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## THE EFFECT OF CONCENTRATION DEPENDENT VISCOSITY ON PERMEATE FLUX LIMITATION IN ULTRAFILTRATION

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**Abstract.** *Ultrafiltration of macromolecular solutions was analyzed by using the osmotic-pressure model in which the influence of the solute concentration on the mass-transfer coefficient is taken into account. A mathematical analysis of the model clearly shows that an increase of the solution viscosity with the solute concentration plays the main role in the existence of a limiting permeate flux at a finite pressure difference. The membrane resistance has no effect on the limiting permeate flux except on the value of the pressure difference necessary to reach the limiting flux. In order to define the  $J_s$  vs.  $\Delta p$  plots, a system of equations was derived, the parameters of which are to be experimentally determined. A simulation results are presented for the ultrafiltration of whey protein solutions in recirculating UF units under turbulent flow.*

**Key words:** *ultrafiltration, flux limitation, osmotic-pressure model, whey proteins, mass-transfer coefficient.*

### INTRODUCTION

The major problem in ultrafiltration applications is the limitation of the permeate flux to far below the pure solvent flux under the same pressure difference [1]. This limitation is a consequence of the accumulation of retained solutes at the surface of the membrane. During ultrafiltration of macromolecular solutions this increased solute concentration causes a rise in the osmotic pressure which partially cancels the applied pressure difference. The aim of this paper is to examine the main features of the modified osmotic-pressure model based on the assumption that the mass-transfer coefficient depends on the interfacial solute concentration.

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## THEORY

The stationary permeate flux  $J_s$  through a totally retentive UF membrane ( $C_p = 0$ ) is deduced from the film theory [2, 3]:

$$J_s = K \ln \left( \frac{C_m}{C_b} \right) \quad (1)$$

where  $C_m$  and  $C_b$  are the solute concentration at the surface of the membrane and in the bulk solution, respectively, and  $K$  is the mass-transfer coefficient (Fig. 1).

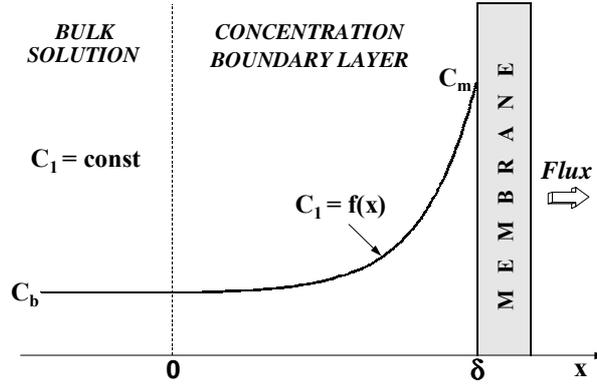


Fig. 1. Boundary layer formation in ultrafiltration.

Thus,  $C_m$  increases with increasing  $J_s$  and the limiting permeate flux is reached at the limiting solute concentration at the membrane surface,  $C_{ml}$ :

$$J_l = K \ln \left( \frac{C_{ml}}{C_b} \right) \quad (2)$$

Since  $C_{ml}$  and  $C_b$  are constants for a given solution, the limiting flux  $J_l$  can only be increased by increasing  $K$ . The mass-transfer coefficient  $K$  can be estimated from correlation:

$$Sh = A' Sc^x Re^y (d_h/L)^z \quad (3)$$

where  $A'$  is a constant, depending only on the module geometry and flow regime (laminar or turbulent),  $d_h$  is the hydraulic mean diameter of the feed channel,  $L$  is the length of the feed channel,  $Sh = Kd_h/D$ ,  $Sc = \eta/(D\rho)$ ,  $Re = vd_h\rho/\eta$  are the Sherwood, Schmidt and Reynolds number, respectively,  $v$  is the average velocity of the feed solution, and  $\rho$ ,  $\eta$ , and  $D$  are the density, dynamic viscosity, and solute diffusivity of the solution at the membrane surface. We can thus write:

$$K = Av^y D^{(1-x)} \rho^{(y-x)} \eta^{(x-y)} \quad (4)$$

where  $A = A'd_h^{(y+z-1)}L^{-z}$  is a constant for any given module geometry and flow regime.

