Mass Transfer Coefficient of Tubular Ultrafiltration Membranes under High-Flux Conditions

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The effect of suction flow on the mass transfer coefficient of tubular ultrafiltration membranes, in particular that under a high-flux condition, was studied. We pointed out that N_{Sh} is proportional to ${N_{Re}}^{0.875}{N_{Sc}}^{0.25}$ under turbulent conditions, and that the proportional constant, b, exceeds 0.023 when the effect of suction flow is not negligible. We conducted the velocity variation method using ultrafiltration membranes with MWCOs of 20k and 100k and dextrans having molecular weights of 40,000 and 70,000 at the conditions, where N_{Re} exceeded 3.6×10^3. We demonstrated that the effect of suction flow includes not only flux but also the diffusion coefficient of solute, and that the ratio of the flux to the diffusion coefficient, expressed as N_{Pe_w}, is an important index. Finally, we concluded that $b=0.023$, when N_{Pe_w} is smaller than 2.23×10^3, giving the Deissler equation itself, and that $b=2.04\times10^{-6}\times N_{Pe_w}^{1.21}$, when N_{Pe_w} exceeds 2.23×10^3. © 2017 American Institute of Chemical Engineers *AIChE J*, 64: 1778–1782, 2018

Keywords: mass transfer coefficient, wall Peclet number, suction, flux, diffusion coefficient

Introduction

Transport equations for reverse osmosis and ultrafiltration are essential in evaluating intrinsic membrane performances and in developing practical processes. On the basis of non-equilibrium thermodynamics, transport equations were first formulated by Kedem and Katchlsky[1] and then modified by Spiegler and Kedem[2] as

$$J_V=L_p(\Delta P-\sigma\Delta\pi) \tag{1}$$

$$R=\frac{(1-F)\sigma}{1-\sigma F} \tag{2}$$

$$F=\exp\left(-\frac{(1-\sigma)J_V}{P}\right) \tag{3}$$

where J_V is the volume flux of pure water, ΔP is the pressure difference, $\Delta\pi$ is the osmotic pressure difference, R is the real objection, L_p is the pure water permeability, P is the solute permeability, and σ is the reflection coefficient. The latter three of these parameters in the transport equations characterize the intrinsic membrane performances, and pore structures can be also discussed in ultrafiltration using these three parameters[3]. L_p can be simply obtained in a pure-water permeation test in which the applied pressure difference is varied, while parameters P and σ can be obtained by curve fitting the relationship of R versus ${J_V}^{-1}$. We thus have to estimate R, which is independent of flow rate but dependent on pressure, in a proper manner to obtain P and σ.

In estimating the real rejection, which is calculated from the ratio of the concentrations of solute at the permeate (C_p) to that at the membrane surface (C_m) and expressed as R or R_{real} to distinguish it from the observed rejection, the concentration polarization should be considered. Rejection of the solute by the membrane surface results in a value of C_m higher than the concentration in the bulk (C_b). Although there are a few reports of the direct observation of particle deposition onto a membrane surface during microfiltration using a specially fabricated membrane housing with a microscope,[4–6] there is no feasible and widely accepted experimental method of measuring C_m directly, in particular when we deal with macromolecules or much smaller substances, and C_m is thus generally estimated using the concentration polarization model, which can be easily derived by applying the mass balance concept as

$$\frac{C_m-C_p}{C_b-C_p}=\exp\left(\frac{J_V}{k}\right) \tag{4}$$

where k is the mass transfer coefficient. This equation indicates that we need the value of k to estimate C_m in a proper manner.

The mass transfer coefficient can be estimated experimentally using the velocity variation (VV) method[3,7,8] or the osmotic pressure method.[9,10] In contrast, correlation equations have been established to predict mass transfer coefficients from experimental data. Among them, Sherwood relations are generally used.[11,12] The empirical formula used in the case of turbulent flow is

$$N_{Sh}=\frac{k\ d_h}{D}\propto {N_{Re}}^p{N_{Sc}}^q \tag{5}$$

where N_{Sh}, N_{Re}, and N_{Sc} are, respectively, the Sherwood number, Reynolds number, and Schmidt number, d_h and D

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are, respectively, the equivalent diameter and diffusion coefficient, and p and q are exponents. Among the empirical formulas, the Deissler equation, written as

$$N_{Sh}=0.023\ N_{Re}{}^{0.875}N_{Sc}{}^{0.25} \tag{6}$$

is often employed.[3,13] However, this correlation is not applicable for higher permeate flux.[14–17] Indeed, it is well known that the mass transfer coefficient predicted using the Deissler equation under a higher flux condition is underestimated, compared with the real coefficient obtained using the VV method. In such cases, $C_{\rm m}$ estimated using the Deissler equation often exceeds 100%, which is an unrealistic situation. This leads to an inaccurate evaluation of intrinsic membrane performances. Thus, the effect of suction flow, which has been attributed to permeate flux (in particular higher permeate flux) passing through the porous membrane, on the mass transfer coefficient should be more carefully considered.

