THEORETICAL DESCRIPTIONS OF MEMBRANE FILTRATION OF COLLOIDS AND FINE PARTICLES: AN ASSESSMENT AND REVIEW

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ABSTRACT

Membrane separation technology is a novel and highly innovative process engineering operation. Membrane processes exist for most of the fluid separations encountered in industry. The most widely used are membrane ultrafiltration and microfiltration, pressure driven processes which are capable of separating particles in the approximate size ranges of 1 to 100 nm and 0.1 to 10 \( \mu \text{m} \), respectively.

The design of membrane separation processes, like all other processes, requires quantitative expressions relating material properties to separation performance. The factors controlling the performance of ultra- and microfiltration are extensively reviewed. There have been a number of seminal approaches in this field. Most have been based on the rate limiting effects of the concentration polarisation of the separated colloids at the membrane surface. Various rigorous, empirical and intuitive models exist, which have been critically assessed in terms of their predictive capability and applicability. The decision as to which of the membrane filtration models is the most correct in predicting permeation rates is a matter of difficulty and appears to depend on the nature of the dispersion to separated. Recommendations are made as to which of the existing models can be most appropriately applied to different types of dispersions.

1. INTRODUCTION

Membrane separation technology is a novel and highly innovative process engineering operation. Membrane filtration processes are nowadays used as an alternative to conventional industrial separation methods such as distillation, centrifugation and extraction, since they potentially offer the advantages of highly selective separation, separation without any auxiliary materials, ambient temperature operation, usually no phase changes, continuous and automatic operation, economical operation also in small units, modular construction and simple integration in existing production processes, as well as relatively low capital and running costs. The former advantages make membrane processes even more interesting for certain types of materials which have been inherently difficult and expensive to separate, such as,

1. Dispersions of colloids and fine particles, especially those which are compressible, have a density close to that of the liquid phase, have high viscosity, or are gelatinous.

2. Biological materials, which often fall in the colloidal size range and are very sensitive to their physical and chemical environment.

3. Low molecular weight, non-volatile organics or pharmaceuticals and dissolved salts.

The various membrane separation methods can be divided according
to their separation characteristics which may be classified by the size-
range of materials separated and the applied driving force. Ultrafiltra-
tion and microfiltration separate solutes and colloidal particles of the size
of approximately 1 to 100 nm and 0.1 to 10 μm respectively, using a
pressure difference of usually 100 to 500 kPa as the driving force.

Today ultrafiltration and microfiltration membranes are most com-
monly made of polymeric materials such as polyamide, polysulphone,
cellulose-acetate, polycarbonate and a number of other advanced poly-
mers. However, recent developments of inorganic membranes composed
of materials such as ceramic, aluminium-oxide or silica-glass show ad-
vantageous properties compared to polymeric types, like higher tempera-
ture stability, increased resistance to fouling and narrower pore size
distribution, despite their high capital costs.

Various configurations exist to support or contain the membranes.
Which configuration, for instance tubular modules, hollow fibre modules,
plate-and-frame modules or spiral-wound modules is to be used depends
both on the solution which should be filtered and the operating condi-
tions.

In general, three filtration modes can be distinguished: unstirred and
stirred dead-end filtration and cross-flow filtration. In the unstirred
dead-end filtration the solution is put under pressure without any agita-
tion in the liquid. In the stirred dead-end mode agitation is provided with
a stirring bar. In cross-flow filtration the solution is pumped to flow
tangentially over the membrane surface.

In recent years, membrane separation processes have found wide
application. Membrane processes exist for most of the fluid separations
encountered in industry. The development of quantitative predictive
models is, therefore, of great importance for the successful application of
membrane separation processes in the process industries. The design and
simulation of membrane separation processes, like all other processes,
require quantitative expressions relating material properties to separa-
tion performance. The physical theories governing the filtration models
principally describe the effect of the concentration polarization phenom-
enon at the membrane surface.

This article presents a review and assessment of the existing filtration
models for colloidal and fine particle dispersions with emphasis on their
qualitative and quantitative predictive capability, their scientific basis,
and their limitations.
1.1 Theory of Ultrafiltration

Ultrafiltration is a pressure-driven membrane process by which macro-molecular solutes and/or colloidal particles are separated from a solvent, usually water. The relationship between the applied ultrafiltration pressure and the rate of permeation (flux) for a pure solvent feed flowing under laminar conditions in tortuous membrane channels may be described by the Carman–Kozeny equation (Carman (1938))

\[ J = \frac{\Delta p}{\mu R_m} \]

where \( J \) is the flux (volumetric rate per unit area), \( \Delta p \) the transmembrane pressure difference, \( \mu \) the solvent viscosity and \( R_m \) the membrane resistance. The general approach to describe ultrafiltration in the presence of a solute is given by

\[ J = \frac{\Delta p - \Delta \pi}{\mu(R_m + R_s)} \]

where \( \Delta \pi \) is the difference in osmotic pressure across the membrane, and \( R_s \) represents reversible (concentrated layer, filter cake) and sometimes irreversible (foulants) deposition of solute (or solids) onto the membrane surface.

1.2 Concentration Polarization

The separation of solute and solvent takes place at the membrane surface where the solvent passes through the membrane and the retained solute causes the local concentration to increase, an effect which is known as concentration polarization. Thereby a concentration profile is established within a boundary film generated by the hydrodynamic conditions (see Fig. 1).

With the higher concentration at the membrane surface, there will be a tendency of solute to diffuse back into the bulk solution according to Fick's law of diffusion. A solute mass balance above the membrane surface at steady state condition and with the assumption that
Fig. 1. Concentration polarization at a membrane surface.

1. solvent and solute densities are similar,
2. the diffusion coefficient is constant and
3. concentration gradients parallel to the membrane are negligible compared with the concentration gradients orthogonal to the membrane,

gives the rate of convective transport of solute towards the membrane surface equal to the rate of solute leakage through the membrane plus the rate of solute due to back-diffusion,

\[ J_c = D \frac{dc}{dy} + J_{cp} \]  

(3)

where \( c \) and \( c_p \) are solute concentrations in the boundary layer and in the permeate, respectively and \( D \) is the diffusion coefficient of the solute in the solvent. An integration of Eq. (3) over the boundary layer thickness \( \delta \) with the boundary conditions

\[ c(y = \delta) = c_b \quad c(y = 0) = c_m \]

gives the well-known film model relationship:
\[ J = k_s \ln \left( \frac{c_m - c_p}{c_b - c_p} \right) \]  

where \( k_s = D/\delta \), the overall mass transfer coefficient of the solute in the boundary layer, and \( c_m \) is the concentration at the membrane surface.

The overall mass transfer coefficient is usually obtained from correlations of the form

\[ \text{Sh} = \frac{k_s d_h}{D} = K \text{Re}^a \text{Sc}^b \left( \frac{d_h}{L} \right)^c \]  

where the constants \( K, a, b, c \) vary with the flow regime (see Blatt et al. (1970), Porter (1972a), Gekas and Hallström (1987)). The above boundary layer theory applies to mass transfer controlled systems where the permeate flux is independent of pressure (no pressure term in the model).

2. GEL-POLARIZATION MODEL

Several investigators (Blatt et al. (1970), Porter (1972a,b), Henry (1972), Kozinski and Lightfoot (1972), Fane et al. (1981)) found from plots of flux versus applied ultrafiltration pressure of most macromolecular solution and colloidal dispersion experiments that the steady state flux reaches asymptotically a limiting value where further increase in applied pressure results in minimal increase in permeate flux. The existence of this flux plateau cannot be explained from the basic principle of the film model of concentration polarization.

Michaels (1968) and Blatt et al. (1970) forwarded a hypothesis that as the concentration at the membrane surface increases due to polarization the macrosolute reaches its solubility limit and precipitates on the membrane surface to form solid or thixotropic gels. For colloidal dispersions the gel layer is expected to resemble a layer of close-packed spheres (Porter (1972a)). As a consequence, a constant gel layer concentration is rapidly reached, which is expected to be virtually independent of bulk solution concentration, applied pressure, fluid flow conditions or membrane characteristics. According to Michaels' model, an increase in applied pressure produces a temporary increase in flux, which brings more solute to the gel layer and increases its thickness (increase in hydraulic resistance to solvent flow), thereby reducing the flux to the original level.
For 100% solute rejection \((c_p = 0)\) Eq. (4) can thus be written in the form

\[
J_{\text{lim}} = k_s \ln \left( \frac{c_g}{c_b} \right)
\]

where \(c_m\) has been replaced by the constant gel layer concentration \(c_g\) and \(J_{\text{lim}}\) is the limiting flux.

Blatt et al. (1970), Porter (1972a,b), Goldsmith (1971), Henry (1972) and Madsen (1977), among many others, who have studied macrosolute and colloidal ultrafiltration, have obtained experimental results for the limiting flux which give strong support to the gel-polarization model, namely,

- \(J_{\text{lim}}\) is independent of applied pressure,
- \(J_{\text{lim}}\) is semi-logarithmically related to \(c_b\),
- \(J_{\text{lim}}\) approaches zero at a limiting bulk concentration \(c_{b,\text{lim}}\) which is equal the gel concentration \(c_g\) (or the cake-concentration in the case of colloidal dispersions),
- \(J_{\text{lim}}\) may be modified by factors which alter the overall mass transfer coefficient \(k_s\).

Porter (1972a) reported that the agreement between theoretical and experimental ultrafiltration rates for macromolecular solutions can be said to be within 15 to 30% using the Lévêque (laminar flow) and Dittus–Boelter (turbulent flow) mass transfer relationships for flow in non-porous channel and tubular systems. The success of the relationships in indicating the variation (power dependence) of ultrafiltration flux with channel geometry and fluid velocity is gratifying. However, for colloidal dispersions, experimental flux values are often one to two orders of magnitude higher than those indicated by the Lévêque and Dittus–Boelter relationships. Similar discrepancies for colloidal ultrafiltration were noted by Blatt et al. (1970).

2.1 Limitations of the Model

The gel-polarization model has proved to be useful in the interpretation of ultrafiltration performance (Fane (1986)). However, it bears some implications which put its foundation in doubt.

Porter (1972a) noted that for the ultrafiltration of human albumin in spiral flow channel plates with 10 mm and 30 mm channel-height for
laminar and turbulent flow, respectively, the plots of flux versus bulk concentration gave surprisingly different gel concentrations. A fact which cannot be accounted for by the gel-polarization model.

Nakao et al. (1979), who have measured the concentration of the gel layer at steady state conditions for macromolecular solutions, found that the gel concentration is not constant but rather a function of bulk concentration and cross-flow velocity, which confirmed Porter's experimental observations. Furthermore, they found from experiments using a bulk concentration equal to the calculated gel concentration (obtained by extrapolation of flux versus \( \ln c_b \) plots), that the permeate flux was not zero as predicted by the gel-polarization model.

Fane et al. (1981) observed experimentally that the gel-polarized behaviour with identical solutions and hydrodynamics produced different limiting flux values when membranes of differing permeability were used. This is in contradiction to the gel-polarization model, since no dependence of membrane properties is implied.

Blatt et al. (1970) and Porter (1972a) ascertained that colloidal ultrafiltration flux is several times higher than predicted by the gel-polarization model and the conventional mass transfer correlations. This has been termed, the \textit{flux paradox} for colloidal dispersions. Porter related this finding to the tubular-pinch effect (Segré and Silberberg (1962)), which is sometimes known as the radial migration phenomenon. The tubular-pinch effect as well as further explanations are presented in Section 5.

Thus the gel-polarization model appears to have some physical limitations although in some cases it still remains the most convenient model from a practical point of view (Fane (1986)).

2.2 Model Developments with Variable Physical Properties

Kozinski and Lightfoot (1972) developed a theoretical model for predicting permeate flux through a rotating disk ultrafiltration membrane taking into consideration concentration dependent diffusivity and viscosity. The model is based on the concentration–diffusion equation (in a form equivalent to Eq. (7)) and the equations of motion (in a form equivalent to Eq. (8), (9)), which have to be solved simultaneously to give a numerical solution. Experimental studies were performed with bovine serum albumin (BSA) solutions and the results compared quite well with the numerical
predictions for situations in which the protein has not denatured. However, the model could not account for the experimental finding that the filtration rate increased by 50% when ionic strength was lowered by a factor of 5. They suggested that these effects might result from the instability of bovine serum albumin at low ionic strength and in particular its tendency to undergo polymerization.

Their numerical solution was found to be very sensitive to the exact behaviour of the diffusivity, osmotic pressure and viscosity at high concentrations. The lack of sufficient diffusion data (see Fig. 2) may be the greatest source of uncertainty in the prediction of ultrafiltration performance (Kozinski and Lightfoot (1972), Shen and Probstein (1977), Probstein (1978), Trettin and Doshi (1980a,b), Vilker et al. (1981)). Kozinski and Lightfoot extended their model to parallel plate systems.
and considered the limitations of Blatt's et al. (1970) thin channel ultrafiltration data for bovine serum albumin. The predictions were low but well within the accuracy of Blatt's data.

