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THE RELEVANCE OF FOULING MODELS TO CROSSFLOW MICROFILTRATION

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ABSTRACT

An overview of models for crossflow microfiltration is presented, and several are compared with experimental pseudo-equilibrium permeate flux values. Models requiring curve fitting procedures do not describe the physics of the process, and require the answer to be known *a priori*. Predictive models were generally found to give fluxes which were in error by several orders of magnitude. The shear-induced hydrodynamic self-diffusion model was found to give the best predictions of permeate flux, but the errors found when comparing predictions with experimental values suggest that improvements need to be made to the model or that it is not a good description of the mechanisms which actually occur in microfiltration.

INTRODUCTION

Crossflow microfiltration (MF) is a technique which has increasing importance for the removal of fine particulates from dilute suspensions to yield a particulate free filtrate or permeate. In a microfilter the feed suspension flows with a high velocity (up to 6-8 m s⁻¹) tangentially to the permeable surface, and the permeate velocity through the porous membrane is low (≈ 0.1 m s⁻¹). The high shear stress associated with the feed stream, together with the scouring action of the bulk flow, reduces the tendency of particles to accumulate near the membrane surface. Although MF is often associated with no filter cake formation, the convective flow of suspension towards the surface does cause an accumulation of particles to occur, and if the concentration rises sufficiently a cake may form. Despite the apparent technological importance of membrane fouling, models previously developed for MF often prove inadequate when compared with observed behaviour.

In recent years many workers have attempted to model the phenomena associated with MF, in some cases simply extrapolating models developed for UF without giving due regard to differences between the physics underlying the two processes. Existing MF models may be divided into two categories:

- i) Curve-fitting models, which are fitted to experimental data through the use of an adjustable parameter, and
- ii) Predictive models, which require no adjustable parameters, and which forecast the permeate flux behaviour from the process conditions.

In this paper, some of the more appropriate existing theoretical membrane fouling models are briefly reviewed. Using data obtained from the filtration of calcite suspensions obtained under carefully controlled conditions using a computerised filter rig¹, comparisons are made between this data and the models in order to assess their relevance to MF. Table 1 shows the range of experimental values used.

CURVE FITTING MODELS FOR PERMEATE FLUX DECLINE

Although the ultimate aim is to develop a MF model which predicts the filtrate flux behaviour from process and operating conditions, the mechanisms responsible for membrane fouling are still not fully understood. This is perhaps the reason why many previous workers have used curve fitting

models applicable to other filtration techniques to describe the permeate flux decline observed in MF.

The simplest attempts to model the behaviour of MF systems are based upon the concentration polarisation concept, which had previously been applied to the UF of macromolecules by Blatt *et al.*². Interpreted in the context of MF, an accumulation of particles occurs near the membrane surface due to convection of the suspension towards the membrane, and a cake forms if the concentration at the membrane surface reaches a sufficiently high level. The convective flux of particles towards the membrane is assumed to be balanced at steady state by the Brownian back-diffusion of particles away from the membrane. The governing equation is then

$$\frac{1}{A} \frac{dV}{dt} = k(x) \ln \left(\frac{c_w}{c_b} \right) \quad (1)$$

where A is the area of the filter and dV/dt the filtrate flow rate. c_w and c_b are the particle concentrations in the suspension adjacent to the membrane surface and in the bulk respectively and $k(x)$, the adjustable parameter, is the local mass transfer coefficient between the bulk suspension and the layer next to the membrane surface. However, the relatively large particle sizes that exist in MF systems compared with the molecular sizes dominant in UF systems mean that this model is not strictly applicable to MF. UF permeation rates calculated from equation (1) were found to be up to two orders of magnitude below experimental observations³, giving rise to the so-called 'flux paradox'. In an attempt to explain this it was suggested by some workers, including Porter³ and Green and Belfort⁴, that Brownian back-diffusion is augmented by a lateral migration away from the membrane due to an inertial lift force. However, for MF systems the inertial lift velocity is much less than the permeate velocity, and the 'inertial lift' correction does not properly account for the permeate flux decline.

