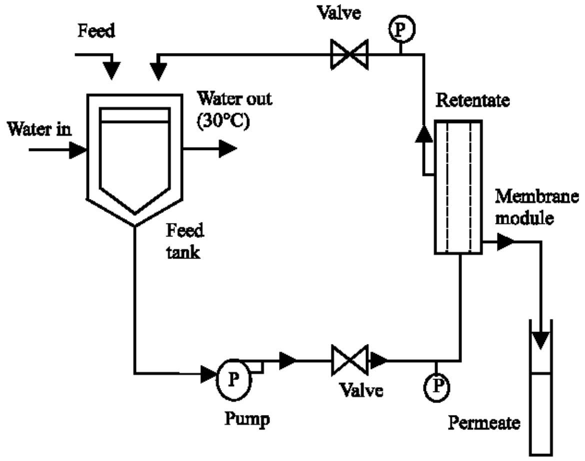

Fig. 1: Schematic diagram of cross-filtration experimental set-up for applied pressure driving force study

Crossflow filtration technique experiment: Figure 1 shows a schematic diagram of a cross flow filtration set-up used in the applied pressure driving force experiment. It consists of a feed tank, gear pump and membrane module. Prior to the experiment, the membrane was precompacted by circulating distilled water at a temperature of 30°C and $\Delta P = 0$ kPa for 30 min. Then, the water was discharged and replaced with salt solution at a concentration of 0.9 g L^{-1}. The salt solution was prepared by mixing the sodium chloride powder with distilled water to 0.9 g L^{-1} at ambient temperature. Then, the salt solution was pumped into the membrane via a variable-speed gear pump (PQM-1/230, Greylor Co) at a temperature of 30°C.

The TMP was monitored using two pressure gauges at both ends of the membrane and was varied between 17 and 69 kPa. The temperature was controlled by using a temperature-controlled waterbath (Model 9101, Polyscience, Niles 1L60714). The experiment was run for about 1 h. The permeate flux was monitored during the filtration by measuring the volume of permeate in the measuring cylinder for 1 min. At the end of the experiment, the membrane was cleaned and regenerated with surfactant (Decon 90) for 1-2 h and subsequently with pure water for 1 h. The membrane resistance was recalculated to determine the membrane restoration.

Concentration gradient technique experiment: Figure 2 shows a schematic diagram of the experimental set-up used in the concentration gradient experiment. It consists of two jacketed feed tanks, two gear pumps and a membrane module. The sodium chloride powder was mixed with distilled water to three different concentrations (i.e., 20, 25 and 30 g L^{-1}) at ambient temperature. The sodium chloride solution was pumped into the membrane lumen from the jacketed feed Tank 1, via a variable-speed

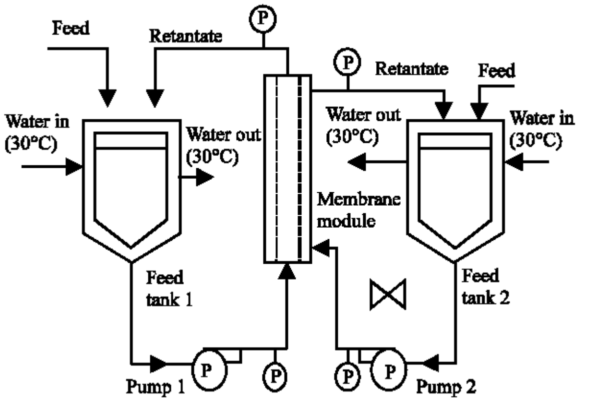

Fig. 2: Schematic diagram of experimental set-up for concentration driving force study

gear pump (PQM-1/230, Greylor Co). The temperature was controlled by using a temperature-controlled waterbath (Model 9101, Polyscience, Niles 1L60714). Distilled water (solvent) was pumped into the outside membrane lumen from the jacketed feed Tank 2 and the temperature was controlled by using a temperature-controlled waterbath (Model Typ E100, Lauda and Co. Kg, Germany).

The TMP was monitored by using two pressure gauges at both ends of the membrane. The operating temperature was maintained at 30°C by using the circulated water. The pressure was constantly controlled throughout the experiment while the feed concentration of the salt solution was varied between 20 and 30 g L^{-1}. At each concentration condition, the permeate flux concentration was measured by using a conductivity meter every 1 min for a period of 12 min. Finally, the membrane was cleaned and regenerated with surfactant (Decon 90) for 1-2 h and subsequently with pure water for 1 h. The membrane resistance was recalculated to determine the membrane restoration.

