$W = M^{-1}R$
$W_k = M_k^{-1}R_k$
w_k = k^{th} column of W
$w_{k,j}$ = j^{th} column of W_k
x = unknown vector

Superscript

T = transpose (row vector)

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Characteristics of Macromolecular Gel Layer Formed on Ultrafiltration Tubular Membrane

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Characteristics of the gel layer were investigated by direct measurement of its concentration treating polyvinylalcohol and ovalbumin aqueous solutions, using cellulose acetate tubular ultrafiltration membranes. The concentration of the gel layer was not constant but a function of bulk concentration and feed velocity. The mass transfer coefficient obtained agreed with Deissler correlation. There was a definite correlation between the resistance of the gel layer and its concentration.

SCOPE

Ultrafiltration is now not only a laboratory curiosity, but it is also an important industrial process. The recent development of many asymmetric high flux membranes has made practical applications possible in a large number of fields. Porter and Michaels (1971*a*, *b*, *c*, *d*, 1972) have reviewed the applications of ultrafiltration in the areas of concentration, recovery, and so on. In the field of wastewater treatment, many applications of ultrafiltration have been reported.

The development of high flux membranes, however, has brought with it the problem of a gel layer formation of macromolecules on the membrane surface. This layer has such a large permeation resistance that the product flux decreases considerably.

Ultrafiltration fluxes have been treated theoretically using the gel polarization model, which deals with the boundary layer between the bulk solution and the gel layer on a membrane surface. But there are two questionable points in this model. One is the assumption that the concentration of the gel layer (C_g) is constant. If C_g is, in fact the gelling concentration, it should change only with the kind of materials used, not with the types of apparatus or modules, or with the experimental conditions. But there have been no experimental results to support this assumption, and the value of C_g has never been measured directly by the previous experiments. The assumption that C_g is constant is quite doubtful and needs experimental verification.

The other problem has to do with the mass transfer coefficient (k). The gel polarization model itself does not specify what k should be used. It is not certain whether the usual k can be applied to ultrafiltration, where the Schmidt number is very large. Moreover, k of macromolecules is considered to change with the bulk concentration when the viscosity and diffusivity also change.

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The objective of this study is to measure C_g directly by treating polyvinylalcohol and ovalbumin, using cellulose acetate ultrafiltration tubular membranes. Mass transfer coefficient, calculated by the gel polarization model using this C_g, is compared with the correlation of mass transfer in a turbulent pipe flow. Finally, the relationship between the permeation resistance of the gel layer and its concentration is studied.

CONCLUSIONS AND SIGNIFICANCE

The concentrations of the gel layer were measured with sufficient accuracy when the gel layer had fully developed on the membrane surface. From the results of these measurements, it became clear that C_g was not constant but a function of bulk concentration and feed velocity, both in the case of polyvinylalcohol and of ovalbumin.

Mass transfer coefficients, calculated from this C_g, agreed very well with the Deissler correlation, which is applicable to the fully developed turbulent pipe flow in the high Schmidt number region (Deissler, 1955). This agreement was obtained in all experiments.

The permeation resistance of the gel layer could be further analyzed using the Kozeny-Carman equation. In this equation, an increase in resistance is expressed by a decrease in the specific surface area in the gel layer on the membrane surface and a corresponding increase in its concentration.

For practical applications of ultrafiltration, an estimation of fluxes under various operational conditions is very important. If the relationship between the gel layer resistance and its concentration is ascertained, ultrafiltrate fluxes can be estimated easily. In this study, it became clear that there was an empirical equation of the form $R_g = \alpha C_g^{1.7}$, where R_g was the resistance to the product flux of the gel layer, and α was a proportional constant. The power, 1.7, was obtained independent of the kind of macromolecule, the characteristic and diameter of tubular membrane, and experimental conditions. Constant α only depended on the macromolecules used. The calculated fluxes using this relationship and the Deissler correlation agreed with experimental values very well.

Therefore, it is possible to estimate ultrafiltration fluxes under given operational conditions in advance, providing the proportional constant in the law of 1.7 power for the macromolecule to be used is known.

In this study, over 98% of both macromolecules was rejected under all experimental conditions, making a quantitative analysis for product concentration impossible.

BACKGROUND

We have much experimental data from the days when ultrafiltration was still a laboratory curiosity. Schott (1964), for instance, purified nonionic detergent solutions using ultrafiltration through cellophane, and Wang et al. (1969) fractionated the biological materials, trypsin and protein, by ultrafiltration using cellulose acetate membrane.

Recently, in many fields, with the development of various asymmetric membranes, practical applications of ultrafiltration have been made possible. Porter and Michaels (1971*a, b, c, d,* 1972) have reviewed the applications in the concentration of milk, egg white, juice, pectin and sugar, and in the recovery of protein from cheese whey, animal blood, gelatin and glue, and so on. Sugar solutions had been treated by reverse osmosis, but the modern application of ultrafiltration included the concentration and purification of sucrose and raffinose solutions (Baker et al., 1972). Grieves et al. (1973) and Bhattacharyya et al. (1975) have reported ultrafiltration of nonionic surfactants and inorganic salts from complex aqueous solutions as an application for laundry wastewater treatment. They have obtained experimental equations of ultrafiltrate fluxes and concentrations as a function of pressure, bulk concentration, feed velocity, and temperature.