It has been reported⁵ that classical 'cake filtration' theory can be used to model observed MF behaviour and the equation governing this process is

$$\frac{dV}{dt} = \frac{A^2 \Delta p}{\mu_f (\alpha c V + A R_m)} \quad (2)$$

where A is the area of the filter, Δp the hydraulic pressure difference across the membrane, μ_f the viscosity of the clean liquid, c the effective feed solids concentration, V the filtrate volume and R_m the membrane resistance. The adjustable parameter is then α , the mean specific cake resistance. Plotting the experimental data as $(dV/dt)^{-1}$ vs. V would give a straight line if the theory were correct, and the value of α would be adjusted to give the best fit between theory and experiment. To show examples of experimental data representative of MF test results, two sets are plotted on Figure 1, and from these it is apparent that neither set could be approximated by a straight line. During MF the permeate flux declines to a pseudo-equilibrium level; plotting the reciprocal flux against permeate volume invariably produces a curve which approaches a constant value as an equilibrium fouling layer thickness is formed at the membrane surface. From video studies of the formation of 'cakes' in MF carried as part of this work it is thought that the deposit is in a dynamic state locally in so far as particles in the layer jostle for position and there is continued deposition and removal occurring at the surface. The assumptions behind equation (2) are not met in MF, and it is therefore clear that the use of cake filtration theory to model MF is quite inappropriate.

Another possible model for MF membrane fouling is that developed by Hermia⁶ based on mechanisms of pore blocking. It has been reported⁷ that observations of the rate at which a filter medium blinds during filtration suggest that four basic types of particulate blocking mechanism exist, and Hermia has described the derivation of a set of equations relating the permeate flux

decline during filtration to the properties of the process suspension. Like equation (2), these equations cannot purport to describe a crossflow situation unless appropriately modified.

The cake filtration theory was modified to include the effects of a crossflowing feed stream by Sabuni⁸, allowing for operation with complete recycling of the permeate and retentate. (This describes the *modus operandi* of the filter rig used in the collection of the data used here.) It was also assumed that for a given dispersion, filter and operating conditions only a certain fraction of particles, β , from the volume of feed filtered will actually adhere to the filter surface to form a cake, with the remainder being re-entrained into the bulk suspension. The modified equation then becomes

$$\frac{dV}{dt} = \frac{A\Delta p}{\mu_f \left[R_m + (1 - \exp(-\beta V/V_0))(\alpha V_0 c/A) \right]} \quad (3)$$

Sabuni assumed the value of β to be 1. The adjustable parameter in this model is α , and V_0 is the total feed volume (0.023 m³ for the present experiments). When $(dV/dt)^{-1}$ is plotted against V , the value of α would be varied to give the closest agreement between theory and experiment. If the exponential term in equation (3) is replaced by its series expansion, and noting that for all practical purposes $V/V_0 \ll 2/\beta$ and $V \ll V_0$, the theory predicts straight line behaviour for $(dV/dt)^{-1}$ vs. V . This is in no way representative of MF flux decline data (cf. Figure 1). Thus, as expected since Sabuni's model is only an extension of cake filtration theory, agreement between experimental data and theory is poor.

Mikhlin *et al.*⁹ developed a simple mathematical model of cake build-up during filtration based on the assertion that in crossflow filtration two basic mechanisms act to minimise the accumulation of particles at the filter surface; the feed suspension passing over the membrane contains turbulent eddies which carry away individual particles in addition to the removal of chunks of the cake by fluid shear exerted on the surface of the cake. Performing a mass balance and simplifying yields

$$\left(\frac{J_0}{J_\infty} \right)^2 \ln \left[\frac{1 - J_\infty/J_0}{1 - J_\infty/J} \right] - \left(\frac{J_0}{J_\infty} \right) \left(\frac{J_0}{J} - 1 \right) = \frac{\alpha \mu_f c J_0^2 t}{\Delta p} \quad (4)$$

where J_0 and J_∞ are the permeate fluxes at zero time and steady-state respectively, and J the permeate flux at any time t . Once again the value of α is adjusted such that the best possible agreement is obtained between the permeate flux behaviour described by this equation and that observed experimentally; Figure 2 shows the fit of this equation to the data in Figure 1. From Figure 2 it is seen that the agreement between the experimental data and Mikhlin's theory is good.