Determination of salt concentrations: The permeate solutions which were collected from the permeate stream in a measuring cylinder was immediately tested by using conductivity meter (Model PHYWE 07138.00) at a temperature of 26°C. The operating temperature was maintained at ambient temperature (26°C).

RESULTS AND DISCUSSION

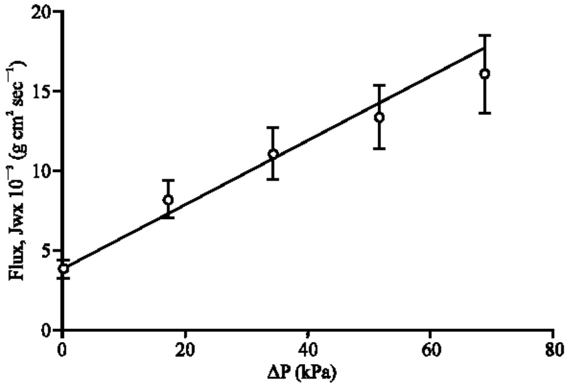

Fig. 3: Water permeability at different pressures

Determination of water permeability: Figure 3 shows the relationship between pure water fluxes at different TMP ranged between 0 and 69 kPa. The water permeability, L_p was calculated from the slope of the graph and it was 1.95×10^{-7} g cm^{-2}. Pa.s. The membrane resistance, R_m, calculated from (1) was 6.4×10^{9} cm^2 g^{-1}. The result also showed that at TMP = 0 kPa, the flux was 3.8×10^{-3} g cm^{-2} sec^{-1}. This indicated that the operating pressure inside the lumen was higher than the outside. After regeneration, the calculated membrane restoration was 98.7%.

Effect of transmembrane pressure (TMP): The Darcy's law is used to describe the effect of pressure on the flux (Kagol, 2000; Paris, 2001) as shown Fig. 4. As can be seen, the flux increased with an increasing TMP because sodium chloride and water have low molecular weights and could easily pass through the membrane. An increase in the TMP had caused more amount of sodium chloride solution could pass through the membrane. Higher TMP had resulted in the higher volume fluxes, which was due to an increase in the pressure driving (Ahmad *et al*., 2006).

The relationship of permeates flux with TMP ranged between 0 kPa and 69 kPa is shown in Fig. 4. The result showed that the salt permeate flux increased with an increasing TMP. At ΔP = 0 kPa, the flux of salt solution was 6.5×10^{-3} g cm^{-2} sec^{-1}. This indicated that pressure was applied from the inside of the lumen. From Fig. 4, the calculated value of the pressure was 24 kPa. This pressure is due to the combination of the osmotic pressure and applied pressure.

Effect of feed salt concentrations: The osmotic pressure gradient had been suggested to reduce the energy required for pumping. The concentration gradient allowed the movement of molecules by diffusion from high to low concentration through a semi permeable membrane. Only molecules that were small enough were able to pass through the membrane boundary and reached the equilibrium when the net mass movements were zero within the entire volume of solution in the system. The equation used for concentration driving force was the Fick's law as shown in Eq. 6.