With the development of high flux membranes, we have had to deal with the problem of formation of the gel layer. We know this phenomenon is not an internal membrane plugging but a deposition on the membrane surface, because the original value of pure water flux is easily recovered by physical washing of the membrane surface. Ultrafiltrate fluxes under gel layer formation conditions have been treated theoretically using the gel polarization model. Goldsmith (1971) has combined the effect of osmotic pressure to the gel polarization model for considering the decrease of ultrafiltrate flux in macromolecular solutions. But, in contrast to applied pressure, the osmotic pressure of these solutions is usually small enough to be neglected.

Expressions of Flux

In membrane separation processes, the concentration polarization phenomenon, where solutes rejected by the membrane accumulate along the membrane surface, resulting in a higher concentration at the membrane surface than in the bulk, always exists. In the boundary layer between the bulk solution and the membrane surface, the concentration gradient is developed, and accumulated solutes diffuse back into the bulk solution along this gradient.

In the case of ultrafiltration, the solutes are macromolecules and colloids and tend to form the gel layer

on the membrane surface. This phenomenon has been explained schematically by the gel polarization model illustrated in Figure 1 (Baker and Strathman, 1970; de Filippi and Goldsmith, 1970; Blatt et al., 1970; Porter, 1972). Its mathematical treatment is as follows.

Solutes are conveyed by permeate flux to the membrane surface, and a portion of them permeate through the membrane. But the rest of them are rejected by the membrane and diffuse back into the bulk solution. At steady state, the quantity of solutes conveyed to the membrane is equal to the sum of those that permeate through the membrane and those that diffuse back. Thus, the mass balance of solute on a differential element in the boundary layer is given as

$$J_s = J_v \cdot C - D\frac{dC}{dx} \tag{1}$$

In the conventional gel polarization model, the concentration of the gel layer C_g is considered to be constant. Therefore, boundary conditions at steady state after gel layer formation are

$$x = 0; \quad C = C_b$$
$$x = \delta; \quad C = C_g \tag{2}$$

Solute flux through membrane J_s is given as

$$J_s = J_v \cdot C_p \tag{3}$$

Using Equations (2) and (3), Equation (1) is integrated as follows:

$$J_v = k \ln\left(\frac{C_g - C_p}{C_b - C_p}\right) \tag{4}$$

where k is mass transfer coefficient defined as

$$k = \frac{D}{\delta} \tag{5}$$

In Equation (4), if the membrane has a very high rejection of solutes, the term C_p can be neglected. Then

$$J_v = k \ln\left(\frac{C_g}{C_b}\right) \tag{6}$$

The rejection of ultrafiltration membrane for macromolecules is generally very high, especially when the gel layer is formed on the membrane surface. So Equation (6) is ordinarily used for the analysis of ultrafiltration fluxes.

In this treatment, C_g is determined by the extrapolation in plots of fluxes vs. logarithmic bulk concentrations at $J_v = 0$. Blatt et al. (1970) have reported the values of C_g of various kinds of macromolecules: flexible chain linear, well-solvated macromolecules; rigid-chain, solvated macromolecules; highly structured spheroidal macromolecules; colloidal dispersions; and polymer latices.

We have analyzed many ultrafiltrate flux data obtained in our laboratory based on this treatment of the gel polarization model. In these analyses, we found that C_g determined by the extrapolation depended most definitely on the types of apparatus or modules and the experimental conditions. Further, we discovered that a solution, whose concentration was made equal to C_g, sometimes had fluidity, in sharp contrast to a nonfluid gellike state, and that the ultrafiltration flux did not become zero when this solution was used as a feed. The values of C_g measured directly by experiments have never been reported, so it seems essential to measure C_g to find out whether it is a constant or not.

Ultrafiltration fluxes can also be determined by the flow resistances due to membrane and gel layer. Under no gel layer conditions there is only membrane resistance, and flux is expressed as

$$J_v = J_w = \frac{\Delta P}{R_m} \tag{7}$$

If the gel layer is formed, it gives resistance to the flow of the solvent in series to that of membrane. Therefore

$$J_v = \frac{\Delta P}{R_m + R_g} \tag{8}$$

In the gel polarization model, the decrease of flux with time is explained by an increase in the gel layer thickness, namely, an increase in the gel layer resistance. Despite the fact that both Equation (6) and (8) give a value of flux, their relationship has not been investigated. To do this, the relationship between R_g and C_g (or C_b) must be established. But an analysis of ultrafiltration data along these lines has never been undertaken because C_g has been thought to be a constant which depends only on materials and not on experimental conditions. It is necessary, therefore, that the relationship between R_g and C_g be established.

Mass Transfer Coefficient

As for the mass transfer coefficient in Equation (6), the usual correlations for the wall mass transfer coefficient in a turbulent pipe flow are considered applicable.

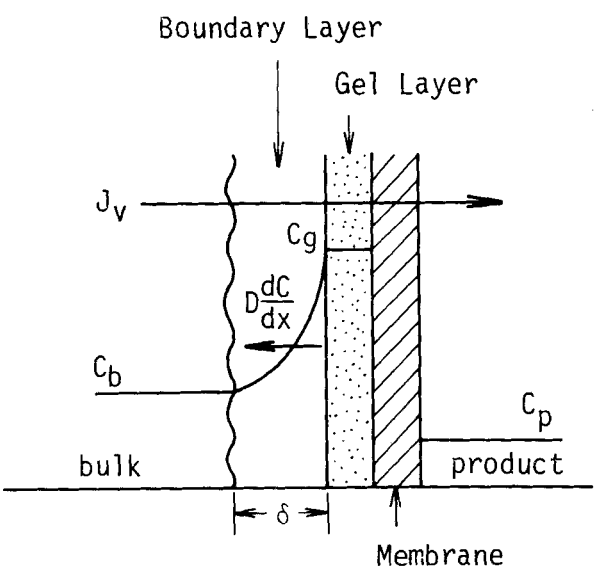

Fig. 1. Gel polarization, schematic.