The specific cake resistance has been measured for these suspensions in a pressure filter test to be 1.3×10^{12} m kg⁻¹ for 2.6 μ m calcite particles, and about 2×10^{10} m kg⁻¹ for 24 μ m particles. These values from an independent test are remarkably close to the values shown on Figure 2 which were obtained from a least squares fit to the experimental data. Thus, whilst the previous MF models based on cake filtration have failed to approximate the general behaviour of the experimental curves, the Mikhlin model yields theoretical curves which are extremely close to those obtained experimentally with the 'best-fit' values of α being of the same order as the correct value. However, the Mikhlin model cannot be used in a predictive sense even if α were known from a pressure filter test as the values of J_0 and J_∞ are not known *a priori*.

PREDICTIVE MODELS FOR PERMEATE FLUX DECLINE

Several workers in recent years have developed MF models with the aim of predicting permeate flux behaviour. In MF systems, after an initial transient period in which the permeate flux declines

relatively rapidly, the flux remains constant or only declines slowly over a long period and an accumulation of particles occurs at the permeable surface. The rate at which particles are added to this layer by convective motion of the permeate through the membrane is then balanced by some other mechanism of particle transport away from the layer.

Semi-Empirical Equations

The most rudimentary model of flux decline is that developed by Zhevnovaty¹⁰, in which a semi-empirical correlation was used to relate permeate flux to crossflow velocity. He found that the permeate flux is given by

$$J = 0.4576 \times 10^{-6} \left(\frac{L}{2H_0} \right)^{-0.412} Eu^{0.415} Re^{0.879} \left(\frac{k_0 \Delta p}{\mu_f h} \right)^{-0.4} \quad (5)$$

where Eu and Re are the Euler number and Reynolds numbers respectively for flow on the feed side of the membrane, k_0 the coefficient of permeability for the membrane and h is its thickness. Results predicted by equation (5) are compared with all the experimental data for calcite filtrations available to date, and the corresponding graph of J_{exp}/J vs. J_{exp} is shown on Figure 3. If the model were to predict accurately the permeate flux the ratio J_{exp}/J would be close to 1. However, from Figure 3 it is clear that the predicted permeate flux J is four to six orders of magnitude too high; it seems likely that the Zhevnovaty equation gives answers which are specific to the experimental facility used to generate the original data, and therefore has no general utility value.

Hydrodynamic Models

Crossflow microfiltration can be modelled by studying the convection and diffusion of particles towards and away from the polarised layer, neglecting the influences of any stationary cake which may exist. Pearson and Sherwood¹¹ argued that any natural analysis of MF should combine these two approaches and went on to formulate a set of equations to describe the behaviour of the entire system, which enabled them to predict not only the filtration rate but also the thickness of the cake. However predictions made using this model do not agree with experimental results¹¹; predictions of the flux decline are in error and the theory suggests that the cake thickness will increase as $x^{1/3}$ (where x is the distance from the leading edge of the membrane. Our experiments to date suggest that a cake of essentially uniform thickness is formed, but this is still being confirmed through further visualisation studies.

Leonard and Vassilief¹² assumed that the velocity profile in the vicinity of the cake layer was linear, that the permeate flux was time independent, and that the cake layer could be treated as a Newtonian fluid with the same viscosity as the bulk suspension. They also assumed that the deposition of particles onto the cake layer is balanced at steady state by the sweeping of this layer along the membrane surface. This model was extended by Birdsell *et al.*^{13,14} who treated the cake layer as a Newtonian fluid with constant viscosity and solids concentration which are, in general, large compared with the viscosity and concentration in the bulk suspension. However, the effective viscosity in this layer depends on the particle concentration, which is not taken into account in these models. Whilst some of the physics governing the particle transport may be correct, these models generally fail to predict the basic structure of the flowing particle layer.