Shen and Probstein (1977) proposed that the physical model of Michaels (1968) is essentially correct in its qualitative behaviour and that the quantitative disagreement is principally a consequence of not considering the variable transport properties of the macromolecular solution, in particular, the variability of the diffusion coefficient. They obtained out of the concentration-diffusion equation for steady, Newtonian, fully developed laminar flow in a parallel plate system

\[ u \frac{\partial c}{\partial x} + v \frac{\partial c}{\partial y} = \frac{\partial}{\partial y} \left( D(c) \frac{\partial c}{\partial y} \right) \]  

(7)

with a concentration dependent diffusion coefficient \( D(c) \) and \( u,v \) the velocity components longitudinal and normal to the membrane surface, respectively, that the limiting flux is proportional to the dimensionless quantity \( (D(c_g) / D(c_b))^{2/3} \). This result suggests simply the replacing of the diffusion coefficient evaluated at \( c_b \) by one evaluated at \( c_g \) in the mass transfer correlation for laminar channel flow of the gel-polarization model. This finding is consistent with Kozinski's and Lightfoot's (1972) observation that the critical region for mass transfer is adjacent to the membrane surface.

Shen and Probstein (1977) compared the calculated results for the limiting flux of their modified film model with that of an exact numerical solution developed out of the boundary layer concentration-diffusion equation with variable diffusivity (Eq. (7)), coupled to the channel momentum equation

\[ \tau = \mu(c) \frac{\partial u}{\partial y} \]  

(8)

with variable viscosity and the continuity equation

\[ \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0 \]  

(9)

The agreement proved to be quite good for a bovine serum albumin solution of known properties despite the rather complex behaviour of the
diffusion coefficient through the boundary layer.

Many measurements of bovine serum albumin diffusivity (Fig. 2) at dilute concentrations (usually below 1 wt.%) have been reported in the literature, where solution properties like buffer type, pH-value (electrical charge on the micro-ions), and ionic strength have negligible effects on the diffusion coefficient. However, there is a lack of data at high concentrations which is especially limiting since the above stated factors cannot be ignored as the interaction among the charged macro-ions becomes more pronounced. Furthermore, Shen and Probstein (1977) and Trettin and Doshi (1980b) stated that even existing data scattered appreciably for the same solution conditions.

Shen and Probstein (1977) tested their numerical model for the sensitivity of the dependence of the limiting flux on viscosity. Their predicted flux values for a constant bulk viscosity were at most 50% higher than the calculations when the dimensionless viscosity was taken to vary from 1 to infinity in the concentration boundary layer as the bulk concentration increases to the gel concentration. Therefore, they concluded that the limiting flux will be much more dependent on the variation of the diffusion coefficient than the viscosity coefficient. This finding further confirmed the assumption of a constant viscosity which was adopted in deriving the modified film model equation.

Shen and Probstein (1977) compared also Blatt’s et al. (1970) experimental data of bovine serum albumin in 0.9% saline-water solution with their calculated data using the modified film-model. The predictions were consistently higher by a small amount, due to uncertainties by the extrapolation of diffusion coefficient data at high gel concentrations and unknown pH-values of Blatt’s et al. (1970) experiments. However, Trettin and Doshi (1980a) noted that Shen and Probstein (1977) as well as Kozinski and Lightfoot (1972) misinterpreted the system design of Blatt’s et al. (1970) experiments, which led to them using a cross-flow velocity four times higher than reality. Use of the correct velocity in the predictions of the modified model of Shen and Probstein would result in 24 to 34% lower flux values than measured by Blatt et al. (1970).

Probstein et al. (1978) developed an integral method based on the partial differential equations of continuity (Eq. (9)) and diffusion (Eq. (7)) with a concentration dependent diffusivity governing the behaviour of a developing concentration boundary layer of constant density fluid flowing under laminar conditions in a plane channel. The result is valid when the gelling concentration is large compared to the bulk concentration and
justifies analytically the result previously obtained by Shen and Probstein (1977), namely the replacing of the bulk diffusivity by the diffusivity at the gel concentration in the widely used formula of Michaels (1968). Comparison of the analytic result for the limiting flux with both exact numerical solution (Shen and Probstein (1977)) and with experiments for bovine serum albumin in 0.15 M saline water at pH 7.4 showed excellent agreement. Possible limitations of the experimental data of Probstein et al. (1978) are that flux measurements were taken only over a narrow range of low solute concentrations.

Trettin and Doshi (1980a) have developed an integral method solution to the solute mass balance equation (Eq. (7)) for a parallel plate system using a variable non-linear concentration profile (Doshi et al. (1971)) in the boundary layer. Excellent agreement was found, for the case of a constant diffusion coefficient, between the calculated limiting flux values of the integral method solution and an exact numerical solution, where Eq. (7) was transformed to an ordinary differential equation by a method described by Shen and Probstein (1977). For a concentration dependent diffusion coefficient the integral method was satisfactory. By a comparison of their integral method solution with the film theory model of Michaels (1968) for constant fluid properties agreement was found for $F_g < 4$, where $F_g$ is the ratio of the gel concentration to the feed concentration. For higher values of $F_g$ the integral solution deviates from the logarithmical behaviour of the film model and predicts higher permeate flux rates. This would mean that extrapolation of experimental values in flux versus $\ln c_b$ plots may not be valid and may underpredict the gel concentration. Despite existing predictive differences, the same upsloping flux behaviour was observed when they compared Probstein's et al. (1978) integral method relationship, which is a special case of Trettin and Doshi's integral method solution, with the film model. However, experimental evidence for their theoretical investigation was not presented.

Trettin and Doshi (1980b) carried on in the development of a constant property integral method solution for ultrafiltration in an unstirred batch cell which agreed well with an exact solution but not with the film theory model. The deviation was more than 25% for all values of $F_g$ greater than 4, which is similar to the results obtained by Trettin and Doshi (1980a) for the parallel plate system. Agreement was found to be excellent between experimental data for bovine serum albumin over a wide concentration and pressure range in the unstirred batch cell and the integral model predictions, with the average error less than 3%, while the film
theory consistently underpredicts the experimental flux measurements. Therefore, the primary conclusion of their work was that the logarithmic permeate flux behaviour predicted by the film theory model is not encountered in the unstirred batch cell system.

Rautenbach and Holtz (1980) solved numerically the mass transfer equation in the boundary layer along the membrane for a laminar flow regime in a rectangular narrow channel with variable and constant physical properties. The measured viscosity and diffusion coefficients of their macromolecular solutions at isoelectric pH showed considerable dependence on concentration. Furthermore, the osmotic pressure of these substances was a non-linear function of concentration. Their ultrafiltration experiments agreed excellently with the numerical predictions for variable physical properties of dextran solutions, where the use of constant physical properties results in excessively high values for the flux. However, for the protein solutions (albumin and hemoglobin) the experimental data lay between the curves calculated by concentration dependent and constant physical properties. They suggested that the deviation could result from a difference between the isoelectric point and the point of zero charge of protein solutions of up to one pH unit. However, no experimental evidence was published.

Vilker et al. (1981) investigated theoretical and experimental concentration polarization in ultrafiltration of bovine serum albumin in saline water with an unstirred cell. The unsteady, one-dimensional concentration–diffusion equation

$$\frac{\partial c}{\partial t} + \nu \frac{\partial c}{\partial y} = \frac{\partial}{\partial y} \left( D \frac{\partial c}{\partial y} \right)$$

was solved by regular perturbation theory for constant density and diffusion coefficient. The general features of the ultrafiltration model were consistent with experimental data for both the ultrafiltrate flux and for the concentration profile near the membrane surface. The occurred discrepancies were thought to be associated with the constant diffusion coefficient in the model (no concentration and pH dependence). The primary conclusion of their work was that under the experimental conditions no gel formation was observed, even for albumin ultrafiltration of isoelectric pH solutions with an applied pressure of 276 kPa. Rather the concentration polarization limits the flux driving force relation-
ship via an osmotic pressure mechanism ($|Δp| - |Δπ| = 0$), which has been confirmed experimentally. Their observation may not apply to other macromolecular solutions since they might display much lower osmotic pressures in concentrated solutions.

Aimar and Sanchez (1986) developed a semi-empirical model based on the film theory model and osmotic pressure model (see Section 3) considering the dependence of the mass transfer coefficient on permeate flux, which has already been proposed by Jonsson (1984) as a qualitative explanation of the existence of a limiting flux. Concentration dependent viscosity is used in the model, since the variation in diffusivity is demonstrated to be of less influence compared to the viscosity variation on the mass transfer coefficient for bovine serum albumin solutions (data used from Kozinski and Lightfoot (1972) and Phillips et al. (1976)). They compared the values of the limiting flux of their model predictions with experimental data from several authors for different solutes, module geometries and membranes after determining a few parameters depending on hydrodynamic conditions and solute properties first. Excellent agreement was found for all kinds of ultrafiltration experiments. Furthermore, a complete model is deduced which calculates the permeate flux from zero to the limiting value. Again, the experimental data was well represented by the model over the whole range of applied pressures. Therefore, the assumptions made in the model seem to be valid, especially the decrease of the mass transfer coefficient with increasing applied pressure (increasing membrane wall concentration) for the limiting flux case. They concluded that the limiting membrane wall concentration (for limiting flux) is a function of the bulk concentration, the hydrodynamic conditions and the physicochemical solution properties.

Gill et al. (1988) modelled the effect of viscosity on the concentration polarization in the entrance region of a thin rectangular channel module until gel-conditions are reached on the membrane surface. They mentioned that Shen and Probstein (1977) neglected the entrance length in their model by assuming that the gel concentration is present at the channel inlet. However, Gill’s et al. model could only predict that the gel concentration is reached after a shorter distance from the channel entrance when a concentration dependent viscosity is used rather than a constant viscosity. Quantitative predictions were limited because of several assumptions made in the model.
3. OSMOTIC PRESSURE MODEL

At typical ultrafiltration feed concentrations, macrosolutes have a negligible osmotic pressure and consequently osmotic effects are frequently ignored (Blatt et al. (1970), Porter (1972a)). However, with the effect of concentration polarization at the membrane surface the surface concentration could be of one to two orders of magnitude higher, where osmotic pressures may be important (see Table 1), as long as the boundary layer remains Newtonian and gelation or precipitation does not occur (Fane (1986)).

Kedem and Katchalsky (1958) derived the osmotic pressure model, which describes the permeate flux in relation to the osmotic pressure difference $\Delta \pi$ created by the concentration difference between the two sides of the membrane:

$$J = \frac{|\Delta p| - |\Delta \pi|}{\mu R_m}$$  \hspace{1cm} (11)

where $R_m$ is the membrane resistance, $\mu$ the solvent viscosity and $\Delta \pi = \pi(c_m) - \pi(c_p)$ with the concentrations $c_m$ and $c_p$ at the membrane surface and in the permeate, respectively. The osmotic pressure $\pi$ is often represented in terms of a polynomial.
\[ \pi = a_1 c + a_2 c^2 + a_3 c^3 \]  

(12)

where \(a_1\) is the coefficient in van't Hoff's law for infinitely dilute solutions and \(a_2, a_3\) represent the non-ideality of the solution.

Goldsmith (1971) who first applied the osmotic pressure model has shown that under circumstances where no gel layer is expected, that is for a low molecular weight polyethylene glycol (mean Mwt = 15500 Daltons) as the solute with a limiting concentration less than 10 wt.%e, an almost limiting flux can be obtained for laminar and turbulent flow conditions, whereby the \(J\) versus \(\ln c_b\) plot is linear for a given pressure drop \(\Delta p\).

Kozinski and Lightfoot (1972) examined low polarization ultrafiltration in a rotating disk geometry. They numerically integrated the concentration–diffusion equation (Eq. (7)) and the equations of motion (Eq. (8), (9)) subject to the osmotic pressure boundary condition and found that concentration polarization could account for experimentally observed reductions in permeate flux when ultrafiltering bovine serum albumin solutions.

Leung and Probstein (1979) developed an integral solution method based on the two-dimensional concentration–diffusion equation (Eq. (7)) and the osmotic pressure model with concentration dependent diffusivity and osmotic pressure for the ultrafiltration of bovine serum albumin in steady, plane laminar channel flow under low polarization conditions. The integral solution was checked with a finite difference solution for the case of a linear osmotic pressure–concentration relation and a constant diffusivity, where the agreement was excellent. Experimental ultrafiltrate fluxes were found to agree very well with the theoretically calculated values of the integral solution method using Vilker’s et al. (1981) non-linear osmotic pressure data and a diffusivity relation obtained by linearly interpolating the diffusivity values between the gel and dilute concentration limits.

Vilker et al. (1981) concluded from osmotic pressure measurements and ultrafiltration experiments of bovine serum albumin in an unstirred cell, that the permeate flux is limited by the osmotic pressure, since also no gel formation was observed under the applied operation conditions.