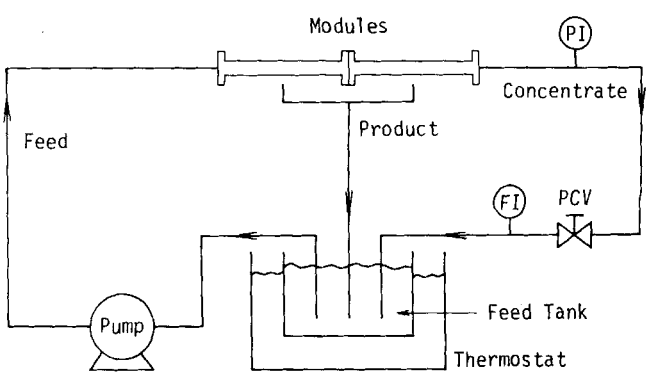

Fig. 2. Schematic flow sheet of experimental apparatus.

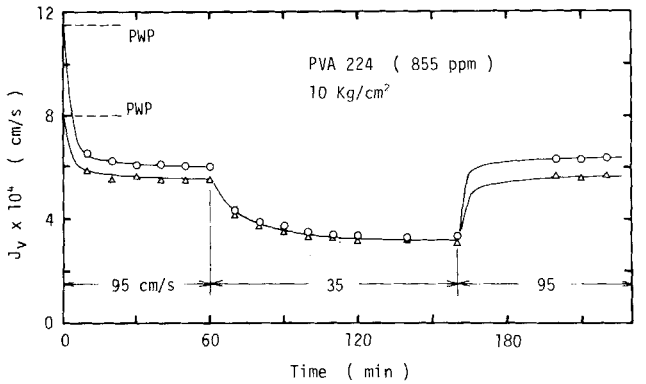

Fig. 3. Flux changes with time and its reproducibility for PVA 224 aqueous solution under two different feed velocities using T2/A (△) and T4/A (○) membrane.

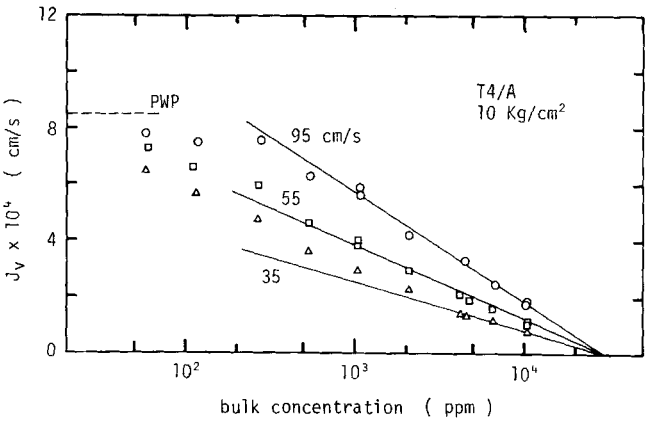

Fig. 4. Experimental fluxes for T4/A membrane, with a 1.25 cm diameter, treating PVA 224 aqueous solutions under three kinds of feed velocities.

The famous correlation given by Chilton and Colburn (1934) is expressed as

$$N_{Sh} = 0.023\, N_{Re}^{0.8} N_{Sc}^{0.33} \qquad (9)$$

Deissler (1955) proposed the following treatment at high Schmidt number. To explain the fully developed turbulent mass transfer, eddy diffusivity is ordinarily used, neglecting the effect of kinematic viscosity. But at high Schmidt number it may be expected that, in the region very close to the wall where turbulence level is low, the effect of kinematic viscosity becomes important. Therefore, he introduced the kinematic viscosity term into the expression of eddy diffusivity and arrived at the following correlation:

$$N_{Sh} = 0.023\, N_{Re}^{0.875} N_{Sc}^{0.25} \qquad (10)$$

In ultrafiltration systems, as Schmidt number is usually very large, it is expected that Deissler's correlation, Equation (10), is more applicable than Chilton and Colburn's, Equation (9).

According to the conventional gel polarization model, mass transfer coefficient in Equation (6) is given as a slope of linear part in flux vs. log (C_b) plots and constant, not depending on C_b, because C_g is assumed constant. But if diffusivity and viscosity change with bulk concentration, mass transfer coefficient also changes, as expressed in Equation (9) or Equation (10). It is necessary, therefore, that this inconsistency is explained. To do this, two kinds of solutes were employed. One was linear chain polymer, polyvinylalcohol, whose diffusivity and viscosity change widely with concentration, and the other was spheroidal macromolecule, ovalbumin, whose properties change only slightly in aqueous solutions.

EXPERIMENTAL

Apparatus and Materials

Two experimental apparatus systems were employed. System 1 consisted of two pairs of two kinds of cellulose acetate ultrafiltration tubular membranes which were available commercially. One pair was T2/A and T4/A membranes, and the other pair was T4/A and T5/A membranes. All three kinds of membranes were supplied by Paterson Candy International, Limited, England. Tubular modules (I.D. 1.25 cm, effective length 30 cm) were made in our laboratory by placing these membranes in brass pipes, which had two lines of small pores on the wall, and by sealing both ends by rubber gaskets. System 2 consisted of two kinds of Abcor tubular modules (I.D.