An alternative model was developed by Hoogland *et al.*¹⁵ based on the concept that the removal of solids approaching the filter surface is achieved by the translation of these solids along the membrane and eventually out of the flow channel. The permeate flow resistance is increased by the resistance of a mobile layer at the membrane surface and no stagnant deposit is formed. Balancing the volume of solids entering and leaving an element of 'cake', rearranging and using the Carman-Kozeny equation for the resistance of the flowing cake leads to the following expression for the permeate flux through the membrane

$$J = \frac{(1 - \varphi_c)^2}{2\varphi_c^2} \left[\frac{(1 - \varphi_b)\tau_w}{\varphi_b\mu_c L} \right]^{1/3} \left[\frac{x_{av}^2 \Delta\rho}{60\mu_f} \right]^{2/3} \quad (6)$$

where τ_w is the wall shear stress, μ_c the cake viscosity, L the channel length and x_{av} the mean particle diameter. According to Romero and Davis¹⁶, for the laminar flow of a suspension in a two-dimensional, non-porous channel the wall shear stress τ_w in equation (6) can be calculated by

$$\tau_w = \frac{3U\eta(\varphi_0)\mu_f}{H_0} \quad (7)$$

where $\varphi_0 = c/(\rho_s + c)$ and ρ_s is the density of the solid. U is the crossflow velocity, H_0 is half the height of the flow channel and $\eta(\varphi)$ the effective relative viscosity, which depends on the local particle volume fraction φ . Recent viscosity measurements on suspensions of rigid spheres in a Couette viscometer by Leighton *et al.*^{17,18} suggest that a reasonable approximation to $\eta(\varphi)$ is

$$\eta(\varphi) = \left[1 + 1.5 \frac{\varphi}{(1 - \varphi/0.58)} \right]^2 \quad (8)$$

The cake viscosity μ_c is estimated following Einstein¹⁹, namely

$$\mu_c = \mu_f (1 + 2.5\varphi_w) \quad (9)$$

where φ_w is the particle volume fraction at the wall. Using equation (6), this model was compared with the same experimental data as the previous model. The ratio of the observed steady-state permeate flux J_{exp} to the calculated value J is shown plotted against J_{exp} in Figure 4. From Figure 4 it is apparent that the calculated values are generally two to four orders of magnitude too high.

The behaviour of a particle in a shear flow is complex, with particles rotating and producing a rotational flow in the nearby fluid which then exerts drag forces on other particles in the vicinity. These particle-particle interactions cause the particles to be displaced from their time-averaged streamlines. Since many such interactions are likely to occur, the motion of a single particle might be expected to exhibit 'random walk' behaviour, which may be characterised by an effective diffusion coefficient. Zydney and Colton^{20,21} proposed that the discrepancy between the UF model of Blatt *et al.* and MF data is due to an augmentation of particle motion by this diffusivity. They balanced the rate of convective deposition of particles onto the layer with the rate of diffusion of particles towards the bulk, but with a shear enhanced diffusion coefficient measured experimentally by Eckstein *et al.*²², whose results were later shown to be deficient due to wall effects in their experiments^{17,18}. Although Leighton and Acrivos¹⁸, Einstein¹⁹, Eckstein, Bailey and Shapiro²² and Batchelor²³ have produced alternative expressions to account for the presence of particles and their effect on viscosity, it seems that the Krieger-Dougherty²⁴ expression provides one of the simpler and more reliable equations. It may therefore be preferable to use this in further MF modelling studies.

Davis and Leighton²⁵ developed a MF model in which the deposition of particles into the polarised layer is balanced by the tangential flow of this fluidised layer of particles. The layer is fluidised by shear induced hydrodynamic self diffusion, or viscous re-suspension of the particles. The shear stress exerted by the bulk suspension on the concentrated particle layer causes particle-particle interactions to occur, and displacement of the particles from their time-averaged streamlines. They migrate in the general direction of decreasing concentration. Thus, an initially stationary particle layer expands and re-suspends under the action of the crossflowing suspension. It is argued that