Wijmans et al. (1984) provided an interesting analysis of the osmotic pressure model. Assuming complete rejection \((c_p = 0)\) the osmotic pressure difference in equation (11) is approximated by

\[ \Delta \pi = \pi(c_m) = a c_m^n \]  

(13)
with \( n > 1 \). \( c_m \) itself is dependent on permeate flux according to the film model (Eq. (4)), which gives

\[
J = \frac{|\Delta p| - a c_b^n \exp (nJ/k_s)}{\mu R_m}
\]  

(Wijmans et al. 1984) demonstrated how the derivatives of Eq. (14) provide insight into the ultrafiltration process. The flux–pressure derivative

\[
\frac{\partial J}{\partial |\Delta p|} = \left[ \mu R_m + \frac{n}{k_s} (|\Delta p| - J \mu R_m) \right]^{-1}
\]

(15)

gives the asymptotes

\[
\frac{\partial J}{\partial |\Delta p|} \rightarrow (\mu R_m)^{-1} \quad \text{for} \quad |\Delta p| \rightarrow 0, \text{or} \quad |\Delta \pi| \rightarrow 0
\]

and

\[
\frac{\partial J}{\partial |\Delta p|} \rightarrow 0 \quad \text{for} \quad |\Delta p| \rightarrow \infty, \text{or} \quad |\Delta \pi| \gg J \mu R_m
\]

Thus the flux-pressure profile commences at low \( \Delta p \) with a slope similar to pure solvent flow and as \( \Delta p \) increases the slope declines and approaches zero at high pressure, which is similar to the gel-polarization model.

The flux–concentration relationship could be examined by rearranging Eq. (14), taking the logarithm and differentiating to give

\[
\frac{\partial J}{\partial \ln c_b} = - \left( \frac{1}{k_s} + \frac{1}{n \left( \frac{|\Delta p|}{\mu R_m} - J \right)} \right)^{-1}
\]

(16)

\[
= - k_s \left( 1 + \frac{\mu R_m k_s}{n |\Delta \pi|} \right)^{-1}
\]
which shows that when polarization is significant, that is,

\[ |\Delta p| \gg J \mu R_m \quad \text{or} \quad \frac{n |\Delta \pi|}{\mu R_m k_s} \gg 1, \quad \text{then:} \]

\[ \frac{\partial J}{\partial \ln c_b} \to -k_s \]

The same prediction for the limiting slope in a \( J \) versus \( \ln c_b \) plot is given in the gel-polarization model.

The limiting concentration \( c_{b,\text{lim}} \) for \( J \to 0 \) is obtained from equation (14) when

\[ \Delta p = \alpha c_{b,\text{lim}}^n = \pi(c_{b,\text{lim}}) \quad (17) \]

which gives an osmotic pressure equal to the applied pressure. This also implies that \( c_{b,\text{lim}} = f(\Delta p) \), an important difference from the gel-polarization model which predicts that \( c_{b,\text{lim}} = c_g = f(\Delta p) \).

Wijmans et al. (1984) concluded from their theoretical analysis that,

1. lower values for the membrane resistance \( R_m \) lead to a more pronounced osmotic pressure effect, that is flux limitation at lower applied pressures,
2. the membrane resistance has a diminishing importance as the osmotic pressure increases, that is as the bulk concentration increases, and,
3. the osmotic pressure limitation will be expected in ultrafiltration of medium macrosolutes \((10^4\text{–}10^5 \text{ Mwt})\).

Jonsson (1984) has analysed the ultrafiltration of dextran solutions (macrosolutes of medium molecular weight) and demonstrated that the osmotic pressure model could produce a semi-logarithmic plot of \( J \) versus \( \ln c_b \), which gives a linear relation at bulk concentrations above 7 wt.\%, intersecting the \( \ln c_b \)-axis at a concentration of about 37 wt.\%. However, Jonsson concluded that this is not the gel concentration, but the concentration at which the solution has an osmotic pressure equal to the applied pressure. Further evidence for the absence of a gel layer was obtained by direct observation of the hydrodynamic flow of the polarization layer.

Clifton et al. (1984) solved the two-dimensional concentration–diffusion equation with constant diffusivity, coupled to the momentum equation with variable viscosity for a hollow-fibre membrane geometry in order to
describe the growth of the concentration polarization layer along the membrane. The obtained integral solution was combined with the osmotic pressure model to yield a numerical solution, where the permeate flux, taken separately in several sections along the membrane bundle, was assumed to be completely limited by the osmotic pressure effect. Very good agreement between observed and calculated values was often found, but generally better for dextran T70 than PVP (polyvinylpyrrolidone) solutions. They proposed that this was due to a better representation of the viscosity variations by their concentration dependent viscosity relationship for the dextran solutions. Furthermore, for high bulk concentrations of PVP a distinct divergence between measured and calculated flux values was observed beyond a certain distance along the membrane. Clifton et al. (1984) also suggested that the polarization layer breaks down due to instabilities in the boundary layer after a certain point along the membrane, becoming thinner and thus allowing a more rapid mass transfer to take place resulting in the observed higher flux values. Such hydrodynamic disturbances have been observed directly by Madsen (1977) and Jonsson (1984) using a coloured macromolecular solute.

Choe et al. (1986) confirmed Jonsson's (1984) observations that in ultrafiltration with dextran solutions, the osmotic pressure is the only resistance which needs to be taken into account to explain the permeate flux decline.

4. RESISTANCE MODELS

Ultrafiltration performance can also be interpreted by a resistance-in-series relationship, which can be obtained from equation (2) by neglecting the osmotic pressure term,

\[ J = \frac{|\Delta p|}{\mu(R_m + R_s)} \]  

(18)

According to the filtration theory the resistance of polarized solids can be written as

\[ R_s = \alpha \frac{m_p}{A_m} \]  

(19)
where \( m_p \) is the mass of deposited particles, \( A_m \) the membrane area and \( \alpha \) the specific resistance of the deposit, which can be approximately related to the properties for spherical particles by the Carman–Kozeny relationship (Carman (1938))

\[
\alpha = \frac{180(1 - \varepsilon)}{\rho_p d_p^2 \varepsilon^3}
\]  (20)

where \( \varepsilon \) is the void volume of the cake, \( \rho_p \) the density of the particles and \( d_p \) the mean diameter of the particles. A general observation would be that the smaller the particles are then the greater the specific resistance will be. Equation (18), (19) and (20) apply equally well to particulate filtration (microfiltration) and colloidal ultrafiltration (Fane (1984)).

For dead-end (unstirred) filtration under constant pressure conditions, without any particle back-transport, \( R_s \) increases with time, because

\[
m_p = V c_b
\]  (21)

where \( V \) is the total volume filtered and \( c_b \) the bulk concentration, so that the combination of Eqs. (18), (19) and (21) with \( J = A_m^{-1} \frac{dV}{dt} \) gives by integration the well-known constant pressure filtration equation (in a form first suggested by Underwood (1926)).

\[
\frac{t}{V} = \frac{\mu R_m}{A_m |\Delta p|} + \frac{\mu \alpha c_b}{2 A_m^2 |\Delta p|} V
\]  (22)

Equation (22) yields a straight line on plotting experimental data of \( t/V \) versus \( V \) which allows determination of the specific resistance \( \alpha \) and the membrane resistance \( R_m \).

4.1 Gel-Polarization Resistance Model

Fane (1986) expressed the gel-polarization model in terms of resistances,

\[
J = \frac{|\Delta p|}{\mu(R_m + R_{bl} + R_g)}
\]  (23)
where $R_g$ is due to the gel layer at limiting concentration $c_g$, and $R_{bl}$ represents the resistance of the viscous, but non-gelled boundary layer. If the concentration polarization at the membrane surface increases from zero to the gel concentration due to an increase in pressure, $R_g$ is zero and the flux is pressure dependent. After the gel concentration is reached $R_g$ is established and any further increase in $|\Delta p|$ simply increases the thickness of the gel layer, and increases $R_g$, which predicts the pressure independent flux behaviour.

Chudacek and Fane (1984) used the filtration model to describe the dynamics of polarization in stirred or cross-flow ultrafiltration. The appropriate form of the resistance model (Eq. (18)) is

$$J = \frac{|\Delta p|}{\mu (R_m + R_{sd} + R_{sr})} \tag{24}$$

where $R_{sd}$ is the resistance which would be caused by deposition of all convectively transported solute (equivalent to $R_s$ in Eq. (19)), and $R_{sr}$ is the resistance removed by stirring or cross-flow. They assumed that the removal of solute (back-transport) is constant and equal to the convective solute transport at steady state ($= J_{ss} c_b$) so that Eq. (21) is not any more valid. Equation (24) becomes

$$J = \frac{|\Delta p|}{\mu \left[ R_m + \left( V/A_m - J_{ss} t \right) \alpha c_b \right]} \tag{25}$$

The quantity $J_{ss}$ can either be expressed by the film model relationship (Eqn. (4)) or by experimentally determined values of $J_{ss}$.

Chudacek and Fane's (1984) numerical solutions of the model using experimentally determined $\alpha$-values and measured $J_{ss}$-values, tended slightly to overpredict initial experimental flux values for their three solutes (bovine serum albumin, dextran T2000, Syton X30 silica) used. They explained the discrepancy as due to membrane-solute interactions where the incoming solute may obstruct the entrance pores of the membrane resulting in an additional resistance for which the model takes no account.
4.2 Boundary Layer Resistance Model

Wijmans et al. (1985) proposed the boundary layer resistance model for cross-flow ultrafiltration of dextran solutions where no gel-formation will occur. The basic principle of the model is the correspondence of the permeability of a concentrated solute layer for solvent flow and the permeability of a solute in a stagnant solution, as occurring during a sedimentation experiment. This relationship (Mijnlieff and Jaspers (1971)) can be described by

\[ p = \frac{\mu s}{c(1 - v_1/v_0)} \]  

where \( p \) is the permeability of a concentrated solute layer of concentration \( c \), \( v_0 \) and \( v_1 \) are the partial specific volumes of the solvent and the solute respectively and \( s \) is the sedimentation coefficient at concentration \( c \) usually described by

\[ \frac{1}{s} = \frac{1}{s_0} \left(1 + K_1 c + K_2 c^2 + K_3 c^3\right) \]  

where \( s_0, K_1, K_2 \) and \( K_3 \) are constants.

The permeability depends on the concentration and since there is a concentration profile in the boundary layer, the permeability will be a function of the coordinate \( y \) perpendicular to the membrane. Since, the hydrodynamic resistance of the boundary layer \( R_{bl} \) is generally defined as

\[ R_{bl} = \int_0^\delta r_{bl} \, dy \]  

where \( r_{bl} \) is the specific resistance of the boundary layer \( (r_{bl} = \alpha p_p (1 - \varepsilon)) \) and equal to the reciprocal of the permeability. Thus, the boundary layer resistance is

\[ R_{bl} = \int_0^\delta \frac{1}{p(y)} \, dy \]  

Substituting Eq. (26), (27) with \( c(y) = c_b \exp(Jy/D) \) into Eq. (28) and integrating over the boundary layer thickness \( \delta \), results in
\[ R_{bl} = \frac{D}{\mu s_0 J} \left( 1 - \frac{v_1}{v_0} \right) \left[ c_m - c_b + \frac{K_1}{2} (c_m^2 - c_b^2) + \frac{K_2}{3} (c_m^3 - c_b^3) + \frac{K_3}{4} (c_m^4 - c_b^4) \right] \]

where \( c_m \) is the solute concentration at the membrane surface, which could be determined via the film model relationship (Eq. (4)) if \( k_s \) is known.

Combining Eq. (18) with \( R_s = R_{bl} \), (30) and (4) for \( c_p = 0 \), the boundary layer resistance model is obtained in which the permeate flux is the single unknown parameter.

\[ J = \frac{|\Delta p|}{\mu \left( R_m + \frac{D}{\mu s_0 J} \left[ c_m - c_b + \frac{K_1}{2} (c_m^2 - c_b^2) + \frac{K_2}{3} (c_m^3 - c_b^3) + \frac{K_3}{4} (c_m^4 - c_b^4) \right] \right)} \]

with

\[ c_m = c_b \exp \left( \frac{J}{k_s} \right) \]

Thus, the flux can be calculated if the process conditions \( (\Delta p, R_m, c_b, k_s) \) and the physical properties of the solute–solvent system \( (s, D) \) are known. Wijmans et al. (1985), who calculated the mass transfer coefficient by applying the osmotic pressure model, after having proven that the resistance model and the osmotic pressure model are equivalent, obtained an excellent agreement between their calculated and experimental flux values.

Nakao et al. (1986) who examined the concentration polarization effect in unstirred ultrafiltration used the boundary layer resistance model adapted to the cake filtration theory to analyse the experimental flux behaviour of dextran and polyethylene glycol solutions. Therefore a step concentration profile and a time-independent concentration in the boundary layer without any solute back-transport were assumed. The simple model worked well in predicting the experimental flux behaviour but only with the need of several other experiments to obtain the necessary parameters.