According to the osmotic-pressure model [4-9] the permeate flux is given by:

$$J_s = \frac{\Delta p - \Delta \Pi_m}{R_m} \quad (5)$$

where  $R_m$  is the membrane resistance,  $\Delta p$  is the applied pressure difference, and  $\Delta \Pi_m$  is the difference between the osmotic pressure of the solution at the membrane surface  $\Pi_m$  and the osmotic pressure of the permeate  $\Pi_p$ . For a totally retentive membrane ( $C_p = 0$ , i.e.  $\Pi_p = 0$ ):

$$\Delta \Pi_m = \Pi_m - \Pi_p = \Pi_m = a_1 C_m + a_2 C_m^2 + a_3 C_m^3 = \sum_{i=1}^3 a_i C_m^i \quad (6)$$

From Eq. (1), (5), and (6) we obtain an expression for the applied pressure difference that is necessary to reach a given steady-state permeate flux

$$\Delta p = J_s R_m + \sum_{i=1}^3 a_i C_m^i = J_s R_m + \sum_{i=1}^3 a_i C_b^i \exp(i J_s / K) \quad (7)$$

The slope of  $\Delta p$  vs.  $J_s$  curve at each point is given by

$$\partial \Delta p / \partial J_s = R_m + [1/K - (J_s / K^2)(\partial K / \partial J_s)] \sum_{i=1}^3 i a_i C_b^i \exp(i J_s / K) \quad (8)$$

Using an analogy with electrical circuits, Aimar and Sanchez [10] called this quantity 'mass-transfer impedance' i.e. overall mass-transfer resistance. According to Eq. (8) the impedance is the sum of two terms: a hydraulic one  $R_m$  that is passive since it is independent of the permeate flux, and a physicochemical one that is reactive since it depends on the permeate flux. According to Eq. (1) we can express the derivative  $\partial K / \partial J_s$  as follows:

$$\partial K / \partial J_s = (\partial K / \partial C_m)(\partial C_m / \partial J_s) = (\partial K / \partial C_m)(C_m / K)[1 + C_m(\partial K / \partial C_m)(J_s / K^2)] \quad (9)$$

Substituting Eq. (9) into Eq. (8) we obtain

$$\frac{\partial \Delta p}{\partial J_s} = I + II + III = R_m + \frac{1}{K} \sum_{i=1}^3 i a_i C_m^i - \frac{(\partial K / \partial C_m) \ln(C_m / C_b)}{K^2 [1 + (C_m / K)(\partial K / \partial C_m) \ln(C_m / C_b)]} \sum_{i=1}^3 i a_i C_m^{i+1} \quad (10)$$

The mass-transfer impedance  $\partial \Delta p / \partial J_s$  can only be infinite, for a finite value of  $C_m$ , if the denominator of the third right-hand term of Eq. (10) is equal to zero, i.e. if the following equation is true [10-12]:

$$\frac{1}{K} \frac{\partial K}{\partial C_m} = - \frac{1}{C_m \ln(C_m / C_b)} \quad (11)$$

Eq. (11) is the condition, which must be satisfied for the limiting flux to be reached at a finite pressure difference. Eq. (11) clearly shows that in this case  $K$  is a decreasing function of  $C_m$ . The logarithmic derivative of Eq. (4) with respect to  $C_m$  is as follows

$$\frac{1}{K} \frac{\partial K}{\partial C_m} = \frac{1-x}{D} \frac{\partial D}{\partial C_m} + \frac{y-x}{\rho} \frac{\partial \rho}{\partial C_m} + \frac{x-y}{\eta} \frac{\partial \eta}{\partial C_m} \quad (12)$$

Densities of macromolecular solutions vary slightly with the solute concentration, and the importance of the diffusional term is generally much lower than of the viscosity term, especially for the high concentrations encountered in the boundary layer [10]. We can thus reduce the infinite impedance condition, Eq. (11), to the expression

$$\frac{x-y}{\eta} \frac{\partial \eta}{\partial C_m} = -\frac{1}{C_m \ln(C_m/C_b)} \quad (13)$$