the resulting loosely packed layer of particles flows along the membrane surface and out of the filter. Under certain conditions a stagnant particle layer is also formed beneath the flowing layer. The co-existence of such stagnant and flowing particle layers has been observed experimentally by Birdsell¹³, but the experimental conditions were not representative of typical MF in practice in so far as large (150-212 μm) acrylic latex particles were used in a 10 mm x 50 mm cross-section channel; in reality particle sizes are more typically within the range 0.05-20 μm . The model of Davis and Leighton was extended from a local treatment of the particle layer to a global model of crossflow microfiltration by Romero and Davis¹⁶. Their model is able to predict the steady state variation of permeate flux with axial distance along the filter, and the analysis yields four coupled equations:

$$\hat{Q} = \frac{16\mu_f^3 J^2 Q}{\tau_w^3 x_{av}^4} = \hat{Q}_{cr}(\varphi_0) \quad (10)$$

$$Q = \int_0^x J \varphi_0 dx \quad (11)$$

$$\tau_w = \tau_{w0} \frac{H_0^2}{(H_0 - \delta_{st})^2} \quad (12)$$

$$J = \frac{\Delta p}{\mu_f (R_m + \delta_{fl} R_{fl} + \delta_{st} R_{st})} \quad (13)$$

where Q and \hat{Q} are the dimensional and non-dimensional excess particle fluxes respectively and τ_w is the wall shear stress given by equation (7). φ_0 is the bulk suspension particle volume fraction, δ_{fl} and δ_{st} are the thicknesses of the flowing and stagnant layers at any distance x along the filter and R_m , R_{fl} and R_{st} are the resistances of the membrane and the flowing and stagnant cake layers respectively. The resistance of the flowing layer is assumed to be negligible in the work of Romero and Davis. Equations (10)-(13) form a system of nonlinear simultaneous equations for the variables Q , v_w , τ_w and δ_{st} which have been solved numerically, and the ratio of the experimental to predicted permeate flux J_{exp}/J vs. J_{exp} plotted on Figure 5. From Figure 5 it is clear that the permeate fluxes predicted by this model are up to three orders of magnitude higher than those observed experimentally.

Comparing Figure 5 with Figures 3 and 4 it is apparent that predictions of the permeate flux using the Romero and Davis model are better than those from either the Hoogland or Zhevnotati models; this does not mean to say that the Romero and Davis model is the correct description of the physics of the process, although elements of truth may exist in the model concepts - as they may also be embodied in the Hoogland model. The steady state model of Romero and Davis has recently been extended²⁶ to predict the transient behaviour of a MF system. Davis and Sherwood²⁷ have presented a similarity solution for the convective diffusion equation governing the steady state polarised boundary layer which agrees with the approximate solutions of Davis *et al.*^{14,16} for dilute suspensions; agreement between these works^{14,16,26,27} does not give any insight into their 'correctness' as a description of MF, but merely points to confirmation that the mathematical techniques used in their solution are correct.

CONCLUSIONS

Existing membrane fouling models are reviewed briefly and those considered most appropriate to crossflow microfiltration are then compared with experimental data. None of the curve-fitting models performed well, except that of Mikhlin which yielded values of α close to experimentally

measured values. Three 'predictive' models were studied in detail, those of Hoogland *et al.*, Zhevnovaty and Romero and Davis. The latter was found to predict most accurately the permeate flux behaviour when the crossflow velocity and concentration take lower values whilst the particle size is largest.

There is still a need to produce a realistic model for crossflow microfiltration. This paper represents part of a continuing study in which further experimental data are being generated over a wider range of variables and using a variety of feed suspensions, some of which will contain biosolids and others macromolecular species. The further work also aims to throw light on the true mechanics and physics predominating in crossflow microfiltration systems through visual observation and recording of the dynamics of particles at the membrane surface, to enable a correct model to be developed and to remove the 'black box' approach which has been adopted in most previous studies.

