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A New Method for Identifying Osmotically Limited and Gel Layer Controlled Pressure Independent Flux in Ultrafiltration

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Ultrafiltration is a pressure-driven membrane separation of chemical species from a solvent, usually water, using a semi-permeable membrane. The solution is brought into contact with the membrane under pressure that causes permeate to pass through the membrane while the solute is retained on the membrane surface. The net result is a layer of solution on the membrane surface with substantially higher solute concentration than that of the bulk solution. This is usually termed as concentration polarization. Concentration polarization has received considerable theoretical and experimental attention over last 25 years. It has been observed that with an increase in feed pressure, permeate flux first increases and then becomes more or less pressure independent.

Trettin and Doshi (1981) concluded that pressure independent filtration was gel layer controlled. These authors ignored the existence of the polarization layer and modelled the flux using Darcy's law, setting the filtration resistance in the pressure dependent filtration regime to the membrane resistance for pure water. The filtration resistance for the pressure independent regime was set to the sum of the membrane resistance for water and an additional resistance termed as the resistance of the so-called gel layer. Wijmans et al. (1984) interpreted flux data based on the osmotic pressure model (van Oers et al., 1992) and concluded that for macromolecules with molecular weight ranging from 10-100 kDa, osmotic pressure limitation was more likely than gel formation. These authors assumed that filtration resistance was wholly comprised of the membrane resistance for pure water.

In ultrafiltration (UF) of macromolecules using a retentive membrane, the polarization ratio (defined as the ratio of the solute concentration at the membrane surface to that in the bulk, i.e. C_w/C_b) is expected to be high due to the low diffusivity of the solute. In such a case, the polarization layer could offer an additional filtration resistance. In this work we propose a method for analysis of unsteady state filtration data to differentiate between osmotically limited and gel layer controlled pressure independent filtration regimes. Also, the proposed method could be used to estimate the additional filtration resistance offered by the polarization layer in macromolecular UF.

An unequivocal determination of whether pressure independent flux regime is osmotically controlled or gel layer dominated, is still open for discussion in the membrane literature. The present work reports a method that could be used to address this issue. It is shown that analysis of post steady state transient filtration data leads to clear demarcation of osmotically limited and gel layer controlled filtration. The method proposed in this work can also be used to estimate the additional filtration resistance offered by the polarization layer to the permeate flow in macromolecular ultrafiltration and has been verified experimentally. It has also been shown that the polarization layer thickness is not sensitive to the feed pressure but varies as a function of the bulk solute concentration; higher the bulk concentration, thicker is the polarization layer.

Une détermination claire de si le régime indépendant de flux de pression est osmotiquement contrôlé ou la couche de gel dominée est encore ouverte pour la discussion dans la littérature de membrane. Le travail actuel indique une méthode qui pourrait être employée pour aborder cette question. On lui montre que l'analyse des données passagères de filtration d'état d'équilibre ultérieur mène à la délimitation claire de la filtration contrôlée par couche osmotiquement limitée et filtration contrôlée par une couche de gel. La méthode proposée dans ce travail peut également être employée pour estimer la résistance supplémentaire de filtration offerte par la couche de polarisation à l'écoulement perméable dans l'ultrafiltration macromoléculaire. La méthode proposée a été vérifiée expérimentalement. Il y a aussi eu démonstration que l'épaisseur de couche de polarisation n'est pas sensible à la pression d'alimentation mais change en fonction de la concentration en bloc de corps dissous; plus élevée est la concentration en bloc, plus profondément est la couche de polarisation.

Keywords: membranes, osmotic pressure, gel layer, ultrafiltration, pressure independent filtration.

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Theory

Permeation of pure solvent through membranes is usually modelled using Darcy's law where the solvent flux is directly proportional to the effective pressure difference and inversely proportional to the filtration resistance. The constant of proportionality is the inverse of solvent viscosity (van Oers et al., 1992). Mathematically, this can be written as:

$$J_w = \frac{\Delta P}{\mu R_m} \quad (1)$$

where, J_w is the flux of pure solvent, R_m is the membrane filtration resistance and μ is solvent viscosity.