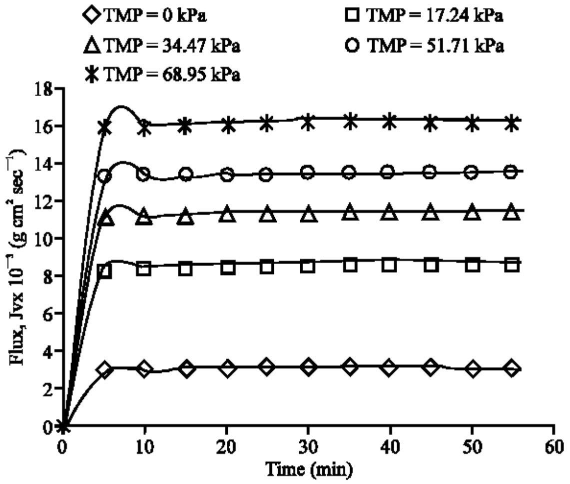

Fig. 4: Effect of TMP on permeates flux in crossflow filtration

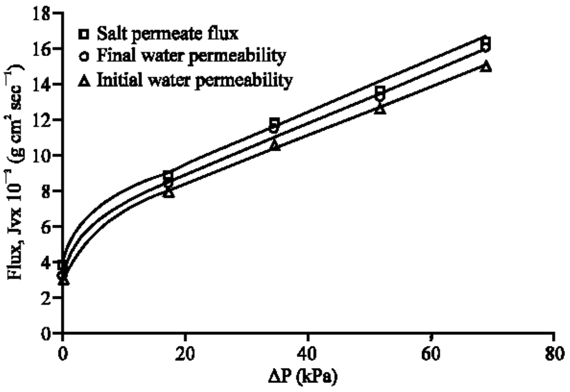

Fig. 5: Fluxed profile of the salt solution at T= 30°C and C_0 = 0.9 g L^{-1}

Figure 5 shows the concentration profile outside lumen at T = 30°C and ΔP = 0 kPa. The salt concentration outside the lumen was initially zero. The salt concentration increased with an increasing initial feed concentration inside the lumen because the solute and solvent flows did not occur independently but were simultaneously, whereby the solvent flow was due to the solute transport and solute flow was due to the solvent transport (Wijmans *et al*., 1984). Furthermore, as the solute flux occurred, the osmotic pressure flow also took place in the opposite direction (from low concentration outside the membrane to the high concentration inside membrane).

Figure 6 shows the effect of salt concentrations ranging between 20 and 30 g L^{-1} on the net calculated mass movement of the salt shown as fluxes. The result indicated that the flux increased with an increasing concentration but it decreased over the filtration time.

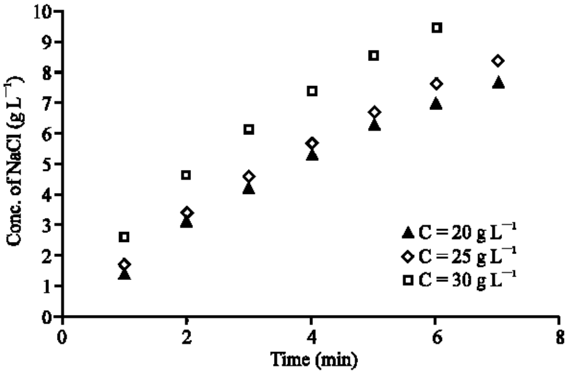

Fig. 6: Concentration profile outside the lumen

This phenomenon is because of the initial high osmotic pressure differences between the inside and the outside of the lumen that had caused high flux. However, over the time, the net mass transfer was reduced and reached the mass transfer equilibrium, hence the flux should become relatively zero. The final salt concentration outside the lumen should reach 0.9 g L^{-1}. From the results, it shows that after 12 min of operation the salt concentration were greater than 0.9 g L^{-1}, namely 1.4, 1.7 and 2.6 g L^{-1} for feed concentrations of 20, 25 and 30 g L^{-1}, respectively. Therefore, the initial feed concentrations should be lower than 20 g L^{-1}, or the operation should be stopped at less 1 min earlier to obtain the targeted concentration of drip solution at 0.9 g L^{-1}.

The net flux at operation of t = 5 min at different initial salt concentrations is indicated in Fig. 7. As can be seen, when the feed salt concentration was zero, the flux was 0.26×10^{-2} g m^{-2} sec^{-1}. This shows clearly that the operating pressure inside the lumen was higher than the outside pressure. The operating pressure inside the lumen was calculated from the y-axis intercept and it was 15 kPa.

Diffusion and mass transfer coefficients: Diffusion is a movement under the influence of a physical stimulus of an individual component through a mixture. The diffusion coefficient of applied pressure driving force technique and concentration driving force technique was determined by using the Nernst-Haskell equation as shown in Eq. 8. Mass transfer coefficient is defined as the rate of mass transfer per unit area per unit concentration difference and is usually based on equal molal flows. The mass transfer coefficients of applied pressure driving force technique and concentration driving force technique was determined by using the Leveque Eq. 3.

Table 1 shows the calculated parameters of the boundary-resistance layer model. Equation 1 was used to calculate R_{CP}, $\Delta\pi$ of distilled water and $\Delta\pi$ of salt solution. The value of membrane resistance is equal to