Van den Berg and Smolders (1989) analysed the concentration polarization phenomena of bovine serum albumin during unstirred dead-end ultrafiltration by adapting the boundary layer resistance model in
combination with the one-dimensional unsteady state concentration–diffusion equation (Eq. (10)). Their numerical approach compared to Nakao's et al. (1986) model required no assumptions concerning the concentration at the membrane, the concentration profile or the specific resistance of the boundary layer. The model predictions agreed very well with the experimental data for all feed concentrations, membrane resistances (retentions) and applied pressures investigated.

5. ALTERNATIVE MODELS FOR COLLOIDAL FILTRATION

Colloidal dispersions have been subjected to unstirred, stirred and cross-flow ultrafiltration in both laminar and turbulent flow systems by many investigators. Flux predictions are frequently described by the widely accepted gel-polarization model of Michaels (1968),

\[ J = \frac{D}{\delta} \ln\left(\frac{c_m}{c_b}\right) = k_s \ln\left(\frac{c_m}{c_b}\right) \]  

(32)

where the diffusion coefficient can be calculated from the Stokes–Einstein relationship for diffusivity,

\[ D_0 = \frac{kT}{3 \pi \mu d_p} \]  

(33)

although this strictly applies only to dilute solutions, where \( k \) is the Boltzmann constant, \( T \) the absolute temperature, \( \mu \) the viscosity of the solvent, and \( d_p \) the particle diameter. The inference from Eq. (32) and (33) is that as particle size increases so flux decreases. However, as particle size increases from colloidal upwards, back-transport becomes non-diffusive and Eq. (32) no longer strictly applies.

Blatt et al. (1970) who have carried out ultrafiltration experiments with skimmed milk, polymer latex, and clay dispersions found that the film theory model expressed by Eq. (32) and (33) drastically underpredicts experimental permeate rates. These observations led them to the conclusion that either: (1) the back-diffusion of particles from the polarized layer is substantially augmented over that expected to occur by Brownian motion or, (2) the transmembrane flux is not limited by the hydraulic resistance of the polarized layer over any reasonable range of
layer thickness. Blatt et al. favoured the second explanation to be the more reasonable, since a cake formed by micron size particles has a relatively low specific resistance compared with a macromolecular cake.

Porter (1972a,b) who observed essentially the same flux augmentation compared with the film model predictions in more than 40 colloidal dispersions he studied, disputed Blatt's et al. (1970) hypothesis and argued that, if the polarized layer is not the limiting factor, the flux should be independent of the bulk concentration and proportional to the applied pressure. However, he observed the opposite to be true, decreasing flux with increasing bulk concentration. Therefore, Porter hypothesized the augmented back-transport of the lateral (radial) migration of particles known as the *tubular-pincho effect* (Segré and Silberberg (1962)) to be relevant for the enhanced flux. But no quantitative predictions of the enhanced mass transfer of particles away from the membrane surface could be made at that stage.

Henry (1972) pointed out the long transient flux decline in the ultrafiltration of colloidal dispersions. He found in general that the dependence of the permeation rate on fluid rate (or shear rate) is greater in colloidal ultrafiltration than that predicted by the film theory model. This finding led Henry to the conclusion, like Porter (1972a,b), that an additional particle back-transport mechanism is present in such systems.

5.1 Inertial Migration Model (Tubular-Pinch Effect)

Segré and Silberberg (1962), working with dilute dispersions of rigid, spherical, neutrally buoyant particles (mean diameters from 0.32 to 1.71 mm) transported along in Poiseuille flow through a non-porous tube, were the first to publish their observations of the *tubular-pincho effect*, whereby the particles are subject to radial forces which migrate them away both from the tube wall and the tube axis, reaching a certain equilibrium position at about 0.6 tube radii from the axis, irrespective of the tube entrance radial position of the spheres. The origin of these effects were found to lie in the inertia of the fluid. Segré and Silberberg developed an empirical equation to correlate their data for the radial migration velocity $v_r$,

$$v_r = 0.17 \bar{u} \text{Re}_t \left( \frac{d_p}{d_t} \right)^{2.84} \frac{r}{d_t/2} \left( 1 - \frac{r}{r^*} \right)$$

(34)
where \( \bar{u} \) is the average fluid axial velocity, \( \text{Re}_t = \frac{\bar{u}^2 R}{\nu} \) the Reynolds number, \( d_p \) the mean particle diameter, \( d_t \) the tube diameter, \( r \) the radial coordinate, and \( r^* \) the equilibrium radial position of the particle which decreases as \( d_p/d_t \) increases. Their observations have spawned a number of theoretical and experimental studies explaining and quantifying this effect (Brenner (1966)).

Cox and Brenner (1968) were the first to attempt a full theoretical treatment of the problem of rigid spheres in laminar flow freely rotating and translating parallel to the tube axis which gave a satisfactory fundamental explanation of the radial migration phenomenon in a tube of finite radius. They obtained the first-order solution of the Navier–Stokes equation including the inertial terms and converted the lateral force, required to maintain the spheres at a fixed \( r \), into an equivalent radial migration velocity by applying Stokes’ law. For the neutrally buoyant case they found

\[
v_r = 0.5 \bar{u} \text{Re}_t \left( \frac{d_p}{d_t} \right)^3 f \left( \frac{r}{d_t} \right)
\]

where \( f (r/d_t) \) is a function of the radial position of the particle in the tube. Equation (35) is of the same form as the empirical one (Eq. (34)) from Segré and Silberberg (1962).

Vasseur and Cox (1976) have analysed the lateral migration, \( v_l \), of a solid spherical particle in laminar channel flow using the method developed by Cox and Brenner (1968). For the case of a neutrally buoyant sphere they obtained

\[
v_l = 4 \bar{u} \text{Re}_{ch} \left( \frac{d_p}{2h} \right)^3 f (\beta)
\]

where \( \text{Re}_{ch} = \frac{\bar{u}^2 h}{\nu} \), \( h \) is the half channel height and \( f (\beta) \) depends on the particle position in the channel.

In general, virtually all studies of the migration phenomena, whether theoretical or experimental (Brenner (1966)) arrive at the following form for the migration velocity expression (Green and Belfort (1980))

\[
v_l = K \bar{u} \text{Re} \left( \frac{d_p}{d_t} \right)^a f (r)
\]
where \( d_t \) is the tube diameter or equal to \( 2h \) the channel height, \( \text{Re} = \text{Re}_t \) or \( \text{Re}_{ch} \), the value of \( n \) lies between 2.84 and 4, and \( f(r) \) is a function of the equilibrium position of the particle in the tube or channel. According to Eq. (37), the lateral (radial) migration velocity depends strongly on the average axial fluid velocity and the particle size.

Madsen (1977) semi-empirically analysed the problem of particle migration with the use of the particle movement formula of Cox and Brenner (1968) and the film theory model. He found that experimental permeate rates for cheese whey and hemoglobin were underpredicted by a factor of 50 by the particle migration model. His primary conclusion was that particle migration can only account for a part of the high permeate flux observed in ultrafiltration of colloidal dispersions.

Green and Belfort (1980) developed a model for colloidal ultrafiltration incorporating the lateral migration effect into the standard filtration theory by using Segré and Silberberg's (1962) empirical relation for the case of flow in a non-porous duct. It was assumed that the permeation drag force and the migration lift drag force could be vectorially added. Further, it was assumed that the thickness of the (immobile) cake layer adjusts itself to the point where the convective flow of particles towards the membrane is balanced by the lift velocity away from the membrane. In order to obtain this condition Altena et al. (1983) found a value for the cake layer thickness of 0.7 times the channel height. They commented that whether such a large value for the cake layer is realistic, is questionable.

However, Green and Belfort's (1980) model predictions for the steady state flux were within the right order-of-magnitude compared with Porter's (1972) thin channel ultrafiltration data of styrene–butadiene polymer latex. They mentioned that Madsen (1977) applied the lift velocity expression in an unsatisfactory manner, by evaluating the radial migration velocity at the original tube radius \( d_t/2 \) and not, as it should be for a steady state flux, at \( d_t/2 - \delta_c \), where \( \delta_c \) is the cake layer thickness. They demonstrated that this slight difference in effective tube radius, neglected by Madsen, provided the margin between correct and grossly incorrect predictions of steady state flux.

Altena et al. (1983) and Altena and Belfort (1984) studied theoretically the effect of lateral migration of spherical rigid neutrally buoyant particles moving in a laminar flow field in a porous channel. They extended Cox and Brenner's (1968) analysis for particle motion in a non-porous duct to include the effect of the wall porosity. Altena and Belfort (1984) showed that the assumption of a no-slip condition for the tangential flow
at the membrane wall and the negligibility of the disturbance velocity within the porous wall are good approximations for typical conditions found in hyperfiltration and ultrafiltration flow channels. They have analysed that the inertially induced velocity (tubular-pinch effect) and the permeation drag velocity due to convection into the porous wall can be vectorially added when $\lambda \equiv \text{Re}_p \kappa^2$, where $\lambda$ is the ratio of wall permeation velocity to the mean axial fluid velocity, $\text{Re}_p$ is the particle Reynolds number based on the mean axial velocity and the particle radius and $\kappa$ is the ratio of particle radius to channel height. If $\lambda << \text{Re}_p \kappa^2$, the particle trajectories converge at an equilibrium position reminiscent of flow in a non-porous duct. If $\lambda >> \text{Re}_p \kappa^2$, the equilibrium position moves closer to the porous wall and finally coincides with it resulting in particle capture. Further, $\lambda << 1$, as also required by the theory, is indeed a practical condition of pressure-driven membrane processes such as nano- and ultrafiltration. However, the assumption above should be invalid when a fouling (or cake) layer is present, since the slip coefficient could be several orders of magnitude higher.

Altena et al. (1983) evaluated theoretically the range of conditions in which the tubular-pinch effect or lateral migration concept is effective. Their results of a calculation of permeation to lift velocity, i.e. $J/\nu_L$, as a function of particle size for three ultrafiltration configurations is given in Table 2. As a general rule they claimed that particles with a radius larger than 1 $\mu$m will be affected by the lift velocity and submicron particles will tend to be transported to the membrane wall by permeation drag. The effect has been confirmed by Bauser et al. (1982) during ultrafiltration of blood in a hollow-fibre membrane, assuming that the red cells behave in whole blood as a dilute solution with a cell size of ~8 $\mu$m in diameter and were observed to move away from the membrane surface. Although, the theory of Altena et al. (1983) and Altena and Belfort (1984) strictly applies only to single particle systems or very dilute dispersions, the experiments of Bauser et al. (1982) indicate a lateral migration in qualitative agreement with the observations of Altena et al. (1983). If their theoretical observations are correct the importance of lateral migration in colloidal ultrafiltration appears to have been overestimated by Porter (1972a,b).

Indirect experimental evidence has been given by Fane (1984), who has studied four sizes of silica-based particulates ranging from 25 nm, 125 nm, 5 $\mu$m to 20 $\mu$m in diameter in an unstirred and stirred ultrafiltration cell. He obtained under stirred conditions (see Fig. 3) that, as the
TABLE 2
Calculations of the ratio of permeation and lift velocity as a function of particle size for a given channel geometry and axial centre line velocity ($J = 4 \times 10^5 \text{ m s}^{-1}$) (Alterna et al. (1983))

<table>
<thead>
<tr>
<th></th>
<th>$h$ or $d_f/2$ (mm)</th>
<th>$u_m$ (m s$^{-1}$)</th>
<th>$d_p/2$ (μm)</th>
<th>$J/v_l$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hollow-fibre</td>
<td>0.1</td>
<td>0.1</td>
<td>0.1</td>
<td>$4 \times 10^4$</td>
</tr>
<tr>
<td></td>
<td>1.0</td>
<td></td>
<td>40</td>
<td></td>
</tr>
<tr>
<td></td>
<td>10.0</td>
<td></td>
<td>0.04</td>
<td></td>
</tr>
<tr>
<td>Flat-plate</td>
<td>1.0</td>
<td>0.5</td>
<td>0.1</td>
<td>$2 \times 10^5$</td>
</tr>
<tr>
<td></td>
<td>1.0</td>
<td></td>
<td>200</td>
<td></td>
</tr>
<tr>
<td></td>
<td>10.0</td>
<td></td>
<td>0.2</td>
<td></td>
</tr>
<tr>
<td>Tubular</td>
<td>10.0</td>
<td>1.5</td>
<td>0.1</td>
<td>$2 \times 10^6$</td>
</tr>
<tr>
<td></td>
<td>1.0</td>
<td></td>
<td>$2 \times 10^3$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>10.0</td>
<td></td>
<td>2</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 3. Influence of particle size on permeate flux in aqueous dispersions of 2.5 vol% and 10 vol% particle concentration (Fane (1984)). (Lines are for illustration purposes only).
particle size increases from 25 nm to 20 μm, the flux passes through a minimum at about 0.1 μm particle size due to the fact that the polarization control changes from diffusive (decreasing with particle size) to non-diffusive (increasing with particle size), such as lateral migration and/or scour effects (Fane et al. (1982)).