In the case of UF with a solute that is retained by the membrane, the driving force and the flow resistance would be modified due to the occurrence of concentration polarization or gel layer formation. Permeate flux for such cases can be obtained using the model reported in literature (Choe et al., 1986; van Oers et al., 1992):

$$J_s = \frac{\Delta P - \Delta \pi}{\mu R_f} \quad (2)$$

where, J_s is the solute flux, ΔP is the applied pressure, R_f is the total filtration resistance and $\Delta \pi$ the osmotic pressure difference across the membrane. Equation (2) has been used to model ultrafiltration since first proposed by Goldsmith (1971). Subsequently, several authors (Bhattarjee et al., 1992; Elimelech et al., 1998) have shown the equivalence of osmotic pressure and gel layer controlled concentration polarization models.

Differentiating Equation (2) with respect to time we get following equation:

$$\frac{dJ_s}{dt} = \frac{1}{\mu R_f} \left[\frac{d\Delta P}{dt} - \frac{d\Delta \pi}{dt} \right] - \frac{\Delta P - \Delta \pi}{\mu R_f^2} \left(\frac{dR_f}{dt} \right) \quad (3)$$

The second term on the right-hand side of the above equation was neglected as it was found to be too small to make any significant contributions to the calculations, e.g. on the right-hand side the second term was found to be 0.1-2.0% of the first term in the present study. For subsequent calculations Equation (4) was used.

$$\frac{dJ_s}{dt} = \frac{1}{\mu R_f} \left[\frac{d\Delta P}{dt} - \frac{d\Delta \pi}{dt} \right] \quad (4)$$

By analyzing post steady state transient filtration data obtained by reducing the driving pressure as a function of time, the actual filtration resistance can be calculated from the slope of the graph obtained by plotting dJ_s/dt versus $d\Delta P/dt$. The slope of the resulting graph would be linear if $d\Delta \pi/dt$ was equal to zero or the difference between $d\Delta P/dt$ and $d\Delta \pi/dt$ was constant. Moreover, since the shape of the flux versus pressure curve is non-linear (linear at low pressure and tapering off at high pressure) the only case where the variation of dJ_s/dt and $d\Delta P/dt$ would yield a linear slope would be at a $d\Delta \pi/dt$ value of zero. In the case of gel layer controlled filtration, the solute concentration at the surface of the membrane remains

constant (time independent) at the gel concentration. This would produce a linear slope for the dJ_s/dt versus $d\Delta P/dt$ graph. In the case of osmotically limited pressure independent filtration, the solute concentration at the surface of the membrane would be a function of the applied pressure. Therefore, after achieving a steady state in the experiment, a reduction in the driving pressure with time would cause a change in the solute concentration at the surface of the membrane with time, resulting in a non-linear graph between dJ_s/dt versus $d\Delta P/dt$. Using the simple technique described above, it is possible to unequivocally determine whether the pressure independent filtration regime is a result of a gel layer controlled filtration or an osmotically limited filtration.

Experimental

Materials

The UF membrane used in this work had a nominal MWCO of 6 kDa and was prepared in the laboratory on a 1079 backing (Tyvek, Supplied by Du-Pont). Membrane casting formulation contained 25% of polysulfone, (Radel-R Amoco, U.S.) and 21% polyvinylpyrrolidone (PVP) (Sigma, U.S.) in N-methyl-2-pyrrolidinone (NMP) (Anachemia, U.S.). The membrane was prepared by the phase inversion method, details of that are given elsewhere (Dal-Cin et al., 1994). Throughout this investigation, Polyethylene Glycol (PEG) (Fluka chemie AG, Switzerland) with 35 kDa molecular weight was used as a standard solute. Concentration of PEG in the feed as well as permeate stream was measured on the basis of Total Organic Carbon (TOC) content. TOC content was estimated using a TOC Analyzer (Shimadzu Corporation, Japan). Feed concentration was varied from 0.2 to 5 kg/m³.