Few reports on methods of predicting the mass transfer coefficient have considered the effect of suction due to permeate flux. De and coworkers developed Sherwood number relations for predicting the mass transfer coefficient with suction from first principles.[16,18] Adopting a basic approach of solving simultaneously the governing solute mass and momentum balance equations, they demonstrated general Sherwood number relations for crossflow reverse osmosis and ultrafiltration, including the effects of suction for different flow geometries; i.e., flows through a rectangular cell, tubular module, and radial crossflow cell. Herath and coworkers developed a modified Sherwood relation that considers the effect of suction flow, and obtained good agreement with estimations made using the VV method.[19] However, they only tested two viruses as model solutes having similar diffusion coefficients using one membrane to demonstrate the developed relation, and the proposed relation thus lacks versatility. Numerical simulations are another option for demonstrating the effect of suction,[20,21] but a simple modification of the exsiting theory, such as the Deissler equation, by adding the effect of suction flow to the mass transfer coefficient would be more attractive, in particular for membrane housings with simpler structures.

The present study employed the VV method to measure mass transfer coefficients using two dextrans with different molecular weights and two ultrafiltration membranes with different molecular weight cutoff (MWCO) values under various flux conditions. In particular, the study focused on mass transfer coefficients under higher flux conditions in which the effect of suction flow is not negligible, and the disparity between the mass transfer coefficients obtained experimentally with the VV method and those estimated using the Deissler equation is discussed in terms of the wall Peclet number, which is an index of the ratio of flux to the diffusion coefficient.

Theory

The rejection characteristics of membranes are discussed using the observed rejection, $R_{\rm obs}$, and the real rejection, $R_{\rm real}$:

$$R_{\rm obs}=\frac{C_{\rm b}-C_p}{C_b} \tag{7}$$

$$R_{\rm real}=\frac{C_{\rm m}-C_p}{C_m} \tag{8}$$

In the VV method, k is considered to be a function of the Reynolds number, and k is thus expressed as a function of the crossflow velocity, u:

$$k \propto u^a \tag{9}$$

Using Eqs. 7–9, the concentration polarization equation, given as Eq. 4, is rewritten as

$$\ln\left(\frac{1-R_{\rm obs}}{R_{\rm obs}}\right)=\ln\left(\frac{1-R_{\rm real}}{R_{\rm real}}\right)+\frac{1}{\gamma}\left(\frac{J_{\rm V}}{u^a}\right) \tag{10}$$

When we set a reasonable value for a, a linear relationship between $\ln\left(\frac{1-R_{obs}}{R_{obs}}\right)$ and $\frac{J_V}{u^a}$ is obtained. Here, we use 0.875 for the value a.[3,13] $R_{\rm real}$ can be obtained by extrapolating linear plots of $\ln\left(\frac{1-R_{obs}}{R_{obs}}\right)$ vs. $\frac{J_V}{u^a}$, and k is then obtained as

$$k=\gamma u^a \tag{11}$$

where γ is the slope of the same linear plots. Using this k value, we can calculate $N_{\rm sh}$ from Eq. 5. When a linear relationship between $\ln\left(\frac{1-R_{obs}}{R_{obs}}\right)$ and $\frac{J_V}{u^a}$ is obtained, we also obtain a linear relationship between N_{Sh} and $N_{Re}{}^{0.875}N_{Sc}{}^{0.25}$. This indicates that the coefficient in the Deissler equation, 0.023, varies when the effect of the suction flow is not negligible. In such cases, we can express the relationship between N_{Sh} and $N_{Re}{}^{0.875}N_{Sc}{}^{0.25}$, as

$$N_{\rm Sh}=b\ N_{\rm Re}{}^{0.875}N_{\rm Sc}{}^{0.25} \tag{12}$$

where b is the slope for the correlation and depends on the effect of the suction flow. The value b thus appears to be a function of $J_{\rm v}$. However, as the conentration polarization model tells us, the ratio of the flux to the diffusion coefficient needs to be reasonable to discuss the transport phenomena. The value b would be a function of the wall Peclet number, defined as