Eq. (13) allows an analytical solution when the variations of the solution viscosity  $\eta$  with the solute concentration  $C$  take the following form:

$$\eta = \eta_0 + \eta_1 C^m \quad (14)$$

where  $\eta_0$  is the solvent viscosity, which can be neglected for the high  $C$  values. Therefore, the solution viscosity  $\eta$  at the membrane surface is given by  $\eta_1 C_m^m$ , from which one obtains

$$\frac{\partial \eta}{\partial C_m} = m \eta_1 C_m^{m-1} = \frac{m \eta}{C_m} \quad (15)$$

Substitution of Eq. (15) into Eq. (13) and rearrangement gives

$$\frac{1}{\ln(C_m/C_b)} = -m(x-y) = n \quad (16)$$

where  $n = m(y-x)$ . Eq. (16) applies to limiting flux conditions and accordingly,  $C_m$  from this equation can be replaced by the limiting solute concentration at the membrane surface  $C_{ml}$ :

$$\ln \frac{C_{ml}}{C_b} = \frac{1}{n} \quad (17)$$

i.e.

$$C_{ml} = C_b \exp\left(\frac{1}{n}\right) \quad (18)$$

The solution viscosity at the membrane surface is given by  $\eta_1 C_{ml}^m$ . The substitution of this equation and Eq. (18) into Eq. (4) gives the mass-transfer coefficient at the limiting flux:

$$K = A v^y D^{(1-x)} \rho^{(y-x)} \eta_1^{(x-y)} C_{ml}^{-n} = B v^y C_b^{-n} / e \quad (19)$$

The relationship for the limiting permeate flux  $J_1$  is obtained by substituting Eqs. (17) and (19) into Eq. (2)

$$J_1 = B v^y C_b^{-n} / (en) \quad (20)$$

where  $B = A D^{(1-x)} \rho^{(y-x)} \eta_1^{(x-y)}$ . Eq. (20) clearly shows that the limiting flux is independent of the membrane resistance, as confirmed experimentally by Michaels [2] and Porter [3], and theoretically by Do and Elhassadi [13]. Eq. (20) also predicts that the logarithm of the limiting flux  $J_1$  is a linear function of the logarithm of the bulk concentration  $C_b$ , with a

slope equal to (-n).

Combining Eq. (7), (18), and (20) leads to an expression giving the applied pressure difference necessary to reach the limiting flux:

$$\Delta p_1 = R_m B v^y C_b^{-n} / (en) + \sum_{i=1}^3 a_i C_b^i \exp(i/n) \quad (21)$$

Eq. (21) suggests that this so-called threshold pressure  $\Delta p_1$  increases with  $R_m$ , as confirmed by Vladislavjević et al. [14,15] within the framework of the gel theory.

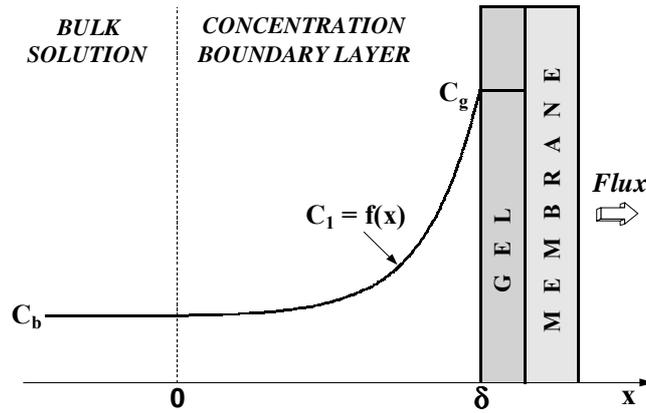


Fig. 2. Concentration polarization with gel formation.

If gelation of the interfacial solution occurs at a solute concentration  $C_g$  lower than  $C_{ml}$  (Fig. 2), then the limiting permeate flux is given by

$$J_1 = B v^y C_g^{-n} \ln(C_g / C_b) \quad (22)$$

In the section below we will discuss the main features of the presented model taking ultrafiltration of whey proteins as an example. Whey created as a by-product of cheese and casein manufacture contains 0.6-0.8 wt% proteins ( $\alpha$  - lactalbumin and  $\beta$  - lactoglobulin) [16]. Whey protein concentrates (WPCs) produced by ultrafiltration are used as additives in dairy, bakery, and confectionery products, soft drinks, infant formula, and specialty dietary foods.