5.2 Shear-Induced Hydrodynamic Convection Model

Madsen (1977), Green and Belfort (1980), Altena et al. (1983) and Altena and Belfort (1984) have found from their hydrodynamic calculations that the inertial lift velocity is often less than the permeation velocity in typical cross-flow filtration systems. As a result, a concentrated layer of deposited particles is formed on the membrane surface. If this cake layer would built up indefinitely, it would plug the tube or channel, but this is not observed in practice. Instead, Blatt et al. (1970) have hypothesized that the cake layer accumulates only until the hydrodynamic shear exerted by flow of dispersion causes the cake to flow tangentially along the membrane surface at a rate which balances the deposition of particles. Although Porter (1972a,b), Henry (1972), and others have reported an increase in the permeation flux with increasing tangential shear, indicating that a high shear rate is effective in reducing the cake layer thickness, this process is not well understood (Davis and Leighton (1987)).

Leonard and Vassilieff (1984), using the method of characteristics, solved the unsteady, two-dimensional convective equation by neglecting the diffusion term (no diffusive particle transport) for the axial movement of deposited particles along a membrane surface. They described the simultaneous convective deposition of particles onto the membrane and the sweeping of this cake layer in response to the fluid shear along the membrane surface where it is discharged with the fluid at the exit of the filtration device. Leonard and Vassilieff were able to obtain analytical solutions for the developing cake layer thickness over time until steady state is reached by assuming that the velocity profile in the vicinity of the cake layer is linear, the permeate flux is invariant with time and position along the membrane and the cake layer is treated as a Newtonian fluid with the same effective viscosity as the bulk dispersion. Their simple convection model has been used to predict the steady state performance of plasmapheresis devices, yielding good correspondence with experimental observations.
Davis and Birdsell (1987) presented a steady stratified laminar flow model which relaxes some of the simplifications imposed by Leonard and Vassilieff (1984). In particular, fully developed parabolic velocity profiles were determined for both the dispersion and the cake layer, the channel pressure and the permeate flux were allowed to vary along the channel, and the cake layer was assigned a concentration-dependent effective viscosity higher than that of the bulk dispersions. They developed out of the non-dimensional Navier–Stokes equation, by neglecting the transient and convective inertia terms, and the continuity equation the solution for fully developed flow in a two-dimensional channel, which yields parabolic flow profiles in both the dispersions and the cake layer. Although the governing flow equations were developed for the flow of the dispersions, they were also valid for the cake layer, provided that the viscous terms are multiplied by the viscosity ratio $\mu = \mu_c / \mu_b$. Additionally, the steady state differential mass balances for the bulk material (liquid plus solids) and for the solid material flowing through the channel served as auxiliary equations in the determination of the cake layer thickness, the pressure drop, and the change of the pressure drop over the channel downstream distance, all as a function of the longitudinal distance from the channel entrance, which resulted further in the permeation flux. For dilute dispersions, that is $\delta_c / h << 1$ and $R_c / R_m << 1$, they obtained an analytical solution for the steady state cake thickness similar to the result found by Leonard and Vassilieff (1984). For non-dilute dispersions a numerical solution of the governing model equations was obtained.

Davis and Birdsell's (1987) experimental data for the acrylic latex dispersions (150–212 μm particle diameter, so that the cake layer could be readily observed) followed the trends predicted in their theory quite well. The cake layer thickness increased with increasing down-channel distance, transmembrane pressure drop, and bulk concentration, whereas it decreased with increasing cross-flow velocity owing to the increasing shear stress exerted on the cake layer. However, the quantitative agreement between the model predictions and the experiments were also good, although the theory underpredicts the cake layer thickness observed in some of the experiments. They explained this as due to the fact that a stagnant cake sublayer could be present below the flowing layer or that the effective viscosity of the cake was somewhat higher than the value used in making the model predictions.

However, the most significant limitation in both, Leonard and Vassilieff's and Davis and Birdsell's, models results from the treatment of
the cake layer as a Newtonian fluid with a particle concentration and a corresponding shear viscosity that must be assigned. While some of the physics governing particle transport may be correct in these models, they fail to predict the structure of the flowing particle layer (Romero and Davis (1988)).

5.3 Shear-Induced Hydrodynamic Diffusion Model

During the cross-flow filtration of particle dispersions a concentrated particle layer will build up on the membrane surface due to the convective fluid flow towards the membrane. The particle layer thickness increases until the cross-flow, which induces a shear stress upon the layer, is large enough so that the outer particles experience re-entrainment leading to an expansion (or dilution) of the cake layer. The re-entrainment arises from particles tumbling over one another as the layer is sheared. These particle-particle interactions result in displacements of particles from the time averaged bulk flow stream lines. The effect of this shear-induced motion is the net migration of particles in the direction of decreasing particle concentration, where the mean migration velocity proportional to the concentration gradient, as described in more detail by Leighton and Acrivos (1987a).

Thus the effect may be described as a diffusion process, which is termed "shear-induced hydrodynamic diffusion" by Davis and Leighton (1987). However, this should not be confused with Brownian diffusion, which results from interactions between particles and surrounding fluid molecules and is present even in the absence of shear, whereas the former is a result of particles interacting with other particles and is only observed in the presence of shear. Since this phenomenon occurs in purely viscous systems it is not thought to be associated with inertia (Davis and Leighton (1987).

Eckstein et al. (1977) were the first who experimentally determined the magnitude of a shear-induced coefficient of self-diffusion of a dispersion of neutrally buoyant spherical particles in a linear shear field using a concentric cylinder Couette device. A single radioactively labelled sphere was placed in a slurry of otherwise identical spheres to determine its radial position (or rather, as an effect of the self-diffusion process, its radial migration) throughout the experiment by a radiation detector. The self-diffusion coefficient was calculated by means of random-walk theory, using the ergodic hypothesis. Two sizes of large polystyrene particles,
3175 μm and 1054 μm mean diameter, were used, to ensure better observation of the marked particle and to make sure that Brownian and electrokinetic forces were negligible. Eckstein et al. found that their calculated values of the self-diffusion coefficient were not of high accuracy owing to the nature and size of the experimental errors, but correct within a factor of two. However, the self-diffusion coefficients showed the trend of increasing linearly in the range of 0 to 20% particle volume fraction, and to be constant in the concentration range of 20 to 50%.

Leighton and Acrivos (1987a) carried out similar experiments but only measured the transit time of a marked particle immersed in the dispersions to complete a circuit of a Couette device, since they developed an utilization method, based on the unsteady two-dimensional density diffusion equation, which obviated the need of measuring the radial particle position in the Couette gap. This greatly simplified the experimental apparatus compared to Eckstein’s et al. (1977) and as a consequence reduced the experimental errors. Leighton and Acrivos (1987a) compared their measured self-diffusion coefficients (for particle volume fractions of 5 to 40%) in dispersions of 645 μm and 389 μm acrylic spheres with that observed by Eckstein et al. (1977). They found that the self-diffusion coefficient is proportional to the shear rate and the square of the particle radius and to be an increasing function of particle volume fraction φ, approximately equal to 0.5φ² at low concentrations, which is in contrast to the linear dependence for low concentrations reported by Eckstein et al. (1977). However, for particle volume fractions above 20% the self-diffusion coefficients deviated rapidly from the constant values of Eckstein et al. (at φ = 40 vol.% approximately 5 times). Leighton and Acrivos (1987a) demonstrated that Eckstein’s et al. (1977) experiments at high concentrations were limited by the presence of the Couette walls, since their particle diameter to gap-width ratio was only 1/8 compared to 1/32 of Leighton and Acrivos.

Leighton and Acrivos (1987b) demonstrated in the course of their experimental study of the behaviour of concentrated dispersions (30–50 vol.%) of neutrally buoyant polystyrene particles (46 μm and 87 μm mean diameter) in Newtonian fluids using the Couette device, that the short-term viscosity-increase and the long-term viscosity-decrease phenomenon observed in the Couette gap is due to particle migration across the gap-width and out of the sheared gap respectively, forced by the shear-induced diffusion mechanism. Their experiments enabled them to infer the values of the “effective” diffusivity, which consists of the sum of the
random self-diffusion (Eckstein et al. (1977) and Leighton and Acrivos (1987a)) and the non-random drift process (migration of particles along concentration gradients) occurring in concentrated dispersions of non-uniform composition. Leighton and Acrivos (1987b) determined that effective diffusivity for a particle volume fraction of 40% was approximately 5 times higher than the measured self-diffusion coefficient in their related paper from 1987a and approximately 25 times higher than the corresponding data of Eckstein et al. (1977).

Leighton and Acrivos (1986) supported their previous analyses (Leighton and Acrivos (1987a,b)) of the shear-induced diffusion mechanism by showing that these are able to describe the viscous re-suspension of a settled layer of rigid spherical particles when it is subjected to shear flow. They balanced the downward flux of particles due to gravity with an upward flux arising from the shear-induced particle diffusion which yields an (increased) equilibrium height of the settled layer where re-suspension starts. Predictions of this equilibrium height were shown to be in only approximate agreement with experimental observations, probably due to errors in the determination of the shear stress corresponding to the observed equilibrium height.

Zydney and Colton (1986) have proposed a concentration polarization model based upon the film theory model and the classical Lévêque solution for mass transfer. They incorporated a shear-induced hydrodynamic diffusivity based on the experiments of Eckstein et al. (1977) in place of the Stokes-Einstein particle diffusivity. They found $D = 0.03 \alpha^2 \dot{\gamma}$, where $\alpha$ is the particle radius and $\dot{\gamma}$ the wall shear rate, as an approximate fit to the data, valid for particle volume fractions greater than 20%. Zydney and Colton (1986) found that their model predictions for the length averaged flux match the experimental data of twelve different solute studies with a greater degree of accuracy than previous plasmapheresis models. The model seemed to be especially accurate for deformable particles such as red blood cells, as shown by a comparison of seven other models with the experimental whole blood data of Porter (1972b). However, Davis and Leighton (1987) found this agreement somewhat surprising since the diffusivity of Eckstein et al. (1977) has been shown by Leighton and Acrivos (1987a) to be too low. They proposed two explanations for this: the existence of a non-linear velocity profile across the cake layer which was not employed in the model, and possibly a lower shear-induced diffusivity for deformable particles in concentrated dispersions.
Davis and Leighton (1987) modelled the transport of a concentrated layer of rigid spherical particles along a flat porous wall under laminar flow conditions, where the assumption of a constant viscosity fluid with linear velocity profile in the layer is relaxed. At steady state, the lateral migration of particles away from the particle layer due to a shear-induced hydrodynamic diffusion mechanism was balanced by the convective flux of particles toward the porous wall owing to fluid flow into the wall. Together with the momentum balance they were able to predict, at any axial position along the filter, the non-linear particle concentration and velocity profiles and the axial excess particle flux (volumetric particle flow rate per unit filter width) in the cake layer, after assuming a local value for the permeate flux. A criterion for predicting whether a stagnant particle layer will form on the membrane surface or not, was found as well. However, no experimental verifications were made for the local model.

Romero and Davis (1988) extended the local model of Davis and Leighton (1987) to describe the axial variation of the particle layer thickness and its structure (stagnant and flowing part) and the permeate flux, all at steady state. They developed a solution technique by which the local model is applied stepwise along the filter, in conjunction with standard filtration theory, in order to generate a global model. Romero and Davis (1988) compared the dimensionless length-averaged permeate flux of a numerically calculated exact solution and a simplified analytical form, along with the predictions of Zydney’s and Colton’s (1986) model for various values of bulk concentration, as a function of the dimensionless filter length \( L/x_{cr} \) (\( x_{cr} \) is the critical point downstream where a stagnant cake layer starts to form). The models agreed quite closely for long filters, but both the analytical asymptotic solution and the model of Zydney and Colton (1986) failed for filters operating below or near the critical point, since the assumption of \( L \gg x_{cr} \) was implicit in both cases. Despite the assumptions made in Zydney’s and Colton’s (1986) model, as discussed earlier, Romero and Davis (1988) explained that these may have compensating effects on the predicted permeate flux. However, no quantitative comparison of their model with experimental data have been made, since the fairly good agreement (with moderate scatter) of Zydney’s and Colton’s (1986) model predictions for various particulate and cell dispersions was obtained.

Romero and Davis (1990) further developed their previous global steady state model by incorporating a time-dependent term in the micro-
scopic two-dimensional particle mass balance. Then the resulting partial
differential equations were solved over time and axial distance using the
method of characteristics for the case of development of the flowing layer
and for the subsequent formation of the stagnant layer. They discussed
their model predictions of the length-averaged permeate flux versus time
for the variation of several parameters like, shear rate, bulk concentra-
tion, particle size and transmembrane pressure drop.