Apparatus

Figure 1 shows a schematic diagram of the experimental setup. As shown in Figure 1, all the experiments were performed on a commercial ultra-filtration stirred cell unit (Cell Model No.: 8050; Amicon, U.S.). The specifications of the stirred cell unit are listed below:

Area of membrane:	$3.2 \times 10^{-3} \text{ m}^2$
Diameter of cell:	0.063 m
Operating pressure:	517 kPa (75 psi)
Feed volume:	$1.6 \times 10^{-4} \text{ m}^3$ (160 mL)

For each experimental run, the feed chamber was purged with nitrogen. The membrane was set up at the base of the feed chamber and permeate was collected at the bottom of

Figure 1. A schematic diagram of the experimental set-up.

the cell unit through a 1.5 mm diameter tube. Membrane flux was measured by recording the change in weight of a standard beaker over time. In order to determine the transient flux through the membrane, permeate weight was recorded continuously using a standard weighing balance (SARTORIUS Model No.: BP221S). The serial port communication program (RS-232) from the weighing balance was developed using LabVIEW software. A static pressure transducer (Ashcroft, Model No.: K5) was installed on the feed side of the cell. The analog output of a static pressure transducer was digitized using a National Instrument PCI 6023E A/D card and a CB-68LP connecting block. A data acquisition system in LabVIEW was developed to synchronize the pressure and mass measurements. The rate of change in flux and pressure were also plotted simultaneously in real-time with the developed software to monitor the transient response.

Experimental Procedure

1. Initially Pure Water Permeation (PWP) was measured for a new membrane. The mass of permeated solvent was recorded every two seconds through the data acquisition software LabVIEW. An error of less than 0.1% in measuring solvent flux was observed.
2. The filtration resistance for pure water was calculated using Equation (1).
3. For a given feed solution, flux versus time data was collected at a constant feed pressure until a steady state was reached, i.e. when the flux values became time independent. Once a steady state was achieved, the bypass valve (item 9 in Figure 1) was opened a crack to let the feed pressure decay as a function of time.
4. The unsteady state filtration data was continuously recorded using a data acquisition system.

Results and Discussions

Figure 2 shows the permeate flux as a function of time for three different (constant) feed pressures (135, 270 and 405 kPa) for a bulk feed concentration of 1 kg/m³. As expected, there is a sharp drop in the permeate flux from the initial value (at time $t = 0$, corresponding to the flux of pure water) for short filtration times. At longer filtration times, the flux decline gradually reduces and eventually attains steady state. It should be noted that during the time required to attain steady state

Figure 2. Variation of permeate flux as a function of time for various values of feed pressure (bulk concentration of PEG = 1 kg/m³).

Table 1. Steady state PEG rejection for the entire pressure and concentration range studied in this work.

ΔP , (kPa)	C_b , (kg/m ³)		
	0.2	1	5
135	99.7	99.6	99.5
270	99.1	98.5	98.3
405	97.9	97.5	97.2

Figure 3. Steady state permeate flux as a function of feed pressure for different feed concentrations of PEG.

the change in the bulk feed concentration was negligible. Only about 20–25 mL of the initial feed volume of 150 mL was collected as the cumulative permeate during these experiments.

Table 1 shows the steady state membrane rejection (R) based on the bulk feed concentration ($R = 1 - C_p/C_b$ where C_p and C_b are respectively the solute concentration in permeate and feed) for the entire concentration and pressure ranges studied in this work. As can be seen from this table, steady state solute rejection was always greater than 97%. The membrane can therefore be treated as being completely retentive to the solute. As a consequence, the osmotic pressure of permeate can be neglected and the effective osmotic pressure difference across the membrane ($\Delta\pi$) can be set equal to π_w .

Figure 3 shows the steady state flux for a solute concentration of 0.2, 1 and 5 kg/m³ as a function of feed pressure. It is clear that permeate flux becomes independent of pressure for a feed concentration of 5 kg/m³. For a feed concentration of 1 kg/m³, permeate flux is only slightly pressure dependent. The pressure range was restricted to 400 kPa due to the limitations of the test cell. After steady state was reached at a constant feed pressure, the bypass valve (see Figure 1) was opened a crack. This resulted in a decay of the feed pressure (pressure reduction) as a function of time. The unsteady permeate flux and feed pressure was recorded automatically using a data acquisition system as discussed in the experimental section.

Figure 4 shows the variation of the unsteady state permeate flux as a function of reducing feed pressure achieved by opening the bypass valve a crack once steady state had

Figure 4. Post steady state transient variation of permeate flux as a function of reducing feed pressure for three different initial pressures (bulk concentration of PEG = 0.2 and 1 kg/m³).