$$N_{\rm Pe_w}=\frac{J_{\rm V}\ d_{\rm h}}{D} \tag{13}$$

The diffusion coefficient, D, of dextran at 25°C is estimated from an equation for chainlike polymers, such as dextrans.[22,23]:

$$D=8.76\times10^{-9}\times M_{\rm w}{}^{-0.48} \tag{14}$$

According to the Stokes-Einstein equation, D is also expressed as a function of temperature, T:

$$\frac{\mu\ D}{T}=\text{constant} \tag{15}$$

where μ is the viscosity. The viscosities of all feed solutions tested are regarded to be the same as the viscosity of pure water at each temperature because the concentrations of the polymers in the feed were only 100 ppm in the present study.

Experimental

Two types of tubular ultrafiltration membranes with MWCOs of 20k and 100k (Daicen Membrane-Systems Ltd., Tokyo, Japan) were used. The inner diameter and length were 10 mm and 0.37 m, respectively, and the effective membrane area was 1.16×10^{-2} m^2. Some of the membranes were used after compaction at 1.0 MPa. Dextrans T40 and T70 (Pharmacosmos), having molecular weights of 40,000 and 70,000 respectively, were used as the solute. The temperature was fixed at 25 or 10°C. Table 1 summarizes four combinations used for the VV method in this study.

Figure 1 is a conceptual diagram of the experimental setup for the VV method. Two different membranes were set in

Table 1. Four Combinations of Solute, Membrane, and Temperature Employed for the VV Method

	Solute	MWCO	Temperature (°C)
A	Dextran T70	100k (after compaction)	25
B	Dextran T40	20 k	25
C	Dextran T40	100 k	25
D	Dextran T70	20 k	10

series, and the flow velocity was well developed inside the membranes. The feed concentration was fixed at 100 ppm in all experiments, and the osmotic pressure was negligible. The retentate and permeate were fully recycled to maintain a constant concentration of the feed during the tests. The concentrations of the dextran in the feed and permeate were measured with a total organic carbon analyzer (TOC-V$_{CSH}$; Shimadzu Corporation, Kyoto, Japan) to calculate R_{obs}. The flow rate was varied in the range 1.5–6.5 L min^{-1}. N_{Re} under each operation condition exceeded 3.6×10^3, and the experiments were thus carried out under turbulent conditions.

Results and Discussion

Mass transfer coefficient at lower and higher flux conditions

Two examples of the data sets obtained using the VV method at different fluxes are shown in Figures 2 and 3. The membrane used was that having an MWCO of 100k (after compaction), and the solute used was Dextran T70. The temperature of the feed was 25°C. As for the lower flux condition (example 1), the flux was 4.7×10^{-6} m^3 m^{-2} s^{-1} at ΔP of 0.02 MPa and the flow rate ranging from 2.0 to 5.0 L min^{-1}, and as for the higher flux condition (example 2) the flux was 2.0×10^{-5} m^3 m^{-2} s^{-1} at ΔP of 0.06 MPa and the flow rate ranging from 3.5 to 6.5 L min^{-1}. These flux values were independent of the flow rate. This is important for the VV method. Figure 3 shows the relationship between $\ln\left(\frac{1-R_{obs}}{R_{obs}}\right)$ and $\frac{J_V}{u^{0.875}}$. Good linear correlation is observed in both cases. These two lines give different R_{real} values even though the membrane and the solute used were the same in both cases. As described in the introduction, R_{real} is dependent on pressure, which is well-known and easily understood from Eqs. 1–3. Another reason is the difference in the membranes performances after compaction. Even though the same procedure was used for the compaction, the performance of the resultant membrane slightly differs one by one. However, this is not matter at all because we deal with the mass transport phenomena outside the membranes, not the mass transport phenomena inside the pores of membranes. Thus, we estimated the mass transfer coefficients, which were a function of u, in both cases from the data.