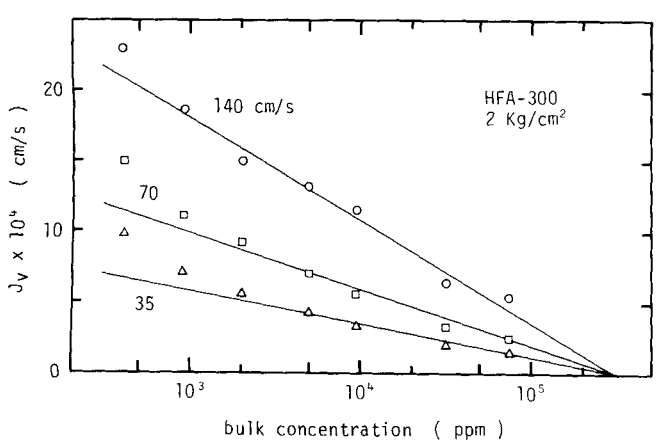

Fig. 5. Experimental fluxes for HFA-300 membrane, with a 2.54 cm diameter, treating ovalbumin aqueous solutions under three kinds of feed velocities.

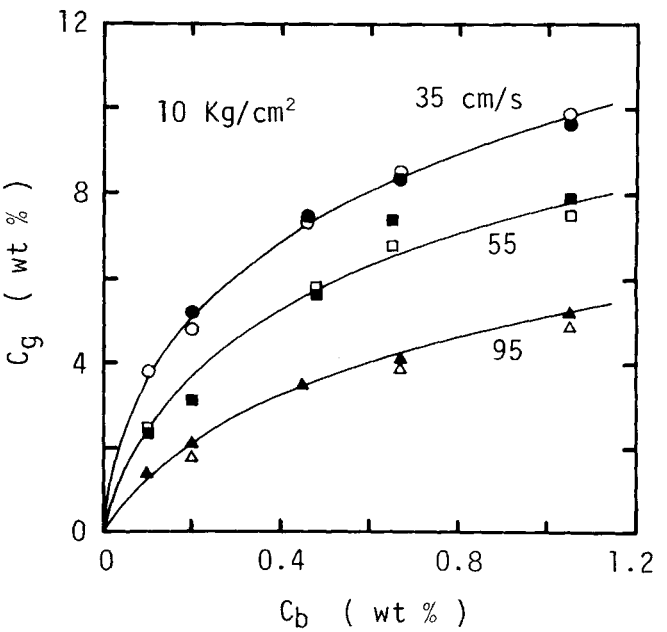

Fig. 6. Gel layer concentrations of PVA 224 measured directly as a function of bulk concentration and feed velocity using T4/A (black) and T2/A (white) membranes.

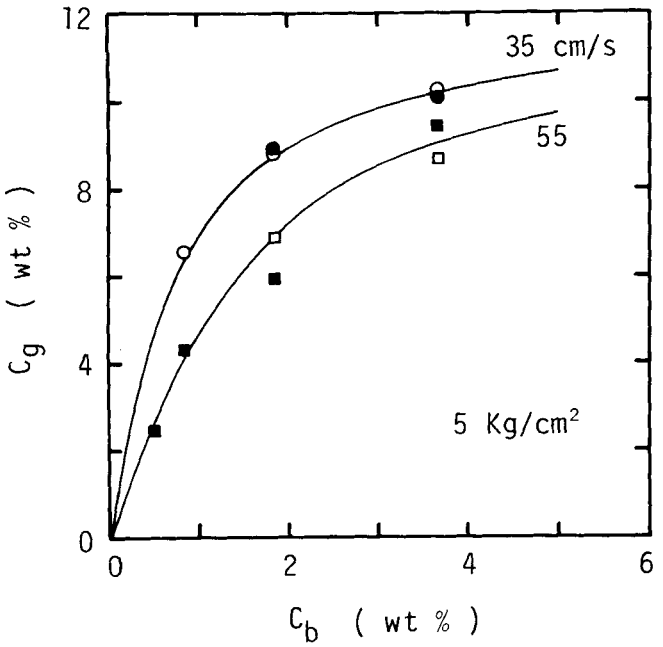

Fig. 7. Gel layer concentration of ovalbumin measured directly as a function of bulk concentration and feed velocity using T5/A (white) and T4/A (black) membranes