Figure 5. Post steady state transient variation of permeate flux as a function of reducing feed pressure for three different initial pressures (bulk concentration of PEG = 5 kg/m³).

been reached for a feed concentration of 0.2 and 1 kg/m³. A similar plot for 5 kg/m³ is shown in Figure 5. These figures are remarkable because they clearly show the contribution of the polarization layer to the overall filtration resistance. It should be noted that as the pressure decays from a higher initial value, the transient flux at any given pressure is slightly lower than the flux at the same pressure when the pressure decay is initiated from a lower initial value. For example in Figure 4, if a straight line is drawn vertically at 100 kPa, the flux resulting from a pressure decay starting from 405 kPa is slightly lower than that resulting from a pressure decay starting at 270 kPa, which in turn is lower than the flux at 100 kPa resulting from a pressure decay initiated at 135 kPa. This observation could be explained as follows: In the case where steady state is reached at a higher pressure, the amount of solute accumulated in the polarization layer is larger. This has resulted in higher filtration resistance to the solvent (permeate) flow. Consequently, when the feed pressure is rapidly reduced from a high initial value, the polarization layer offers a higher resistance to the permeate flow, resulting in a slightly lower flux than if the pressure decay was initiated from a lower initial value.

Furthermore, Figure 5 shows that irrespective of the initial value of the pressure, once the bypass valve is cracked open

Figure 6. A plot of dJ_s/dt versus dP/dt for data presented in Figure 5.

to reduce the feed pressure, the balance registers a loss in weight once the feed pressure drops to less than about 90 kPa. The only reason the balance could register a loss in weight (a negative flux) is because the feed pressure drops to a value less than the osmotic pressure corresponding to the accumulated solute at the membrane surface. This is indeed remarkable. To the best of our knowledge, this is the first time that such a phenomenon of negative flux has been demonstrated experimentally.

The transient data presented in Figure 5 can be further analyzed by plotting the time derivative of the unsteady flux (dJ_s/dt) versus the time derivative of the feed pressure (dP/dt). This data is shown in Figure 6. It is obvious that there is a very good linear correlation for each value of starting pressure. Using the slope values of these straight lines the filtration resistance (R_f) for each of the three starting pressures, 135, 270 and 405 kPa was 1.84, 2.95 and 4.30 $\times 10^{14}$ m⁻¹, respectively. The resistance for pure water was measured to be 3.75 $\times 10^{13}$ m⁻¹. Utilizing this estimated filtration resistance and Equation (2) the osmotic pressure (π_w) of the solute accumulated at the membrane surface was calculated as a function of the unsteady pressure. The osmotic pressure at the membrane surface is found to be independent of the feed pressure and it clearly indicates the gel layer dominated filtration. Considering that the osmotic pressure is independent of applied pressure and is only dependent on solute concentration, increased resistance at a higher applied pressure was due to an increase in gel layer thickness on the membrane surface. Further, it should be noted that using Equation (2), the osmotic pressure of the PEG gel was found to be about 87 $\pm$ 3 kPa. This value is in very good agreement with the experimental evidence of the onset of the negative flux when the feed pressure drops to values below 90 kPa as shown in Figure 5.

A similar analysis of the data presented in Figure 4 results in Figure 7 (for a PEG concentration of 1 kg/m³). As can be seen, the graph between dJ_s/dt versus dP/dt is no longer linear, which clearly indicates that the solute concentration (or osmotic pressure) at the membrane surface varies as a function of the feed pressure. The filtration resistance at different feed pressures can be calculated by drawing tangents to the curves in Figure 8. Using these resistance values at each feed pressure, the osmotic pressure of the solute at the membrane surface (or π_w) can be calculated using Equation (2). The results are shown in Figure 8. It is clear from this figure that π_w is a function of the feed pressure and is always less than 90 kPa, indicating an

Figure 7. A plot of dJ_s/dt versus dP/dt for data presented in Figure 4.

Figure 8. PEG osmotic pressure at the membrane surface (π_w) for a PEG bulk concentration of 1 kg/m^3 calculated as detailed in the text.

almost osmotically limited permeate flow. However, at higher applied pressures the contributions of the gel layer towards controlling permeate flow will play a prominent role.