## RESULTS AND DISCUSSION

The use of presented model requires a knowledge of some parameters, which will be either arbitrary chosen or taken from previously published papers:

$B = 2.1 \times 10^{-5}$	According to Aimar and Sanchez [10];
$y = 0.875$	From Deissler equation (e.g., Wijmans et al. [17]);
$n = 0.44$	As suggested by Aimar and Sanchez [10];
$R_m = 5 \times 10^{10}$ Pas/m	A typical value of the membrane resistance.

The values of virial coefficients  $a_i$  in the osmotic pressure relationship (Eq. 6) will be taken from experiments by Jonsson [18]:  $a_1 = 4400$ ;  $a_2 = -17$ ;  $a_3 = 7.9$ , where  $C_m$  is to be replaced in wt% for  $\Pi_m$  to be obtained in Pa.

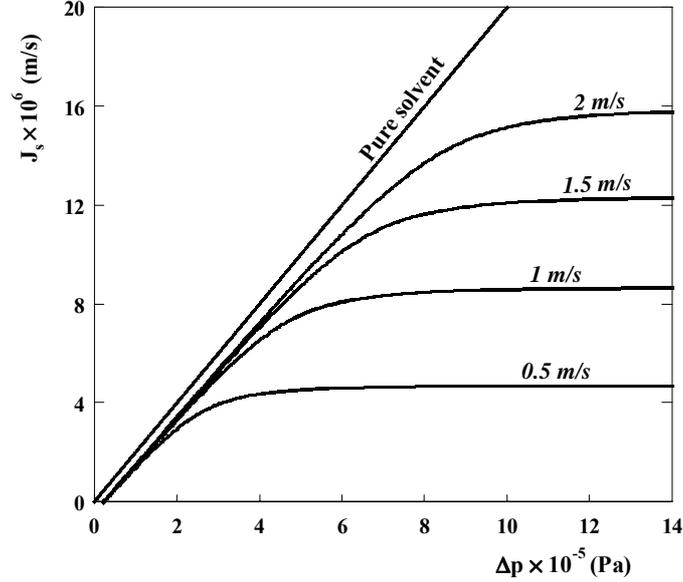


Fig. 3. Stationary permeate flux as a function of applied pressure difference for different feed flow velocities ( $C_b = 5$  wt%).

The stationary permeate flux is plotted against the pressure difference in Figs. 3 and 4. For small pressure differences (when  $\partial\Delta p/\partial J_s \approx R_m$ ), the permeate flux is a linear function of the pressure difference and virtually independent on the bulk solute concentration or feed flow velocity. This is a pure filtration or Darcy's law region where the permeate flux is limited by the membrane permeability. The intercept on the abscissa of each  $J_s$  vs.  $\Delta p$  curve corresponds to the osmotic pressure of the feed solution. As the pressure difference is increased the flux reaches a limiting value  $J_l$ , beyond which a further increase in the pressure does not produce any increase in the steady-state flux. This is an ultrafiltration region where the permeate flux is independent of the membrane resistance and limited by the mass-transfer conditions in the boundary layer. The limiting flux increases with increasing flow velocity and with decreasing bulk concentration, as predicted by Eq. (20). The pressure difference necessary to reach the limiting flux also increases with increasing flow velocity, as suggested by Eq. (21). The pure solvent flux is proportional to the pressure difference and the proportionality constant is  $R_m^{-1}$ .