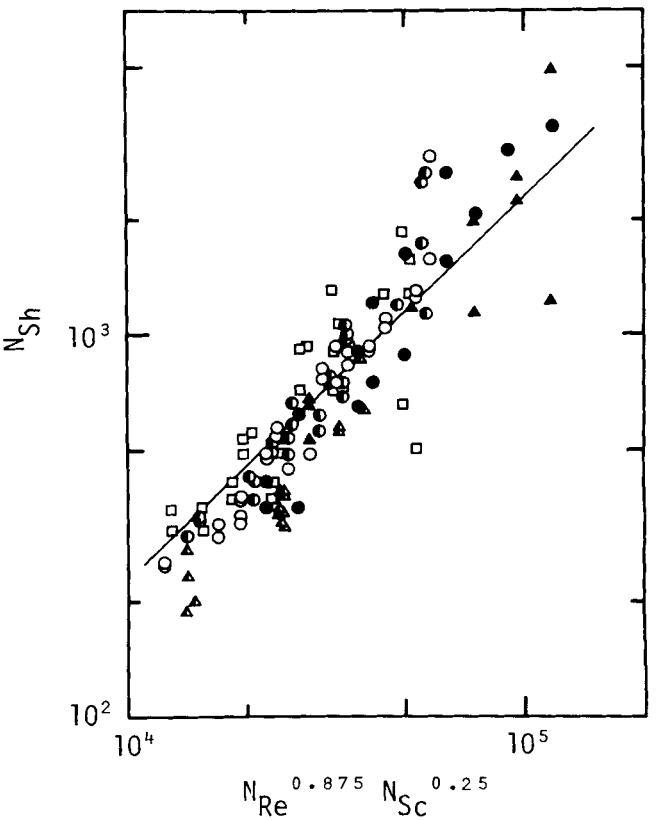

Fig. 8. Comparisons of mass transfer coefficient between experimental and calculated values using Deissler correlation for PVA 224 (○), PVA 205 (□), and ovalbumin (△) under three conditions, white: pressure 10 Kg/cm², membrane tube I.D. 1.25 cm, black: 2 Kg/cm², I.D. 2.54 cm, half black: 5 Kg/cm², I.D. 1.25 cm, and a line indicating Deissler correlation.

2.54 cm, effective length 130 cm), which employed two types of cellulose acetate membranes (Abcor, HFA-180 and HFA-300).

The flow sheet of apparatus system 1 is shown in Figure 2. The two modules were connected in series, and the feed solution was circulated by a high pressure pump with variable speed motor. Product water was returned to the feed tank to prevent concentration of the feed solution. System 2 was similar to system 1, except that system 2 had a constant capacity pump, so the flow rate was controlled by a by-path valve.

Two kinds of solutes were used. One was the linear polymer polyvinylalcohol, and the other was the spherical polymer ovalbumin. Two types of polyvinylalcohol PVA 224 (polymerization degree: $P = 2\,400$, mean molecular weight: $M_w = 100\,000$) and PVA 205 ($P = 500$, $M_w = 20\,000$) were employed. These were supplied by Kurare Company, Japan.

Conditions and Procedure

The experimental conditions were as follows. In system 1, the pressure was controlled at 10 kg/cm² or 5 kg/cm², and three feed velocities (35, 55, 95 cm/s) were employed in the tubular module. In system 2, the pressure was only 2 kg/cm², and the feed velocity was 35, 70, and 140 cm/s in the module. In all experiments, the feed solution temperature was controlled at 25°C by a thermostat. The concentration ranges of the feed solution were from 0.005 to 1% of PVA 224 to 4% of PVA 205 and from 0.04 to 8% of ovalbumin. The concentration of polyvinylalcohol was determined by a total carbon analyzer, and ultraviolet absorption photometry was used for ovalbumin.

The experimental procedure was as follows. At the start of each experiment, pure water permeability (PWP) was measured using ion exchange pure water, and the resistance of the membrane was calculated from Equation (7). Then, the feed water was changed to the polymer solution. The permeate flux decreased rapidly because of the deposition of polymer on the membrane surface and reached the constant value of a steady state within 30 to 50 min. After this steady state flux was measured, the feed solution was depressurized, stopped from circulating, and drained from the modules. Polymer deposited on the membrane surface in high concentration was quickly and completely scratched out by a round rubber plate (one end of which had a slightly larger diameter than that of the membrane tube) and weighed immediately in a wet state. The polymer was weighed again in a dry state, and using these two values, the concentration of the gel layer was calculated. The membrane was washed again with sponge balls before the next experiment.

RESULTS AND DISCUSSION

Changes of Flux

In the beginning, experiments for changes of flux with time were conducted and reproducibility checked. As illustrated in Figure 3, fluxes decreased rapidly and reached the constant values of steady state after 30 to 50 min. At the feed velocity of 95 cm/s, the gel layer did not completely control the flux, so the values of steady state fluxes were different between the two kinds of membranes. But at 35 cm/s, as the gel layer completely controlled permeation, the flux values of the two membranes were almost the same. Changing the feed velocity again to 95 cm/s, fluxes recovered to the steady state values which were obtained in the first experiment. It is clear from these results that the gel layer formed reaches a steady state in which polymers arriving at the gel layer by flow are equal to those leaving by diffusion. Changes with time and reproducibility of fluxes for ovalbumin solutions were almost identical to those of polyvinylalcohol.

According to the conventional treatment of the gel layer, namely, gel polarization model, flux vs. log (C_b) plots have linear parts in the region where fluxes are controlled by the gel layer, as explained by Equation (4) or Equation (6), and these lines merge at one point on a concentration axis when J_v is zero, which gives the gel layer concentration. In this study, these same tend-

sodium chloride solution, lines which converge at one point on the C_b axis can be drawn, but this concentration has no significance and, of course, is not C_g. Most important, experimental results show that C_g is not constant. The intersection in the plots of flux vs. C_b has no significance.

Concentration of Gel Layer

Results of measurements of gel layer concentrations are illustrated in Figures 6 and 7. These concentrations were the function of bulk concentration and feed velocity; they increased with a decrease in feed velocity and an increase in bulk concentration. These tendencies were obtained for both kinds of solutes, Figure 6 for polyvinylalcohol and Figure 7 for ovalbumin, regardless of the membranes used, and properties of solutes to the concentration change.

These results show that the previous assumption of the gel polarization model, namely, C_g as constant, is not true.