It is instructive to calculate the steady state solute concentration profile in the polarization layer. A correlation between the solute osmotic pressure and its concentration correlation has been provided by Bhattacharjee et al. (1996). They demonstrated that Flory's equation for the change in chemical potential (the osmotic pressure) of PEG, as a function of volume fraction was an accurate correlation for osmotic pressure and concentration of PEG. Flory's equation relates the osmotic pressure with a solute volume fraction as

$$\pi = -\frac{RT}{V_1} \left[\ln(1 - \gamma_p) + \left(1 - \frac{1}{n}\right) \gamma_p + \chi_{12} \gamma_p \right] \quad (5)$$

where, $\gamma_p = c/\rho_p$ is the volume fraction of polymer, c is the polymer concentration, ρ_p is the polymer density and n is the number of monomer sub units in the polymer chain. The Flory-Huggins interaction parameter χ_{12} is a linear function of the solute concentration for linear chain polymers, and the concentration dependence is best expressed as

Figure 9. Variation of steady state PEG concentration as a function of distance from the membrane surface for three different feed pressures (bulk PEG concentration = 5 kg/m^3).

Figure 10. Calculated thickness of the polarization layer (δ) as a function of bulk PEG concentrations.

$$\ln|\chi_{12}| = p + qC \quad (5a)$$

where p and q are constants having values of -0.604 and 1.7452 respectively.

Equation (5) can be used to calculate C_w (the solute wall concentration) from the estimated π_w values from experimental data. Using this value of C_w and the steady state flux, J_s , the solute concentration profile as a function of distance perpendicular to the membrane surface can be estimated using the film theory (Zydney, 1997; van den Berg et al., 1989; Bhattacharjee and Datta, 2001) and the following equation:

$$C_x = C_b + (C_w - C_b) \exp\left(\frac{-J_s x}{D}\right) \quad (6)$$

where D is diffusivity and is calculated using literature correlation (Bhattacharjee and Datta, 2001). The boundary layer thickness (δ) can then be estimated as that value of x at which the solute concentration becomes almost equal to the bulk solute concentration.

Figure 9 shows the steady state solute concentration profile within the polarization layer for three different steady feed

pressures and a bulk feed concentration of 5 kg/m³. From this figure, it can be seen that for a given bulk feed concentration, the extent of the polarization layer is independent of the feed pressure. The extent of the polarization layer for each value of bulk feed concentration is plotted in Figure 10. This clearly shows that the extent of the polarization layer is a function of the bulk solute concentration and that the makeup of the polarization layer could be such as to provide significant hydraulic resistance compared to the membrane resistance. It is therefore necessary to account for the additional filtration resistance of the polarization layer when calculating the overall filtration resistance. Application of this analysis will be limited to those gel layers that are not sufficiently diffused and offer significant resistance to flow of solvent.

Conclusions

In this work we present a method to unequivocally determine whether the pressure independent filtration regime is osmotically limited or gel layer controlled. This is done by the analysis of unsteady state filtration data measured after a steady state had been reached at constant feed pressure.

The method of analysis has been validated by the experimental observation of a negative flux induced as a result of gel layer controlled filtration when the feed pressure is reduced to a value less than the osmotic pressure of the gel.

It was shown that using the method proposed in this work, it is possible to estimate the filtration resistance offered by the polarization layer in UF of macromolecular solutions. It is shown that the thickness of the polarization layer in such situations can be significantly large. Further, it was shown that the thickness of the polarization layer is a function of the bulk solute concentration and a higher bulk concentration led to a thicker polarization layer.

Nomenclature

C	solute concentration, (kg/m ³)
D	diffusivity, (m ² /s)
J_w	water flux, (m ³ /m ² .s)
J_s	solute flux, (m ³ /m ² .s)
k	mass transfer coefficient, (m/s)
n	number of monomer subunits in polymer chain
PEG	polyethylene glycol
R	universal gas constant, (J/mol.K)
R_f	filtration resistance, (m ⁻¹)
T	temperature, (K)
t	time, (s)
V_1	molar volume of solvent, (m ³ /mol)
x	distance from membrane or gel surface, (m)
ΔP	trans-membrane pressure, (kPa)

Greek Symbols

γ_p	volume fraction of polymer
χ_{12}	Flory-Huggins interaction parameter
π	osmotic pressure of solute, (kPa)
ρ	density of polymer or solution, (kg/m ³)
δ	thickness of polarization layer, (m)
μ	solvent viscosity, (kg/m.s)

Subscripts and Superscripts

b	bulk
w	wall
p	polymer

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