Pearson and Sherwood (1988) have outlined a continuum mechanics
approach to the modelling of cake filtration under cross-flow conditions,
for a wide class of assumptions concerning the dispersion and cake
rheology and lateral shear-induced diffusivity of particles. A comparison
with experimental results from Fordham and Ladva (1989) showed that
the model predictions agreed well with the theory in the quasi-static
filtration regime (early stages of filtration), but for the dynamic regime
(steady state filtration) the predictions of the convection–diffusion bal-
ance model, which follows the need of a cake growing axially as $x^{1/3}$
(Zydney and Colton (1986)), were not conformed with the experimental
results, that is cake height was independent of $x$, independence of time
was inconsistent with a simultaneous independence of axial distance $x$,
and boundary conditions at “infinity” (i.e. for into the bulk dispersion)
could not be satisfied.

Davis and Sherwood (1990) developed a similarity solution like the
approach taken by Shen and Proebstein (1977) and Trettin and Doshi
(1980a) for ultrafiltration of macromolecular solutions, based upon the
two-dimensional concentration-diffusion equation (where the axial con-
vection term is retained in contrast to Romero and Davis’ (1988, 1990)
model), the continuity equation and the momentum equation governing
the steady-state concentration-polarization boundary layer of fine parti-
cles, under conditions where a thin stagnant layer of particles deposited
on the membrane surface provides the controlling resistance to filtration.
The analysis employed the same concentration-dependent shear viscosi-
ties and shear-induced hydrodynamic diffusivities based on empirical
correlations of rigid spheres used by Davis and Leighton (1987) and
Romero and Davis (1988, 1990). Davis and Sherwood (1990) have dem-
onstrated that the predictions of the approximate solution of Davis and
Leighton (1987) and Romero and Davis (1988, 1990) are exact in the
dilute limit and agree to within a few percent for non-dilute dispersions,
compared with the predictions of their numerical similarity solution for
the permeate flux as well as the concentration and velocity profiles.
Romero and Davis (1991) carried out cross-flow microfiltration experiments in a rectangular channel unit under laminar flow conditions for the case of membrane-limited filtration with negligible flux decline, in order to observe the stagnant particle layer thickness profiles, and in a commercial tube-bundle under conditions for which a moderate flux decline will occur depending on both the membrane and cake layer resistance. They compared their experimental data with their previously developed shear-induced hydrodynamic diffusion models (Romero and Davis (1988, 1990)). The particle layer thickness profiles at steady state agreed reasonably well with the (Romero and Davis (1988)) predictions of the model and the transient data of particle layer build-up was also in good agreement with the theoretical predictions (Romero and Davis (1990)), which are essentially the same as those for dead-end filtration. For the tube-bundle, the transient and steady-state permeate flux agreed with the predicted ones under varying operating conditions. Romero and Davis (1991) permeate flux results, which achieved an average absolute value of the deviation between theory and experiment of 11% and 27% for the rectangular channel experiments and the tube-bundle experiments respectively, without any adjustable parameters, provided encouraging support for the model of shear-induced hydrodynamic diffusion, since also the predicted flux values of alternative models like Zydney’s and Colton’s (1986) and Green’s and Belfort’s (1980) were one or two orders-of-magnitude lower than those observed for the conditions of Romero’s and Davis’ experiments.

However, recently Stamatakis and Chi Tien (1993) argued that there is no direct experimental evidence for the presence of a stagnant and flowing cake, nor can their physical structure and properties be independently measured. Mackley and Sherman (1992), who performed direct in situ observations of the cake deposition process during cross-flow filtration, found no evidence for particle back diffusion or flux of particles away from the cake surface (see Section 5.6), which is in contradiction to Davis’ et al. model approaches.

5.4 Erosion Models

5.4.1 Scour Model

Fane et al. (1982) postulated an alternative mechanism for the cross-flow ultrafiltration of larger solids based on the scour or erosion control of the polarized layer by the tangentially flowing feed dispersions. The
scour model is based on the analogy between dispersion flow across a filter cake and the motion of a sediment-laden stream over a layer of settled sediment. Lateral migration may have a role, but was not required by the model.

The rate of convective solids transport towards the membrane is balanced by the rate of scour,\[ J_c b = - \frac{d\delta_c}{dt} c_c \] (38)

where \( \delta_c \) is the cake thickness and \( c_c \) the cake concentration.

According to Raudkivi (1967) the rate of scour of sediment is proportional to the shear rate which is a function of the cross-flow velocity \( u_b \). Thus,

\[ \frac{d\delta_c}{dt} = - K_e \dot{\gamma} \] (39)

where \( K_e \) is the erosion coefficient varying with solids concentration, size and nature (discrete, flocculating, etc.). Combining Eq. (38) and (39) and assuming a power-law relationship for \( K_e \),

\[ K_e = k'_e c_b^\gamma \quad \text{with} \quad \gamma \leq 1 \] (40)

gives

\[ J = k'_e c_b^{\gamma-1} c_c u_b^m = k_e c_b^n u_b^n \] (41)

where the exponent \( n = \gamma - 1 \leq 0 \) and \( m = 1 \) for laminar flow.

Fane et al. (1982) demonstrated that the predicted log–log relationship between flux and concentration effectively fits the data for a range of dispersions containing rigid and deformable solids and droplets in various membrane systems. However, the scour model is more intuitive rather than rigorous and a more quantitative model would need to establish a theoretical basis to Eq. (38) and (39). Furthermore, the effect of particle size on scour and flux is not stated in the model, but it could be inferred from analogy with momentum transfer (Raudkivi (1967)) that scour and hence flux should increase with particle size.
5.4.2 Turbulent Burst Model

The re-entrainment of particles from a cake or fouling layer has been ascribed to the occurrence of turbulent bursts which sweep down into the laminar sub-layer and remove the fouling layer from a small area of the wall.

The deposition and re-entrainment of suspended particles, from a turbulent stream onto a smooth impermeable wall, has been studied in great detail by Cleaver and Yates (1973, 1976). They suggested that deposited particles could be re-entrained by the updraft generated during the gentler sweep process. The turbulent bursts occurred at random over the surface of the wall, each burst removing the fouling layer from a fraction \( \beta \) of the surface. Cleaver and Yates (1976) found the bursts to occur on average at time intervals of \( \theta \), which yield the empirical relationship

\[
\theta \propto 100 \frac{\mu}{\tau} \quad (42)
\]

where \( \mu \) is the solvent viscosity and \( \tau \) the wall shear stress.

Gutman (1977) modelled the change of permeate flux performance with time of reverse osmosis membranes which were fouled by suspended and colloidal material in the feed stream. He determined the net rate of fouling of the membrane surface to be equal the difference of the rate of deposition and re-entrainment, caused by the turbulent burst mechanism,

\[
\frac{dm}{dt} = r_d - r_e \quad (43)
\]

where \( m \) is the mass of fouling per unit area of surface, \( r_d \) the rate of transport of fouling material towards the membrane surface estimated from the Reynolds flux relationships,

\[
r_d = c_b (k_s + J/2) \quad (44)
\]

and \( r_e \) the rate of re-entrainment given by

\[
r_e = \frac{m\beta}{\theta} \propto \frac{m\beta\tau}{100\mu} = \frac{B}{\rho} \tau m \quad (45)
\]

where \( B/\rho = \beta/(100 \mu) \).
He summed up with an equation for the permeate flux as a function of time

\[
\frac{J}{J_f} = 1 + \frac{\alpha c_b}{B u_b} \left( 1 - \exp \left( \frac{-\left( \frac{J}{J_f} - 1 - J B u_b t \right)}{1 + \frac{\alpha c_b}{B u_b}} \right) \right)
\]

(46)

for the flux of the fouled layer \( J_f > 2k_s \), where \( \alpha = K/R_m \) with \( K \) the hydraulic resistance of the fouling layer per unit mass. Gutman (1977) compared his model predictions with experimental results of the fouling of tubular reverse osmosis modules in turbulent flow by sewage effluent after determining the parameters \( B \) and \( \alpha \) (or \( K \)). The final flux values, \( J_{\text{lim}} \) under different operating conditions were predicted quite accurately, but the rates of flux decline were less accurate.

However, Yung et al. (1989), who experimentally investigated the role of turbulent bursts in particle re-entrainment in aqueous systems, suggested that turbulent-burst activity is insignificant in re-entrainment of deposited particles completely submerged within the viscous layer, since the lifting forces exerted on the particles during the bursting activity were not as strong as proposed and the rate of re-entrainment is not directly proportional to the frequencies of the bursting action (see Eq. (42)), as stated in the Cleaver and Yates (1976) theory respectively.

5.5 Friction Force Model

It is generally accepted that the shear stress associated with tangential flow is responsible for keeping cake growth to a minimum (Green and Belfort (1980), Fane et al. (1982), Leonard and Vassilieff (1984)), while the action of that shear stress is not clearly defined (Blake et al. (1990). Another approach to cross-flow filtration is the friction force model which is based upon the drag force \( F_y \) induced by the convective flow towards the membrane and the drag force \( F_x \) induced by the cross-flow, originally proposed by Ebner (1981).

Figure 4 shows the hydrodynamic conditions in idealized form: the flow around a spherical particle consists of a plug flow in the membrane orthogonal direction and a linear velocity profile parallel to the membrane. The mean membrane-parallel velocity for the particle is given at a wall
distance of \(d_p/2\). For the given velocity profiles the mean velocity normal to the membrane is equal for small and big particles, but the mean velocity parallel to the membrane increases with increasing particle diameter. As a consequence of the assumptions of the friction force model, large particles are calculated to move at a lower membrane-parallel flow velocity than small particles, which indeed is experimentally observed by Fane (1984), Fischer and Raasch (1986) and Blake et al. (1990).

Ebner (1981) showed by balancing the two drag forces that the dynamic pressure of the liquid stream flowing parallel to the membrane surface must be greater than the product of the ratio between the largest \(Q\) and the smallest \(q\) cross-section of the particles to be filtered (equal 1 for spheres), the static (applied) pressure and the frictional coefficient of the particles relative to the membrane, to prevent the deposition of particles onto the membrane surface:

\[
\frac{1}{2} \rho \bar{u}^2 > \frac{Q}{q} \Delta p f
\]  

(47)

Fischer and Raasch (1986) who investigated the particle deposition at the filter medium in cross-flow filtration used a slightly different schematic representation, where a single spherical particle of diameter \(d_p\) is embedded in a sieve mesh of mesh size \(d_m\) being smaller than \(d_p\). After balancing the drag forces normal and parallel to the membrane surface they ended up with a ratio of the applied pressure and the wall shear stress, which in turn depends on the cross-flow velocity and the permeate flux, and is equal a dimensionless number. A critical value of the dimensionless number, that is a critical selective cut-diameter of the deposited particles exists, such that above this value, the filter medium is clogged and filtration rate reduced, and below this value high filtration rates can be maintained over a long period of time.
Rautenbach and Schock (1988) applied the same formulation as Ebner (1981) in their investigation of clay-water and quartz-water dispersions. But, they reduced the equilibrium of drag forces to an equilibrium of velocities, which yield for the condition of particle movement

\[ \bar{u} \geq fJ \]  

(48)

where \( f \) is the friction factor. They related the permeate flux to the fluid Reynolds number and the particle size by

\[ J = Re^{1.26} \left( \frac{d_p}{d_h} \right)^{0.44} \]  

(49)

where the exponents followed out of the boundary layer velocity distribution if the centre of the sphere is assumed to be in the buffer layer (viscous and turbulent fluid friction). The Reynolds exponent was found to be in good agreement with microfiltration experiments of different solute systems and operating conditions from the literature. Rautenbach and Schock performed experiments with different module types, particle dispersions and operating parameters which yield results reasonably accurately predicted by the model.

Blake et al. (1990) developed a model to predict the steady state flux of a cross-flow filter based upon the frictional force balance. Unlike previous friction models both hydrodynamic and interparticle forces (Van der Waals attraction and double layer repulsion) were considered and found to be of similar magnitude for surface separations less than 0.1 \( \mu \)m under the conditions: Hamaker constant of \( 10^{20} \) J, particle radius of 0.5 \( \mu \)m, ionic strength of 0.05 M and zeta potential of 50 mV. Experiments were performed with spherical polystyrene latex particles of 0.55 or 1.8 \( \mu \)m diameter as feed dispersion and different tubular membranes, in order to check the model. Generally, agreement was found between the predicted linear equilibrium lines of permeate flux dependence on wall shear stress and the experimental results. Furthermore, the slope of the lines was an increasing function of particle diameter, as successfully predicted.

However, interparticle forces will have greater importance in the submicron particle size range compared to that for micron sized particles (see Section 5.9).
5.6 Particle Adhesion Model

During cross-flow filtration experiments observations show that after an initial period of flux decline, a steady state flux is reached. In general, this steady state value is found to increase with increasing applied cross-flow velocity, due to the additional shearing forces generated at the higher velocities, which cause less particulates to accumulate at the membrane surface and thus facilitate reduced filter cakes. However, in some cases, for example, Fischer and Raasch (1986), Lu and Ju (1989), Wakeman and Tarleton (1991) and Mackley and Sherman (1992), reduced values have been reported, which are explained in terms of the selective deposition of fine particles into the filter cake, thus reducing the permeability of the cake layer, and by a variation of the cake packing structure with cross-flow (Mackley and Sherman (1992)).