Fig. 5 illustrates the effect of the applied pressure difference  $\Delta p$  on the mass-transfer coefficient  $K$  and the solute concentration at the membrane surface  $C_m$ . The mass-transfer coefficient decreases continuously with  $C_m$ , as predicted by Eq. (11). Observing the shape of the curve representing  $C_m$  vs.  $\Delta p$ , we note a point of inflection (A), whose coordinates can be deduced from an equation:  $\partial\Delta p/\partial C_m^2 = 0$ , from which one obtains

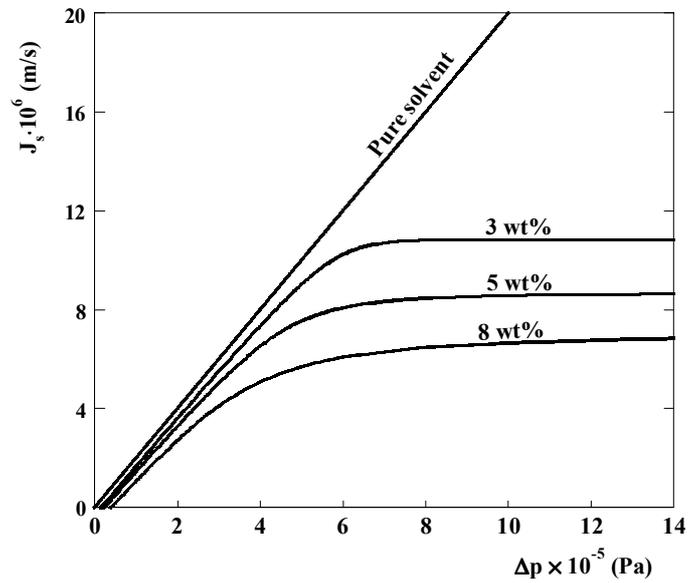


Fig. 4. Stationary permeate flux as a function of applied pressure difference for different feed concentrations ( $v = 1$  m/s).

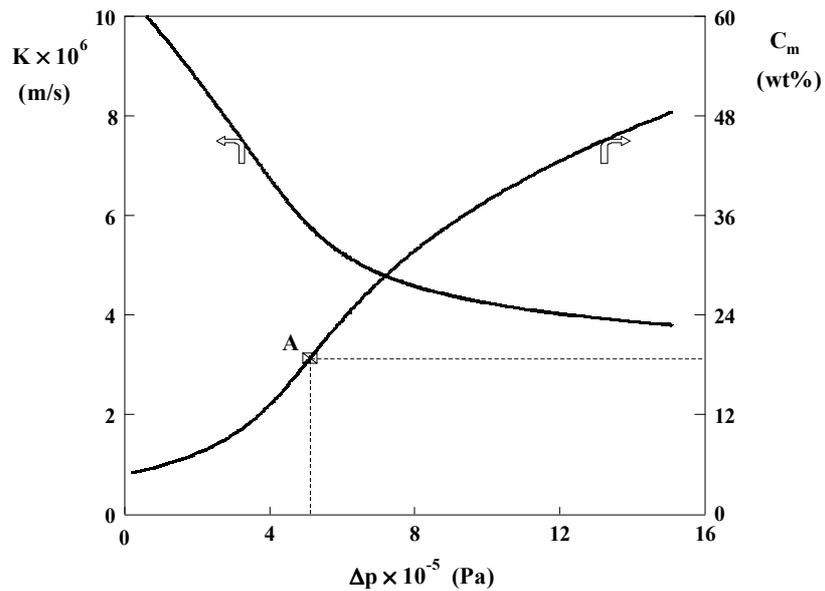


Fig. 5. Effect of pressure difference on mass-transfer coefficient and interfacial solute concentration at  $v = 1$  m/s and  $C_b = 5$  wt%.

$$R_m B v^y C_m^{-n} = \frac{2a_2 C_m^2 + 6a_3 C_m^3}{2n + 1 - n(n+1) \ln(C_m/C_b)} \quad (23)$$

i.e.,  $C_m = 18.76$  wt% and  $\Delta p = 5.11 \times 10^5$  Pa for the conditions as in Fig. 5.

It appears from Fig. 5 that  $C_m$  increases strongly when the stationary permeate flux approaches the limiting value. For an example, if the operating conditions are such that the permeate flux is only 10% below  $J_l$ , the solute concentration at the membrane surface may be reduced by 60% as compared with  $C_{m1}$  (Table 1). Since the membrane fouling phenomena, such as solute adsorption on the membrane surface [9], pore plugging or blocking by solutes [19] or gelation of the interfacial solution [1,2,20] are more pronounced at the higher solute concentrations, it would lead to a much slower fouling kinetics.