Mass Transfer Coefficient

From the concentration of the gel layer measured directly, mass transfer coefficient can be calculated using Equation (6). In this study, it was compared with two mass transfer correlations, that of Chilton and Colburn, Equation (9), and of Deissler, Equation (10). Experimental data agreed with the Deissler correlation better than with that of Chilton and Colburn, as expected earlier, for both polyvinylalcohol and ovalbumin independent of the characteristics and diameters of tubular membranes, pressure, and bulk concentrations (Figure 8).

Diffusivities and kinematic viscosities used for this calculation were as follows. Diffusivity and its change with concentration of polyvinylalcohol were obtained using results of Dialer et al. (1952). For PVA 224, diffusivity is almost constant, 1.8×10^{-7} cm²/s, but for PVA 205, it varies with concentration as $D = 4.1 \times 10^{-7}\ (1 - 0.08 \times C)$, where C is concentration (weight percent) of PVA 205. Tiselius and Gross (1934) measured diffusivity of ovalbumin in acetate buffer at pH 4.7 and 20°C and reported the value 7.67×10^{-7} cm²/s for aqueous solution which was estimated from the measured value by correction of viscosity using Einstein relation. In this study, the value of 8.7×10^{-7} cm²/s, which was the corrected value at 25°C, was used for the calculation. This value is assumed to be nearly constant independent of concentration. Kinematic viscosities of polyvinylalcohol were measured at various concentrations at 25°C. For PVA 224, it is about 2.4 cst at 1 wt % aqueous solution and increases rapidly with an increase in its concentration. Kinematic viscosity of PVA 205 is much smaller than that of PVA 224. For ovalbumin, the changes of viscosity are neglected, and the value of that is 1.0 cst.

As illustrated in Figure 8, data for both polyvinylalcohol and ovalbumin agreed well with Deissler correlation. The reason for the scattering on the right side of Figure 8 is the inaccuracy of measurements of gel layer concentrations. When the feed velocity and/or bulk concentration was high, namely on the right side of Figure 8, gel layer formation was not complete enough to make accurate measurements.

It can be concluded from these results that the mass transfer coefficient of an ultrafiltration system treating high Schmidt number solution can be estimated using the Deissler correlation, which was derived for a fully developed turbulent mass transfer in a circular pipe.

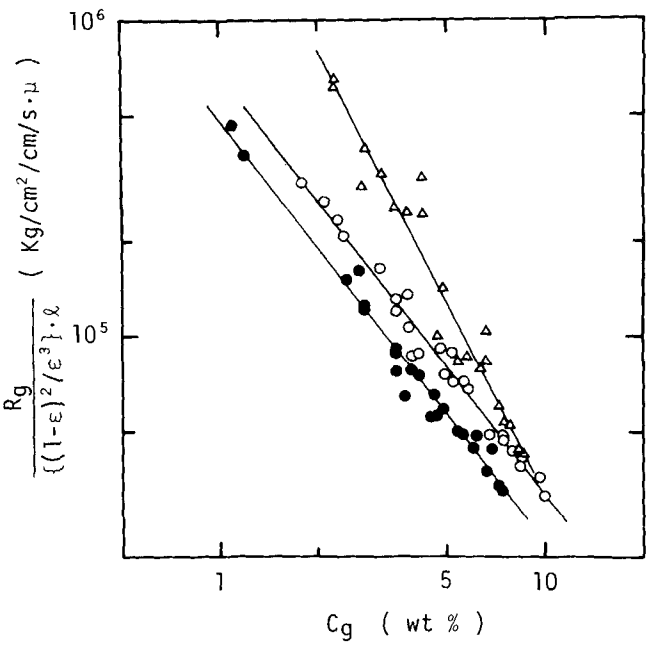

Fig. 9. Changes of specific surface area of gel layer with its concentration for PVA 205 (△) and PVA 224 (○) under two kinds of pressure, white: 10 Kg/cm² and black: 5 Kg/cm².

Resistance of Gel Layer

The permeation resistance of the gel layer, which is in series to that of the membrane and is determined by Equation (8), can be analyzed by the Kozeny-Carman equation. This equation was used for the hydraulic permeability of a packed bed of nearly spherical particles and applied to the analysis of cake filtration. Gel layer formation is similar to that of cake, so this equation was applied to analyze the gel layer of ultrafiltration.

Using the Kozeny-Carman equation, ultrafiltration velocity is determined as follows:

$$J_v = \frac{\epsilon^3}{KS_v^2(1-\epsilon)^2} \cdot \frac{g_c \Delta P}{\mu l} \qquad (11)$$

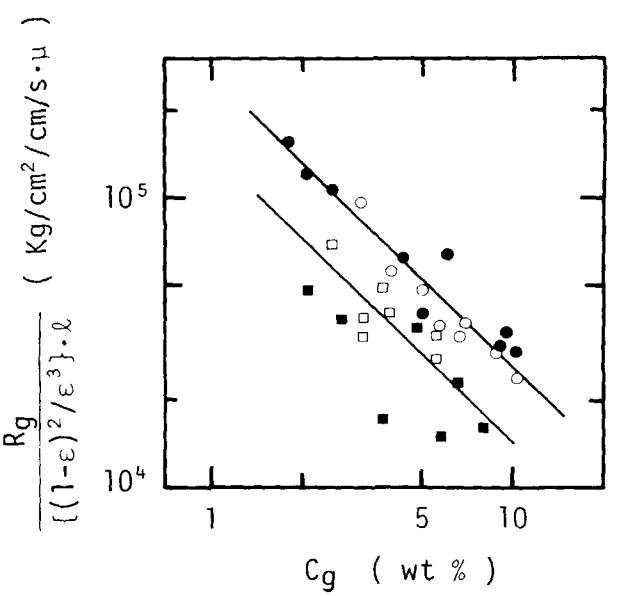

Fig. 10. Changes of specific surface area of gel layer with its concentration for ovalbumin under two kinds of pressures, 5 Kg/cm² (○)—white: T5/A, black: T4/A, and 2 Kg/cm² (□)—white: HFA-300, black: HFA-180.