Fischer and Raasch (1986), Lu and Ju (1989), Wakeman and Tarleton (1991) and Mackley and Sherman (1992) presented experimental evidence that in cross-flow filtration, among the particles transported to the membrane surface only a fraction become deposited. Thus, Mackley and Sherman (1992) developed a model around their direct in situ observations of the cake deposition process and their filtrate flux measurements of 125–180 µm polyethylene particles in aqueous suspension. The relatively narrow size distribution was chosen in order to avoid the effects of deposition of fine particles into the filter cake structure. The model is based on the general static-cake-filtration equation with allowance for the probability of particle capture (adhesion) onto the cake surface, inducing a variation in the specific cake resistance. This was related to particle arrival trajectory by defining a capture probability function $f(\phi)$, based on a capture angle, $\phi$, such that when the filtrate flux dominates all impacting particles are captured and as cross-flow becomes increasingly important the capture probability falls. Their model predictions of permeate flux and cake height versus time have been found to be in good agreement with the experimental results, at least partly because of a trial-and-error fitting procedure needed to evaluate a specific resistance parameter out of their experimental data. However, this parameter, which gives a ratio of the resistance of cake layers formed at high cross-flow with negligible filtrate flux and with no applied cross-flow, was estimated to be within the limits of a random loose packing and a hexagonal close packing, and therefore physically credible. It should be noted that from their optical observations no evidence for particle back
diffusion or flux of particles away from the cake surface could be detected.

Stamatakis and Chi Tien (1993) formulated their cross-flow model on the same premise that only a fraction of the particles convected to the filter medium surface become deposited. The model is based on the friction force model with a criterion of particle deposition related to a protrusion out of the filter cake of finite height, which may be considered as a protruding particle in the cake–slurry interface. The particle drag force normal to the membrane surface is expressed, in contrast to the friction force model, as the sum of the convective hydrodynamic drag force due to the permeating liquid, the lateral migration force (from Vasseur and Cox (1976)) and the buoyant force on the spherical particle. A particle adhesion probability was established as a function of the minimum protrusion height for particle deposition. Stamatakis and Chi Tien's (1993) time dependent model is capable of predicting the right qualitative trends for all variations in operating conditions and particle diameters, as well as taking polydisperse systems into account. The compressible cake version is based on a power law dependency of the compressible drag pressure inside the cake subject to the local permeability and voidage, which results in three empirical parameters needing adjustment. A comparison with microfiltration experiments performed by Murkes and Carlsson (1988) using finely dispersed and stabilized kaolin particles of 1 μm size gave good agreement with their predictions from the incompressible model. However, the good agreement was probably gained by a prior optimization–search procedure to obtain the constant cake voidage and permeability values of Murkes and Carlsson’s (1988) experimental results, which were subsequently implied in the model.

In summary, the particle adhesion model seems to be valid for large particles, since it could describe the at first unexpected effect of decreasing permeate flux with increasing cross-flow velocity. However, Wake- man and Tarleton (1991) found that for their small calcite particles of mean size of 2.7 μm the opposite was true. This leads to the conclusion that the particle deposition and packing selectivity is more marked for larger size particles. Mackley and Sherman (1992) explained this as due to the higher probability of suitable packing sites (capture possibilities) per unit length of filter for smaller particles. Also Brownian motion will become more significant, perhaps allowing more sites to be explored by the particles for capture. A further aspect could be the increased dominance of interparticle forces and especially the attractive van der Waals forces, which for high ionic strength or low particle surface potentials
could maintain the particles in the so called secondary minimum of particle–particle interaction and thus yield a uniform packing independent of cross-flow velocity.

5.7 Pore Blocking Model

Fane et al. (1981) introduced a semi-empirical ultrafiltration model based on the gel-polarization model to allow for the effect of membrane surface properties on gel-polarized ultrafiltration flux. They found experimentally that gel-polarized ultrafiltration flux is dependent on membrane permeability and surface properties. The gel-polarization model has been modified by introducing an effective free area correction factor to account for membrane surface regions of differing permeability. The model is equivalent to the conventional gel-polarization model (Eq. (4)), for the limiting case, where the membrane has a real homogeneously permeable membrane surface.

Le and Howell (1984) proposed a semi-empirical limiting flux model, which is also based on concentration polarization, but postulates that the limiting membrane surface concentration is a result of the interactions between the solute and the surface. Limiting flux occurs when the pores remain in a dynamic blocked–unblocked state responding to convective blockage and reverse mass transfer to clear the blockage. The net flux is related to the relative size of the membrane pore and solute particle or molecule and depends on a totally unblocked and partially blocked fraction of pores. The model fitted well to a range of protein, bacterial cell and polymer latex experiments, requiring parameter estimation of the limiting membrane solute concentration and a pore blocking parameter. However, a theoretical model for their relationship has not yet been developed from first principles.

5.8 Surface Renewal Model

Koltuniewicz (1992) developed a semi-empirical limiting flux model on the basis of a surface renewal concept, which accounts for hydrodynamical impulses such as eddies, bursts or eruptions (turbulent flow) and lifting forces (laminar flow) of fluid elements adjacent to the membrane surface. Infinitesimal (solute) surface elements are randomly, and independently of their age, removed yielding an instantaneous time-
dependent permeate flux (for each element), which led them to the well-known age function, after Danckwerts. The resulting total permeate flux could be predicted as a function of time after initial dead-end and cross-flow experiments were performed in order to obtain the necessary parameters. The model seems to be rather intuitive, but the main advantage of the surface renewal concept is the avoidance of the need for data concerning the diffusive or non-diffusive nature of the solute, osmotic pressure, gel concentration or some other character or phenomenon which controls the flux.

5.9 Particle-Particle Interactions Model

McDonogh et al. (1984) have reported the results of unstirred dead-end ultrafiltration of colloidal silica ($d_p = 24.2$ nm) where the addition of different salts (electrolytes) and pH-adjustments were used to control the zeta potential (measurable surface charge) of the colloid. Strong dependence of the specific resistance, $\alpha$, of the filter cake on zeta potential was found. They correlated their observations with repulsive forces between the colloids, which arise because of double layer interactions. However, the model development is rather questionable due to a probable misinterpretation of the DLVO theory (Verwey and Overbeek (1948)), as well as dimensionally inconsistent equations and untraceable literature quotations.

McDonogh et al. (1989) studied the charge effects in the cross-flow ultrafiltration of monodisperse silica colloids ($d_p = 24.2$ nm). Increasing permeate flux (decreasing hydraulic resistance) with increasing zeta potential for various cross-flow conditions was observed. This led them to the conclusion that the factors which control concentration polarization are charge-dependent. The effect of particle charge was incorporated in the film theory model to predict the experimental flux values. Their model predictions mirrored the trends of the experiments as a function of zeta potential, where the conventional film model was always under-predicting and was in error by a factor of seven at high zeta potentials. However, since the electrokinetic part of the cross-flow model is based on McDonogh et al. (1984) approach results should be taken with caution. McDonogh et al. (1992), in a further publication based on the treatment of McDonogh et al. (1984), showed the compressive drag pressure variation within a filter cake based on empirical power law assumptions for the compressibility.
Bowen and Jenner (1994a) have developed rigorous mathematical models for predicting the rate of ultrafiltration of charged colloidal dispersions which include quantitative calculations of the particle–particle interactions within filter cakes which are responsible for controlling permeation rates. The models were based on an extended pairwise summation of interaction energies between the particles in the filter cake due to electrostatic or double layer forces, London-van der Waals or dispersion forces and hydration forces, through the extended DLVO theory. Electroviscous effects in the filter cake, that is an increased apparent fluid viscosity within the cake, have also been accounted for. The models were a priori models ("from physics to filtration") with no adjustable parameters. They allow quantitative prediction of the rate of filtration of charged colloidal dispersions as a function of zeta-potential, ionic strength, applied pressure, particle radius, particle packing geometry and membrane resistance. In particular, a dynamic model was developed which takes into account the variation of local specific cake resistance (or local cake voidage, or local interparticle separation) as a function of both position in the cake and time. (One feature of the model is, hence, that it may also be considered as providing a fundamental description of a type of compressible cake filtration without empirical power law assumptions for the compressibility). The predictions of the model were systematically investigated, revealing the dynamic nature of the ultrafiltration process and showing the great importance of taking particle–particle interactions into account. Comparison of the predictions of the dynamic model with experimental filtration data for colloidal silica were carried out. For intermediate values of zeta-potential and ionic strength there was good agreement between the theoretical predictions and experimental data. However, the model tended to underpredict filtration rates at low zeta-potentials and overestimated filtration rates at high zeta-potentials, even when hydration forces and electroviscous effects were included respectively. In contrast, neglect of interparticle interactions, as in classical filtration theory (Eq. (22)), led to underestimation of filtration rates by orders of magnitude under the conditions studied.

However, the good agreement between the dynamic model and experimental behaviour for intermediate conditions was satisfying, as the model represented the first rigorous attempt to predict the dynamic rate of filtration of colloids from a physics basis. Bowen and Jenner (1994a) explained that the overprediction of filtration rates at high zeta-potential probably arose from two causes. Firstly, that the extended pairwise
summation of interaction energies approach represented an approximation to the truly multiparticle nature of the interactions in the filter cake. Secondly, that there was some uncertainty in calculating zeta-potentials from electrophoretic mobility measurements using the best available electrokinetic theories (O'Brien and White (1978)). If the O'Brien and White calculations were overestimating the real zeta-potential, as was believed, then the filtration rate predictions would also be overestimates. An important factor in the underprediction of filtration rate at very low zeta-potentials may have been the relatively poor understanding and quantification at present of non-DLVO short-range forces such as hydration forces. Overall, the theoretical work showed the great importance of including an account of interparticle interactions in quantitative descriptions of ultrafiltration.

Bowen and Jenner (1994b) extended their dynamic model by introducing a Wigner–Seitz cell approach for the calculation of the configurational repulsive electrostatic interaction energy to account properly for multiparticle interaction effects. The increase in configurational entropy (also a multiparticle effect) due to the accumulation of particles above the membrane surface and a rigorous solution for fluid flow through porous media by Happel (1958), also based on a cell approach, were also considered. The model was tested against experimental data for the ultrafiltration of well characterized silica colloids and excellent agreement was obtained between the theory and the experiment throughout a broad range of experimental conditions. Further, yet unpublished data, showed an equally good agreement between the dynamic cell model predictions and experiments for protein solutions (BSA). Bowen and Jenner (1994b) remarked that it was especially satisfying that such agreement could be obtained for a model with no adjustable parameters. The excellent agreement was a confirmation of the appropriateness of the descriptions of fundamental particle–particle interactions included as the basis of the model and of its development to account for the dynamic nature of ultrafiltration.

6. CONCLUSION

The decision as to which of the filtration models is the most correct in predicting flux values is a matter of difficulty and a certain amount of controversy. Each model appears to be consistent with selected experimental data.
1. The gel-polarization model has been used to correlate a vast range of ultrafiltration data. However, the work of Jonsson (1984) and Wijmans et al. (1984) showed that an osmotic pressure model describes pressure independent flux and the semi-logarithmic relationship between $J$ and $c_b$ just as well as the gel-polarization model. Furthermore, the dependence of the limiting wall concentration to the applied pressure is in contrast to the gel-polarization model prediction. In particular, solutions of branched chain macrosolutes such as dextrans which do not form a gel or exhibit high viscosity seem most appropriately described by the osmotic pressure model. However, Wijmans et al. (1985) showed that the boundary layer resistance model is essentially equivalent to the osmotic pressure model.

2. The resistance models have the advantages of including a description of non-Newtonian polarized layers, concentrated solutions of solutes which are capable of forming loosely-bound structures, solid cake layers, and of being readily extended to allow for a time-dependent fouling resistance.

3. Since the most widely used colloidal filtration models generally neglect particle diffusion effects as well as interparticle interactions, the particle size appropriate to the theories is usually from 0.1 μm upwards, which is the border-range between ultrafiltration and microfiltration. The inertial migration model is eligible only for particles with radius larger than 1 μm, since only these particles will be affected by the lift velocity (Altena et al. (1983)).

4. The empirical scour model is found to be intuitive rather than rigorous and a theoretical basis has to be established for the governing model equations to enable further developments. Recently, Yung et al. (1989) have experimentally demonstrated that there is some doubt over the ability of turbulent bursts to remove deposited material from a surface, which puts Gutman’s (1977) turbulent burst model in question.

5. The friction force model empirically predicts the results of experiments with different module types, particle dispersions and operating parameters quite accurately. However, the simple geometry has to be extended to a more realistic one in order to substantiate the theoretical basis.