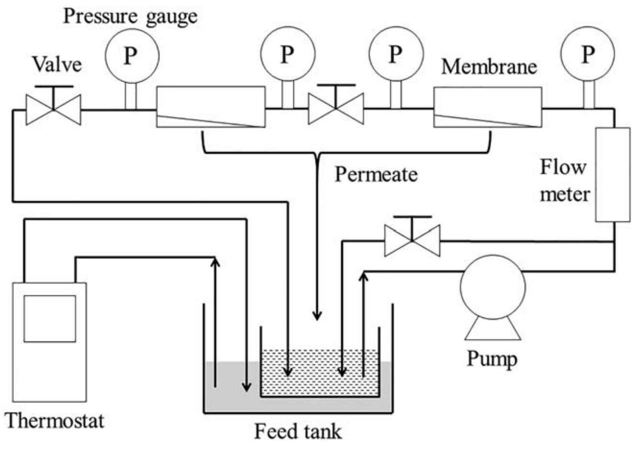

Figure 1. Conceptual diagram of the experimental setup for the VV method.

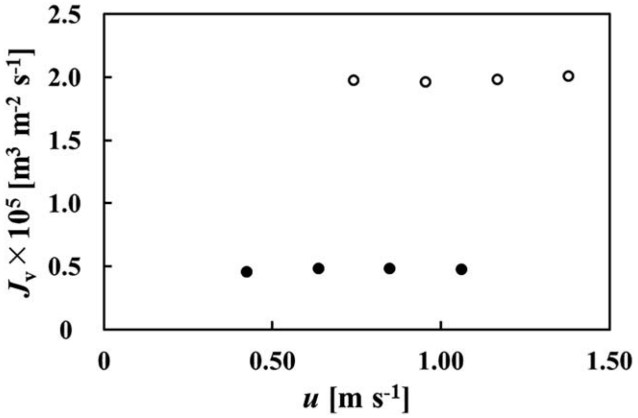

Figure 2. Relationships between flux and the cross-flow. Closed symbols: example 1 (ΔP: 0.02 MPa, Flow rate: 2.0–5.0 L min^{-1}), open symbols: example 2 (ΔP: 0.06 MPa, flow rate: 3.5–6.5 L min^{-1}).

Figure 4 shows relationships between N_{Sh} and $N_{Re}{}^{0.875}N_{Sc}{}^{0.25}$. N_{Sh} was calculated using the k value estimated from Figure 3. As explained by Eqs. 6 and 12, the relationship is linear and the slope of the line corresponds to b in Eq. 12. When b is 0.023, we have the Deissler equation itself and the effect of flux on the mass transfer coefficient is negligible. In contrast, when b exceeds 0.023, the discrepancy is due to the effect of suction flow. Figure 4 shows that the relationship between N_{Sh} and $N_{Re}{}^{0.875}N_{Sc}{}^{0.25}$ almost falls on the Deissler line in the case of the lower flux condition (example 1), which indicates that the effect of suction flow is negligible and that the Deissler equation can be used to estimate the mass transfer coefficient under this condition. In contrast, the relationship between N_{Sh} and $N_{Re}{}^{0.875}N_{Sc}{}^{0.25}$ is far from the Deissler line and b is 0.063 in the case of the higher flux condition (example 2), which indicates that the effect of suction flow is not negligible and

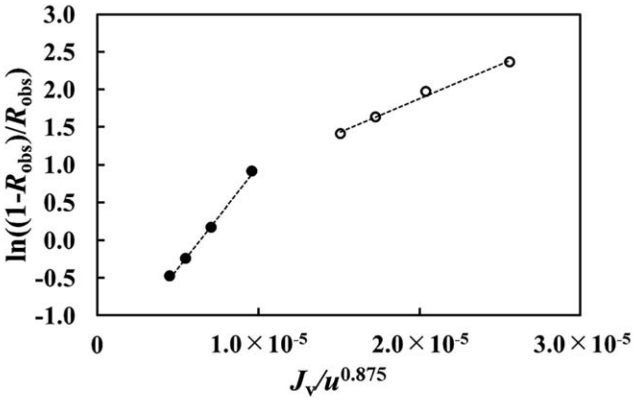

Figure 3. Relationships between ln $\left(\frac{1-R_{obs}}{R_{obs}}\right)$ and $\frac{J_V}{u^{0.875}}$. Closed symbols: example 1 (ΔP: 0.02 MPa, flow rate: 2.0–5.0 L min^{-1}), open symbols: example 2 (ΔP: 0.06 MPa, flow rate: 3.5–6.5 L min^{-1}).