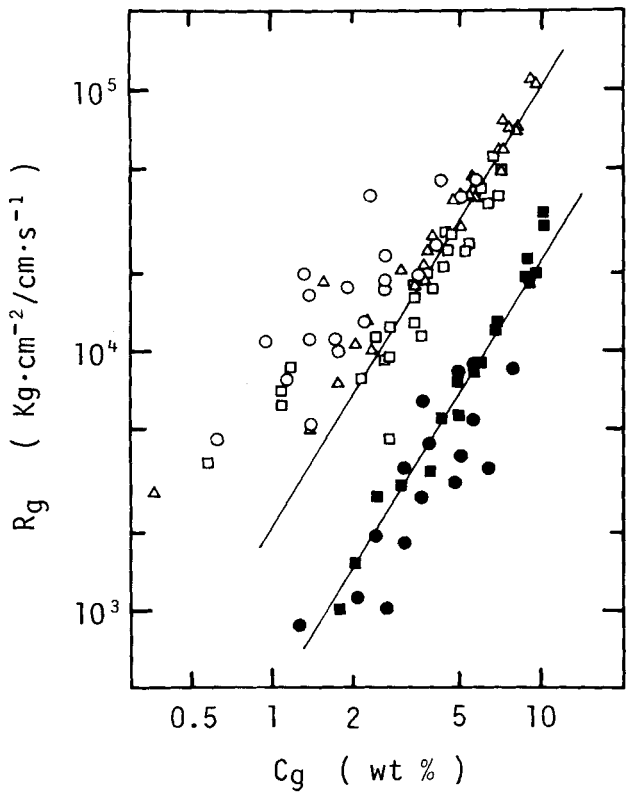

Fig. 11. Relationships between resistance to a flow and concentration of gel layer for PVA 224 (white) and ovalbumin (black) under various conditions, pressure 2 Kg/cm^2, membrane diameter 2.54 cm: (○), 5 Kg/cm^2, 1.25 cm: (□), 10 Kg/cm^2, 1.25 cm: (△).

Viscosity of fluid μ is considered to be the value of water because of high rejection. In the region where the gel layer controls fluxes completely, it can be regarded that the resistance of gel layer is affected by its porosity,

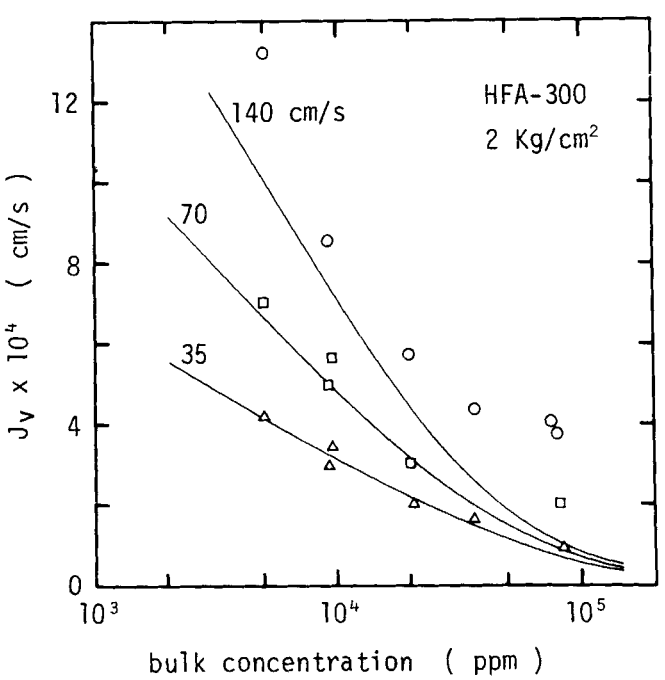

Fig. 13. Comparisons between experimental and calculated fluxes for ovalbumin aqueous solutions under three kinds of feed velocities, curves calculated from Equations (6), (8), (10), and (16).

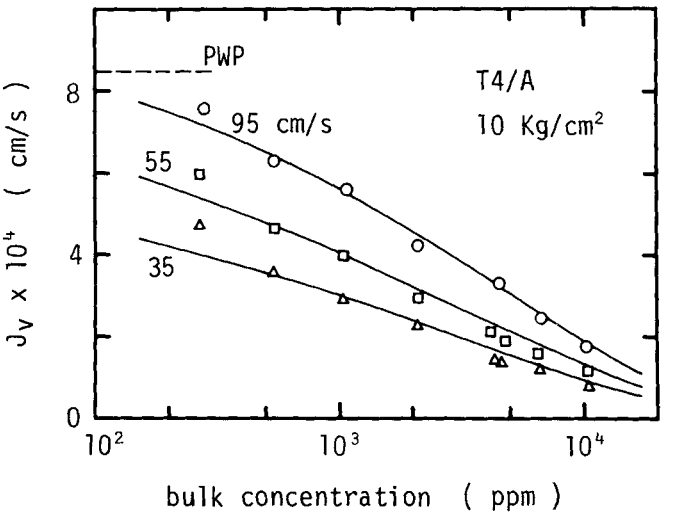

Fig. 12. Comparisons between experimental and calculated fluxes for PVA 224 aqueous solutions under three kinds of feed velocities, curves calculated from Equations (6), (8), (10), and (15).