6. The empirical particle adhesion model, as a kind of extension to the friction force model or cake filtration model, are even capable of predicting decreasing flux rates with increasing cross-flow, as recently experimentally observed (Lu and Ju (1989), Wakeman and Tarleton (1991), Mackley and Sherman (1992)). However, the model is formed
on a statistical adhesion probability function which must be determined, qualitatively at least, from experimental observations. The semi-empirical pore blocking model is an advance on the gel-polarization theory since it takes membrane-solute interactions into account. However, a theoretical model for the pore blocking relationship has not yet been developed from first principles.

7. The recently developed shear-induced hydrodynamic diffusion models by Romero and Davis (1988, 1990) and Davis and Sherwood (1990) look quite promising, since they incorporate many fundamental facts about the cross-flow filtration process, for instance, concentration dependent effective diffusivity and viscosity in the boundary layer and the existence of a stagnant and flowing cake layer, although specific experimental verification is not published so far due to the lack of sufficient effective diffusivity data. Also, recent optical observations of the cake deposition process of Mackley and Sherman (1992) revealed no evidence for particle back diffusion or flux of particles away from the cake surface, which will put the formulation of the model in question.

8. The particle–particle interaction model from McDonogh et al. (1984, 1989, 1992) is the first model which attempts to take electrostatic interactions between the particles into account. Even though the model development is lacking to a great extent, the fundamental idea behind makes it suitable for colloidal ultrafiltration, since variations in ionic strength and zeta potential are considered. However, the recently developed rigorous dynamic mathematical models from Bowen and Jenner (1994a,b) for colloidal filtration, which account, in a sophisticated manner, for the underlying fundamental particle–particle interactions in a filter cake, are excellent descriptions for the prediction of dead-end ultrafiltration rates. Experimental confirmation of the dynamic cell model predictions (Bowen and Jenner (1994b)) showed excellent agreement for the tested colloidal silica system. As yet unpublished experimental data showed equally good agreement for the ultrafiltration of protein solutions (BSA). The models are physics based with no adjustable parameters. However, from an industrial point of view a cross-flow model is needed, which is currently under development by the authors.

In the following the most appropriate cross-flow models for different types of process feeds are listed:
1. Feed type: Macromolecules, Mwt = 10^5 and higher.
   a) Theory used: Gel-polarization theory with concentration dependent diffusivity and viscosity (Shen and Probstein (1977)).

   Model equation: For laminar flow, solved numerically

   \[
   J_{\text{lim}} = 1.5 \left( \frac{u_b D_b^2}{hL} \right)^{1/3} \bar{J}
   \]

   \[
   \bar{J} = \left( 1 - \frac{c_b}{c_g} \right) \left[ \int_{0}^{c_g} D(c)^{-1} \exp \left( - \int_{0}^{c_g} \frac{\eta^2 + \bar{J}}{D(c)} \, d\eta \right) \, d\eta \right]^{-1}
   \]

   \[
   \bar{D}(c) = D(c) / D_b(c_b)
   \]

   \[
   \eta = y \left( 3 x D_b / \left( \frac{du}{dy} \right)_b \right)^{-1/3}
   \]

   Model parameters: \( c_g, D(c) \)

   for example: \( D(c) = \frac{7.1 \times 10^{-7} \tanh (0.159 \, c)}{0.159 \, c} \) [cm^2 s^{-1}]

   (see Fig. 2, Keller et al. (1971))

   Comments: Accuracy of the prediction is limited due to available physical property data.

b) Theory used: Gel-polarization theory with \( D = D(c_g) \) (Probstein et al. (1978)).

Model equation: For laminar flow, solved analytically

\[
J_{\text{lim}} = 1.31 \left( \frac{u_b D(c_g)^2}{hL} \right) \ln \left( \frac{c_g}{c_b} \right)
\]

Model parameters: \( c_g, D(c_g) \)

Comments: This is an approximation of a) valid for \( c_g/c_b >> 1 \); the predictive difference when compared to a) is \( \pm 10\% \) between 0.05 g cm\(^{-3}\) < \( c_b < 7 \) g cm\(^{-3}\).
2. **Feed type:** Macromolecules, $10^4 < \text{Mwt} < 10^5$.

   a) **Theory used:** Osmotic pressure theory.

   **Model equation:** Solved by trial and error

   \[
   J = \frac{| \Delta p | - | \Delta \pi |}{\mu R_m}
   \]

   \[
   \Delta \pi = \pi(c_m) - \pi(c_p) = \sum_{i=1}^{3} a_i c_m^i = \sum_{i=1}^{3} a_i c_b^i \exp \left( \frac{iJ}{k_s} \right)
   \]

   **Comments:** Requires knowledge of the osmotic pressure as a function of concentration to get at least the coefficients $a_i$; generally good agreement with experimental data.

   b) **Theory used:** Boundary layer resistance theory (Wijmans et al. (1985)).

   **Model equation:** Solved by trial and error

   \[
   J = \frac{| \Delta p |}{\mu(R_m + R_{bl})}
   \]

   \[
   R_{bl} = \frac{1 - v_1 / v_0}{\mu s_0} \frac{D}{J} \left[ c_m - c_b + \frac{K_1}{2} (c_m^2 - c_b^2) + \frac{K_2}{3} (c_m^3 - c_b^3) + \frac{K_3}{4} (c_m^4 - c_b^4) \right]
   \]

   \[
   c_m = c_b \exp \left( \frac{J}{k_s} \right)
   \]

   **Comments:** No gel-formation at the membrane surface is required; excellent experimental agreement with extran solutions.

3. **Feed type:** Macromolecules.

   **Theory used:** Empirical model which includes the osmotic pressure theory with a flux dependent mass transfer coefficient and concentration dependent viscosity (Aimar and Sanchez (1986)).
Model equation: 

\[ J_{\text{lim}} = \frac{K \bar{u}_b \mu_b^{-X} \exp(1) X}{\mu} \]

\[ K = AD^{(1-x)} \rho^{(y-x)} \mu_b^{(x-y)} \]

Model parameters: \( K, X, y \) or \( A, x, X, y \)

Comments: Parameters have to be obtained through an experiment; experimental agreement is very good for different solute and membrane types and hydrodynamic conditions.

4. Feed type: Fine particles (and colloids).

Theory used: Shear-induced hydrodynamic diffusion theory with concentration dependent viscosity and effective diffusivity (Romero and Davis (1988)).

Model equation: For laminar channel flow, solved numerically

\[ J_{\text{lim}} = J_0 + \frac{1}{L - x_{cr}} \int_{x_{cr}}^L J_0 \left( \frac{3}{2} \frac{x - x_{cr}}{x_{cr}} + 1 \right)^{-1/3} dx \]

\[ x_{cr} = \frac{\tau_{w0}^3 d_p^4}{16 \mu_j^3 \phi_b J_0^3} \bar{Q}_{cr}(\phi_b) \]

\[ \tau_{w0} = \frac{3\mu_b \mu \eta(\phi_b)}{h_e} \]

\[ \bar{Q}_{cr}(\phi_b) = \int_{\phi_b}^{\phi_{\text{max}}} \left( \int_{\phi}^{\phi_{\text{max}}} \bar{D}(\phi') \frac{d\phi'}{\phi'} \right) \frac{d\phi}{\eta(\phi)} \]

\[ \phi_{\text{max}} = \text{maximum random packing density (for uniform spheres } \phi = 0.645 \text{ (Onoda and Liniger (1990)))} \]

\[ \eta(\phi) = \frac{\mu(\phi)}{\mu} = \left( 1 + 1.5 \frac{\phi}{1 - \phi/0.58} \right)^2 \]

\[ \bar{D}(c) = 0.33 \phi^2 \left[ 1 + 0.5 \exp(8.8 \phi) \right] \]
Comments: Applies for the case of a large cake resistance compared to the membrane resistance; $J_0$ is not specified in the model, therefore, it is suggested that $J_0$ must be either experimentally determined at limiting conditions or approximated by $J_0 = \Delta p/(\mu R_m)$; good agreement is obtained for many microfiltration experiments under different operation conditions.

As stated previously, which of the membrane filtration models is the more appropriate appears to depend on the solute system. Whilst the film theory model is an important description of the concentration polarization process in ultrafiltration and usually employed in the models for the description of the concentration at the membrane surface or the concentration profile in the boundary layer, there are a number of unresolved questions concerning the importance of concentration-dependent solute properties, the effect of convection on the mass transfer, the influence of a wall slip velocity, the effects of membrane surface heterogeneity and solute–membrane interactions, which are probably responsible for the main uncertainties in the prediction of membrane filtration permeate rates. The further development of reliable methods for the prediction of the rate of membrane filtration is crucial to the more widespread use of such separation technology.

7. ACKNOWLEDGEMENT

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8. APPENDIX: NOMENCLATURE

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_m$</td>
<td>Cross-section area of membrane surface</td>
<td>m$^2$</td>
</tr>
<tr>
<td>$c$</td>
<td>Concentration</td>
<td>kg m$^{-3}$</td>
</tr>
<tr>
<td>$d_h$</td>
<td>Hydraulic diameter</td>
<td>m</td>
</tr>
<tr>
<td>$d_p$</td>
<td>Particle diameter</td>
<td>m</td>
</tr>
<tr>
<td>$d_t$</td>
<td>Diameter of a tube</td>
<td>m</td>
</tr>
<tr>
<td>$D$</td>
<td>Diffusion coefficient</td>
<td>m$^2$ s$^{-1}$</td>
</tr>
<tr>
<td>$D(c)$</td>
<td>Diffusion coefficient as function of $c$</td>
<td>m$^2$ s$^{-1}$</td>
</tr>
<tr>
<td>$f$</td>
<td>Friction coefficient</td>
<td>–</td>
</tr>
</tbody>
</table>
\(F\) Drag force \(N\)
\(h\) Height of half channel \(m\)
\(h_e\) Effective height of half channel \(m\)
\(J\) Permeation rate (flux) \(m\,s^{-1}\)
\(J_0\) Pure solvent flux \(m\,s^{-1}\)
\(k_s\) Overall mass transfer coefficient \(m\,s^{-1}\)
\(L\) Characteristic length \(m\)
\(m_p\) Mass of particles in the cake \(kg\)
\(M_{wt}\) Molecular weight \(kg\,kmol^{-1}\)
\(p\) Permeability of cake deposit \(m^2\)
\(\Delta p\) Pressure difference \(N\,m^{-2}\)
\(A\) Smallest cross section area of particle \(m^2\)
\(Q\) Largest cross section area of particle \(m^2\)
\(r\) Radial coordinate \(m\)
\(r_{bl}\) Specific resistance of boundary layer \(m^{-2}\)
\(R_{bl}\) Resistance of boundary layer \(m^{-1}\)
\(R_c\) Resistance of cake layer \(m^{-1}\)
\(R_g\) Resistance of gel layer \(m^{-1}\)
\(R_m\) Resistance of membrane \(m^{-1}\)
\(R_s\) Resistance due to solute \(m^{-1}\)
\(Re\) Reynolds number
\(Re_t\) Reynolds number in Eq. (34)
\(Re_{ch}\) Reynolds number in Eq. (36)
\(s\) Sedimentation coefficient as function of \(c\) \(s^{-1}\)
\(sc\) Schmidt number
\(Sh\) Sherwood number
\(v\) Axial velocity component (tangential to the membrane) \(m\,s^{-1}\)
\(\bar{u}\) Axial average velocity \(m\,s^{-1}\)
\(u_{rn}\) Axial centreline velocity \(m\,s^{-1}\)
\(v\) Velocity component normal to the membrane \(m\,s^{-1}\)
\(\bar{v}\) Partial specific volume \(m\,kg^{-1}\)
\(V\) Total volume filtered \(m^3\)
\(x\) Coordinate tangential to membrane \(m\)
\(\gamma\) Coordinate perpendicular to membrane \(m\)

**Greek letters**

\(\alpha\) Specific resistance of cake deposit \(m\,kg^{-1}\)
\(\delta\) Thickness of boundary layer \(m\)
\(\varepsilon\) Fractional voidage of cake deposit
\(\phi\) Volume fraction of particles \((= 1 - \varepsilon)\)
\(\gamma\) Shear rate \(\text{s}^{-1}\)
\(\mu\) Dynamic viscosity of solvent \(\text{kg m}^{-1}\text{s}^{-1}\)
\(\mu(c)\) Dynamic viscosity of solution as function of \(c\) \(\text{kg m}^{-1}\text{s}^{-1}\)
\(\nu\) Kinematic viscosity of solvent \(\text{m}^2\text{s}^{-1}\)
\(\Delta\pi\) Osmotic pressure difference \(\text{N m}^{-2}\)
\(\rho_p\) Particle density \(\text{kg m}^{-3}\)
\(\tau\) Shear stress \(\text{N m}^{-2}\)

**Subscripts**

\(b\) Bulk
\(c\) Cake
\(f\) Fouling
\(g\) Gel
\(l\) Lateral
\(\text{lim}\) Limit
\(m\) Membrane
\(p\) Permeate or particle
\(r\) Radial
\(ss\) Steady state
\(0\) Solvent or dilute
\(1\) Solute

9. REFERENCES


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