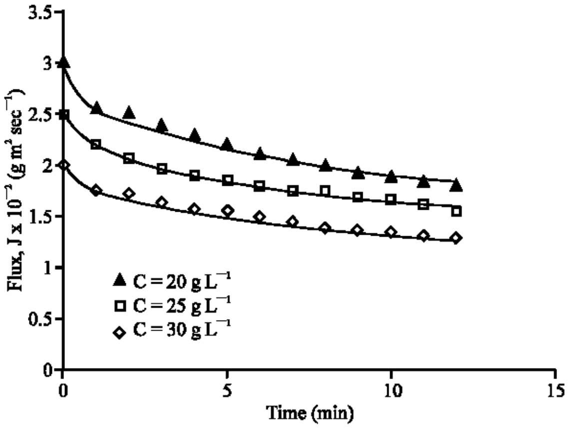

Fig. 7: Effect of concentration on fluxes profile

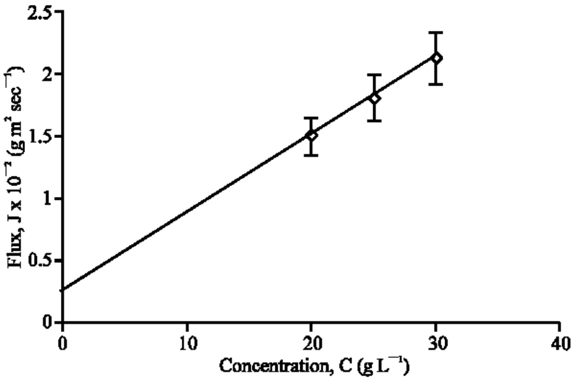

Fig. 8: The flux when t = 5 minutes at different feed concentrations (Condition: T = 30°C, ΔP = 0 kPa)

Table 1: Estimated parameters for the boundary-resistance layer model with (condition: temperature = 30°C, solute concentration in the retentate, $C_l = 0.9$ g L^{-1})

Parameter	Value
R_m (cm^2 g^{-1})	6.4×10^9
R_{cp} (cm^2 g^{-1})	6.2×10^8
$\Delta\pi$, water (kPa)	19.5
$\Delta\pi$, salt (kPa)	24.1
L_p (g cm^{-2} sec^{-1}.Pa)	1.86×10^{-7}
D (m^2 sec^{-1})	1.8×10^{-9}
k_i (m sec^{-1})	1.19×10^{-4}

Table 2: Estimated parameters for the solution-diffusion model, with condition: Temperature = 30°C, Solute concentration in the retentate, $C_i = 20$, 25 and 30 g L^{-1}

Parameter	Value
D (m^2 sec^{-1})	1.4×10^{-9}
k_i (m sec^{-1})	1.1×10^{-4}
$\Delta\pi$ (kPa)	14.6

6.4×10^9 cm^2 g^{-1}, which was in good agreement with the typical value for ceramic membrane reported by Mehta and Zydney (2005). The calculated value of the osmotic pressure of salt solution was 24.1 kPa, which was slightly higher than the osmotic pressure of water, 19.5 kPa. The value of diffusion coefficient and mass transfer coefficient were comparable with the typical value reported by Tung and Jen (2001) when Dextran T500 was used as the feed solution.

Table 2 shows the calculated parameters of the solution-diffusion model. The calculated osmotic pressure of salt for the solution-diffusion model was slightly lower than the osmotic pressure of salt for boundary-resistance layer model. Likewise, the calculated diffusion coefficient and mass transfer coefficient of solution-diffusion model were lower than the diffusion coefficient and mass transfer coefficient of boundary-resistance layer model, respectively.

CONCLUSION

The diffusivities of the salt solutions and the mass transfer coefficients through the membrane were successfully determined. The findings clearly suggested that the applied pressure driving force technique should be chosen for the production of pure and sterile intravenous salt solution as it provided higher diffusivity and mass transfer coefficient compared to the concentration driving force technique. However, further research needs to be conducted to compare the required power needed for pumping the solution for both methods as well as to study the effect of flow regime (i.e., velocity) on the mass transfer.

NOMENCLATURE

C = Solute concentration (kmol m^{-3})
C_i = Solute concentration in the retentate (g L^{-1})
C_p = Solute concentration in the permeate (g L^{-1})
C_f = Solute concentration in the feed (g L^{-1})
D = Diffusion coefficient (m^2 sec^{-1})
J_v = Permeate flux of salt solution (g cm^{-2} sec^{-1})
J_w = Permeate flux of pure water (g cm^{-2} sec^{-1})
k = Mass transfer coefficient (m sec^{-1})
L_p = Pure water permeability (g cm^{-2}.Pa.s)
L = Length of tubular membrane (m)
L = Thickness of the membrane active layer (m)
N_i = Mass of component i (kmol m^{-2} sec^{-1})
P_i = Inlet pressure of tube side (Pa)
P_m = Pressure of tube side (Pa)
P_b = Pressure of permeate phase (Pa)
ΔP = Transmembrane pressure, TMP (Pa)
r_m = Radius of hollow membrane (m)
R_m = Resistance of membrane (cm^2 g^{-1})
R_{CP} = Resistance of concentration polarisation boundary layer (cm^2 g^{-1})
R = Universal gas constant

T = Absolute temperature (K)
t = time (minute)
u_i = Velocity of solute in solvent ($m^2\ sec^{-1}$)
x = Distance (m)

Greek letters

π = Osmotic pressure (Pa)
$\Delta\pi$ = Osmotic pressure difference (Pa)
μ = Viscosity of solution (Pa.s)
μ_I = Chemical potential of diffusing substance (J $kmol^{-1}$.K)

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