specific surface area, and thickness, as compared with Equation (8) and Equation (11). Therefore

$$R_g \propto \frac{(1-\epsilon)^2}{\epsilon^3} \cdot S_v^2 \cdot l \qquad (12)$$

Gel layer thickness can be calculated from the result of its concentration measurement, assuming that the thickness is uniform and the gel layer is almost incompressible. Porosity also can be estimated as follows. If the volume of water in the gel layer corresponds to the void in it and the density of the gel layer is almost 1.0, then

$$\epsilon \simeq (1 - C_g) \qquad (13)$$

Finally, Equation (12) is rewritten as

$$S_v^2 \propto R_g \Big/ \frac{(1-\epsilon)^2}{\epsilon^3} \cdot l \qquad (14)$$

In Figures 9 and 10, the right-hand side of Equation (14) is plotted against the concentration of the gel layer. These figures show that the specific surface area of the gel layer decreased with an increase in its concentration. Slopes depend on the kind of macromolecule. The specific surface area of a linear chain macromolecule decreased more rapidly than that of a spherical one. It can be considered qualitatively that the contact of molecules increases at higher concentration so that overall surface decreases. In the case of a linear chain polymer, chains are entangled more than those of a spherical one, so the decrease of polyvinylalcohol was more rapid than that of ovalbumin. In these figures, the effect of pressure also seemed to exist. Further quantitative analysis, however, is impossible right now because there has been no literature about the specific surface area change of a macromolecule with variation with its concentration.

Relationship between R_g and C_g

For practical applications of ultrafiltration, the estimations of fluxes under given operating conditions are very important. Fluxes are calculated from Equation (6) and/or Equation (8), but the effects of pressure and membrane resistance do not appear in Equation (6)

and the effects of feed velocity and bulk concentration do not appear in Equation (8). Therefore, if the relationship between Equation (6) and Equation (8) can be obtained, fluxes can be estimated under various operating parameters.

The simplest relationship which connected Equation (6) to Equation (8) is that between the resistance of gel layer and its concentration. Figure 11 shows these relationships for PVA 224 and ovalbumin. It is clear that there is an empirical law of 1.7 power regardless of the kind of polymer, the characteristics, and diameter of membrane, pressure, feed velocity, and bulk concentration. The absolute value of resistance depends on the kind of solute. The empirical equations obtained are as follows:

For PVA 224

$$R_g = 2.0 \times 10^3 \cdot C_g^{1.7} \tag{15}$$

For ovalbumin

$$R_g = 4.5 \times 10^2 \cdot C_g^{1.7} \tag{16}$$

With these results, fluxes treating polyvinylalcohol or ovalbumin aqueous solution can be estimated under any operation conditions using Equations (6), (8), and (15) or (16). This calculation for estimation is done by trial and error. Mass transfer coefficient in Equation (6) is determined with the Deissler correlation, Equation (10), from given feed velocity.

Results of these calculations of fluxes are illustrated in Figures 12 and 13 in comparison with experimental data. For PVA 224, the calculation values agreed with experimental ones very well in the region where the gel layer was formed completely (Figure 12). In the case of ovalbumin, Figure 13, the agreement of both values was not so good at high feed velocity. The reason for disagreement might be that in the apparatus, system 2, which was used in this experiment, modules vibrated so violently (an effect of the vibration of the pump at high feed velocity) that mass transfer coefficient became much larger than that estimated from Equation (10), and experimental fluxes became larger than calculated values. At high bulk concentration region in Figure 13, calculated fluxes disagreed with experimental data. This indicates that the gel layer has much less permeation resistance than that estimated in Figure 11 or Equation (16), and one may consider that the vibration of the module prevents the formation of the gel layer on the membrane surface more than a certain thickness.

Though there are a little disagreement in Figures 12 and 13, it can be concluded that fluxes will be estimated using Equations (6), (8), (10) and the relationships between resistance and concentration of gel layer. Hereafter, it will be necessary to investigate the relations given by Equations (15) and (16) theoretically and to develop a convenient method to determine the proportional constants by independent measurement.

NOTATION

C = concentration of solute, wt %
D = diffusivity, cm^2/s
g_c = conversion constant of gravity
J_s = permeation velocity of solute through membrane, cm/s
J_v = volume flux through membrane, $cm^3/cm^2 \cdot s$
J_w = volume flux of pure water, $cm^3/cm^2 \cdot s$
k = mass transfer coefficient, cm/s
K = Kozeny constant
l = thickness of gel layer, cm
N_{Re} = Reynolds number
N_{Sc} = Schmidt number
N_{Sh} = Sherwood number
ΔP = pressure difference, Kg/cm^2
R = resistance to flow, $Kg \cdot cm^{-2}/cm \cdot s^{-1}$
S_v = specific surface area, cm^2/cm^3
x = distance, cm
δ = boundary layer thickness, cm
ϵ = porosity
μ = viscosity, $g/cm \cdot s$

Subscripts

b = bulk
g = gel layer
m = membrane surface
p = product

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