Table 1. Mass-transfer calculations at  $v = 1$  m/s and  $C_b = 5$  wt%  
( $B = 2.1 \times 10^{-5}$ ;  $y = 0.875$ ;  $n = 0.44$ ;  $R_m = 5 \times 10^{10}$  Pas/m).

$C_m$ wt%	$K \times 10^6$ m/s	$J_s \times 10^6$ m/s	$\Delta p \times 10^{-5}$ Pa	$I \times 10^{-10}$ Pas/m	$II \times 10^{-10}$ Pas/m	$III \times 10^{-10}$ Pas/m
5	10.34	0	0.2256	5	0.2331	0
10	7.625	5.285	3.144	5	0.8433	0.3701
15	6.379	7.008	4.392	5	2.169	2.029
20	5.620	7.792	5.340	5	4.697	7.346
25	5.095	8.200	6.328	5	9.010	21.86
30	4.702	8.425	7.512	5	15.77	58.73
35	4.394	8.550	8.994	5	25.68	152.9
40	4.143	8.615	10.85	5	39.55	425.5
45	3.934	8.643	13.16	5	58.18	1693
48.53	3.805	8.648	15.09	5	74.69	$\infty$

In Fig. 6 the different terms composing the transfer impedance (Eq. 10) are plotted as a function of the interfacial concentration  $C_m$ . This plot is obtained using data from Table 1. When the interfacial concentration increases from  $C_b$  to the limiting value  $C_{ml}$ , the third right-hand term of Eq. (10) increases from zero to infinity. The condition to be satisfied for the hydraulic term (I) to be greater than the physicochemical term (II+III) is as follows:

$$R_m K > \frac{\sum_{i=1}^3 i a_i C_m^i}{1 - n [\ln(C_m/C_b)]} \quad (24)$$

For the conditions as in Fig. 6 and Table 1, it is valid for  $C_m < 18.40$  wt%. It should be noted that the  $C_m$  value for which the two resistances are equal is similar to the  $C_m$  value in the point of inflection of  $C_m$  vs.  $\Delta p$  curve. The  $C_m$  value for which the second right-hand term of Eq. (10) is equal to the third right-hand term is given by  $C_b \exp(1/2n) = 15.58$  wt%.

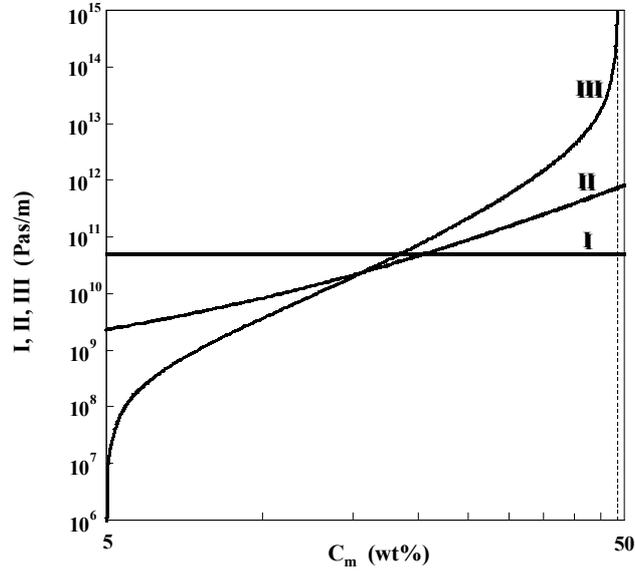


Fig. 6. Different terms composing mass-transfer impedance versus interfacial solute concentration at  $v = 1$  m/s and  $C_b = 5$  wt%.