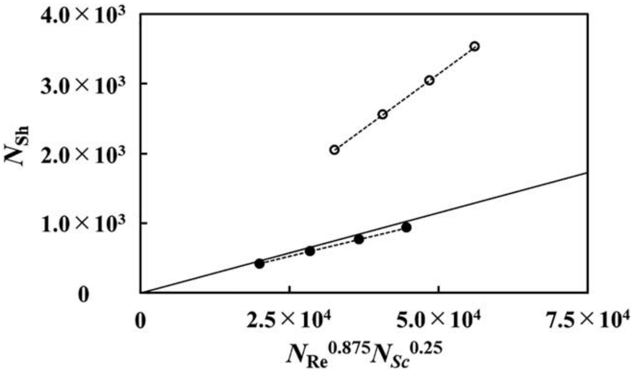

Figure 4. Relationships between N_{Sh} and $N_{Re}^{0.875}N_{Sc}^{0.25}$. The linear line indicates the Deissler equation, given as Eq. 6. Closed symbols: example 1 (ΔP: 0.02 MPa, Flow rate: 2.0–5.0 L $\min^{-1}$), open symbols: example 2 (ΔP: 0.06 MPa, flow rate: 3.5–6.5 L $\min^{-1}$).

that the Deissler equation cannot be used because it underestimates the mass transfer coefficient under this condition. This discrepancy is an issue to be tackled in the present study.

Relationship between the value b and flux

Figure 5 shows the relationship between b and J_v. All data obtained in this study are plotted in the figure. Overall, b is around 0.023 and independent of J_v under the lower flux condition and then increases with J_v under the higher flux condition, regardless of the four experimental combinations given in Table 1. Looking at the data in more detail, we can make the interesting observation that b deviates from 0.023 under the lower flux condition when the molecular weight of the dextran used is larger. Another interesting observation is that b deviates from 0.023 under the lower flux condition when the temperature is lower. A larger molecular weight and lower temperature lead to a smaller diffusion coefficient of the dextrans, and the Deissler equation cannot be used even under the lower flux condition when the diffusion coefficient is smaller. The effect of suction flow includes the effect of flux and that of the diffusion coefficient. In other words, the mass transfer coefficient is affected by the diffusion coefficient of the solute as well as the permeate flux. This is not unexpected because C_m is a function of the ratio of the permeate flux to the diffusion coefficient according to the most fundamental mass balance concept. Additionally, we note that the MWCO of the membrane does not affect the behavior of the value b. This is not unexpected because the mass transfer coefficient should be determined from the principles of transport phenomena outside the membranes, not the principles of transport phenomena inside the pores of membranes.

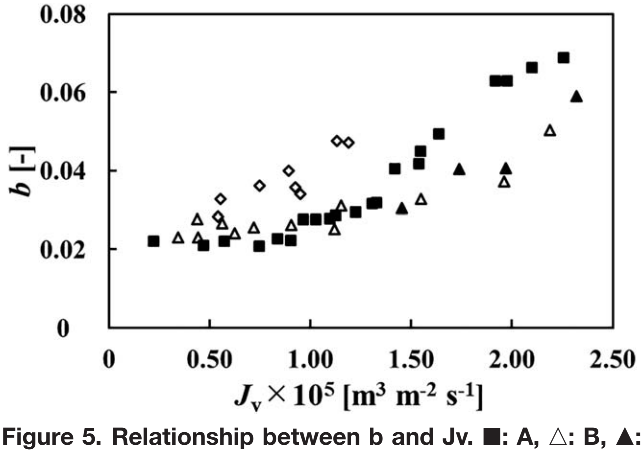

Figure 5. Relationship between b and Jv. ■: A, △: B, ▲: C, ◇: D.

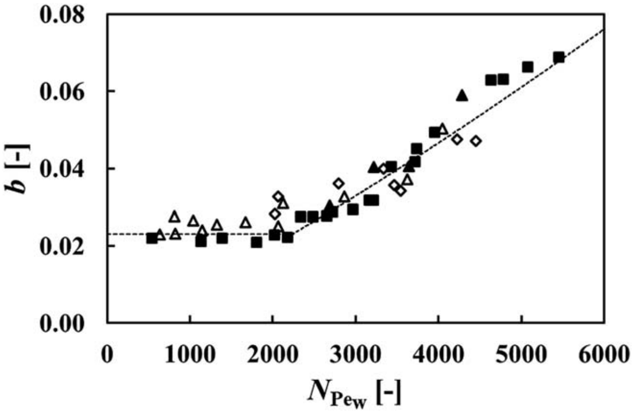

Figure 6. Relationship between b and N_{Pe_w}. ■: A, △: B, ▲: C, ◇: D.

Relationship between the value b and the wall Peclet number

As discussed in the previous section, the ratio of the permeate flux to the diffusion coefficient is an important parameter in understanding the behavior of b, and we thus reorganize the data shown in Figure 5 as a function of the wall Peclet number. The result is shown in Figure 6. Interestingly, regardless of the molecular weight of the dextrans and temperature, the behavior of b can be categorized into two cases. When the wall Peclet number is smaller than 2.23×10^3, b is 0.023. This is because a smaller wall Peclet number means a negiligble effect of the suction flow on the mass transfer coefficient, and we can simply use the Deissler equation in such conditions. In contrast, when the wall Peclet number exceeds 2.23×10^3, b depends on the wall Peclet number and can be expressed as

$$b=2.04\times10^{-6}\times N_{\mathrm{Pe_w}}{}^{1.21} \quad \left(N_{\mathrm{Pe_w}} > 2.23\times10^3\right) \tag{16}$$

The effects of suction flow on the mass transfer coefficient, in particular the effects of the permeate flux and diffusion coefficient of the solute, are included in this empirical equation. This is a simple modification of the Deissler equation and can be used to estimate the mass transfer coefficient even under the higher flux condition.

Herath et al.[19] reported that b can be well expressed as a linear function of J_v, rather than the wall Peclet number. However, their proposal was simply based on their experimental results obtained using two viruses as model solutes having similar diffusion coefficients (i.e., 1.96×10^{-11} and 1.88×10^{-11} m² s⁻¹). It therefore appears that they did not note the effect of the diffusion coefficient on the mass transfer coefficient.

Conclusions

The mass transfer coefficient of tubular ultrafiltration membranes under higher flux conditions was studied, because it has often been pointed out that the Deissler equation is not applicable at higher flux. According to the assumption that k can be expressed as a function of the crossflow velocity, which is used in the VV method, we derived that N_{Sh} is proportional to $N_{Re}{}^{0.875}N_{Sc}{}^{0.25}$, and that the proportionality constant, b, is 0.023 when the effect of suction flow on the mass transfer coefficient is negligible and exceeds 0.023 when the effect of suction flow on the mass transfer coefficient is not negligible. To demonstrate the relationship, we carried out permeation tests using the VV method by changing two dextrans with different molecular weights, two tubular ultrafiltration membranes with different MWCOs, and varying the temperature. The relationship between b and flux depended on the molecular weight of dextran and temperature, and both of these parameters were considered to be closely related to the diffusion coefficient. We thus rearranged the relationship between the wall Peclet number and b, and finally demonstrated that the behavior of b can be classified into two regimes. In the first regime, when the wall Peclet number is smaller than 2.23×10^3, b is 0.023. This is reasonable because a smaller wall Peclet number means a negiligble effect of the suction flow on the mass transfer coefficient, and this gives the Deissler equation itself. In the second regime, when the wall Peclet number exceeds 2.23×10^3, b depends on the wall Peclet number and can be expressed as Eq. 16, which indicates that b is proportional to the 1.21-power of the wall Peclet number. The effect of suction flow on the mass transfer coefficient is the contribution of the integration of the permeate flux with the diffusion coefficient of the solute, and the empirical equation given in this study includes the effect.

Acknowledgment

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Notation

a, p, q = exponent
b = value defined by Eq. 12
C_{b} = concentration of solute in the bulk (mol m^{-3})
C_{m} = concentration of solute at the membrane surface (mol m^{-3})
C_{p} = concentration of solute at the permeate (mol m^{-3})
D = diffusion coefficient (m^2 s^{-1})
d_{h} = equivalent diameter (m)
F = value defined by Eq. 3
J_V = volume flux of pure water (m^3 m^{-2} s^{-1})
k = mass transfer coefficient (m s^{-1})
L_p = pure water permeability (m^3 m^{-2} s^{-1} Pa^{-1})
M_{w} = molecular weight (g mol^{-1})
N_{Pe_w} = wall Peclet number
N_{Re} = Reynolds number
N_{Sc} = Schmidt number
N_{Sh} = Sherwood number
P = solute permeability (m s^{-1})
ΔP = pressure difference (Pa)
R, R_{real} = real objection
R_{obs} = observed rejection
T = temperature (K)
u = crossflow velocity (m s^{-1})
σ = reflection coefficient
$\Delta\pi$ = osmotic pressure difference (Pa)
μ = viscosity (Pa s)

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