#### CONCLUSION

The osmotic-pressure model employed here to examine ultrafiltration of whey protein solutions is based on an approach proposed by Aimar and Sanchez [10], in which the mass-transfer coefficient is considered as a decreasing function of the solute concentration at the membrane surface. It was shown that when the pressure difference increases from zero to  $\Delta p_i$ , the permeate flux is at first limited by the membrane resistance and then by the osmotic pressure of the interfacial solution, but only a decrease in the mass-transfer coefficient can explain the existence of a real limiting flux at a finite pressure difference. Both  $J_l$  and  $C_{ml}$  increase with increasing flow velocity and with decreasing bulk solute concentration, but are independent of the membrane resistance, which influences only the applied pressure difference necessary to reach the limiting flux. When the permeate flux approaches the limiting value, we noticed a drastic increase of the solute concentration at the membrane surface, whose consequence is to enhance membrane fouling phenomena such as protein adsorption or gelation on the surface of the membrane.

#### LIST OF SYMBOLS

A	constant in Eq. (4)
A'	constant in Eq. (3)
$a_i$	virial coefficients in in the osmotic pressure relationship (Eq. 6)
B	constant in Eq. (20)
C	solute concentration, wt%

$C_b$	solute concentration in bulk solution, wt%
$C_g$	solute concentration at which gelation of interfacial solution occurs, wt%
$C_m$	solute concentration at the surface of the membrane, wt%
$C_{ml}$	interfacial solute concentration corresponding to the limiting flux, wt%
$C_p$	solute concentration in permeate, wt%
$C_1$	local solute concentration in boundary layer, wt%
$D$	diffusion coefficient of solute, m <sup>2</sup> /s
$d_h$	hydraulic mean diameter of feed channel, m
$e$	2.71828...
I, II, III	terms in relationship for mass-transfer impedance (Eq. 10), Pa s/m
$J_s$	stationary permeate flux, m/s
$J_l$	limiting permeate flux, m/s
$K$	mass-transfer coefficient, m/s
$L$	length of feed channel, m
$m$	exponent in Eq. (14)
$n$	constant in Eq. (16)
$\Delta p$	pressure difference, Pa
$\Delta p_l$	pressure difference necessary to reach the limiting flux, Pa
$\partial \Delta p / \partial J_s$	mass-transfer impedance, Pa s/m
$R_m$	hydraulic resistance of membrane, Pa s/m
$Re$	Reynolds number, (-)
$Sc$	Schmidt number, (-)
$Sh$	Sherwood number, (-)
$v$	average flow velocity of feed solution, m/s
$x, y, z$	exponents in correlation (3)
$\delta$	thickness of boundary layer, m
$\eta$	dynamic viscosity of solution, Pa s
$\eta_o$	dynamic viscosity of pure solvent, Pa s
$\eta_1$	constant in Eq. (14)
$\Pi_m$	osmotic pressure of interfacial solution, Pa
$\Pi_p$	osmotic pressure of permeate (for an ideal membrane $\Pi_p = 0$ ), Pa
$\Delta \Pi_m$	osmotic pressure difference between interfacial solution and permeate, Pa
$\rho$	density of solution, kg/m <sup>3</sup>

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## UTICAJ KONCENTRACIONE ZAVISNOSTI VISKOZITETA NA LIMITACIJU FLUKSA PERMEATA PRI ULTRAFILTRACIJI

**Goran T. Vladislavljević, Miloš B. Rajković**

*Ultrafiltracija rastvora makromolekula je proučavana primenom modela osmotskog pritiska u kome je uzet u obzir uticaj koncentracije rastvorka na koeficijent prenosa mase. Matematičkom analizom modela pokazano je da porast viskoziteta rastvora sa povećanjem koncentracije rastvorka igra glavnu ulogu u javljanju graničnog fluksa pri konačnoj razlici pritisaka. Otpor membrane ne utiče na granični fluks permeata, već samo na vrednost razlike pritisaka pri kojoj se uspostavlja granični fluks. Radi definisanja zavisnosti  $J_s$  od  $\Delta p$ , izveden je sistem jednačina čiji se parametri moraju odrediti eksperimentalno. Prikazani su rezultati numeričke simulacije procesa ultrafiltracije rastvora proteina surutke u protočnim aparatima sa recirkulacijom napojnog toka pri turbulentnom režimu strujanja.*

*Ključne reči: ultrafiltracija, limitacija fluksa, model osmotskog pritiska, proteini surutke, koeficijent prenosa mase*