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NOMENCLATURE

A	area of filter (m)
c_b	bulk suspension particle concentration (kg m^{-3})
c	solids concentration (kg m^{-3})
c_w	particle concentration at wall (kg m^{-3})
Eu	Euler number
h	membrane thickness (m)
H_0	half-height of the flow channel (m)
J	permeate flux ($\text{m}^3 \text{m}^{-2} \text{s}^{-1}$)
J_0	permeate flux at zero time ($\text{m}^3 \text{m}^{-2} \text{s}^{-1}$)
J_∞	permeate flux at steady state ($\text{m}^3 \text{m}^{-2} \text{s}^{-1}$)
k_0	coefficient of permeability (m^2)
$k(x)$	local mass transfer coefficient ($\text{m}^3 \text{m}^{-2} \text{s}^{-1}$)
L	length of filter (m)
Q	excess particle flux ($\text{m}^2 \text{s}^{-1}$)
\hat{Q}	dimensionless particle flux
Re	Reynolds number
R_{fl}	flowing cake layer specific resistance (m^{-1})
R_m	membrane resistance (m^{-1})
R_{st}	stagnant cake layer specific resistance (m^{-1})
t	time (s)
U	crossflow velocity (m s^{-1})
V	filtrate volume (m^3)
V_0	total feed volume (m^3)
x	axial distance measured from filter inlet (m)
x_{av}	mean particle diameter (m)
α	mean specific cake resistance (m kg^{-1})
β	fraction of particles from volume of feed filtered that adhere to filter surface to form a cake
δ_{fl}	flowing particle layer thickness (m)
δ_{st}	stagnant particle layer thickness (m)
Δp	trans-membrane pressure drop (N m^{-2})
μ_c	cake viscosity (Pa s)

μ_f	viscosity of particle-free fluid (Pa s)
$\eta(\varphi)$	relative viscosity, μ_0/μ_f
φ	particle volume fraction
φ_0	bulk suspension particle volume fraction
φ_w	particle volume fraction at porous wall
τ_w	shear stress at wall (N m ⁻²)

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FIGURES AND TABLES

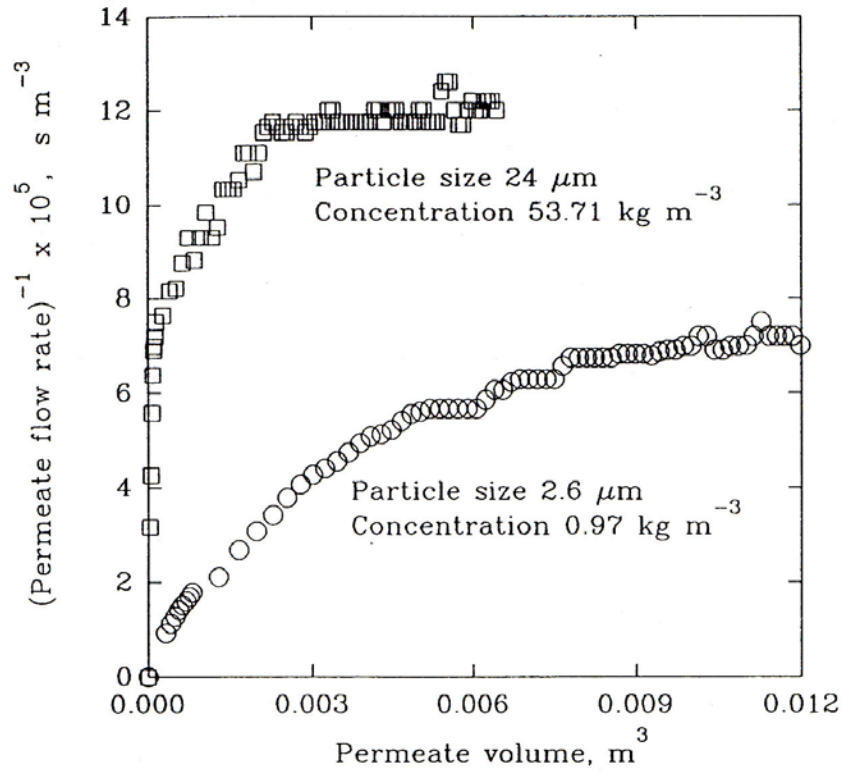


Figure 1: Typical examples of flux decline data plotted as reciprocal permeate flux vs. permeate volume filtered.

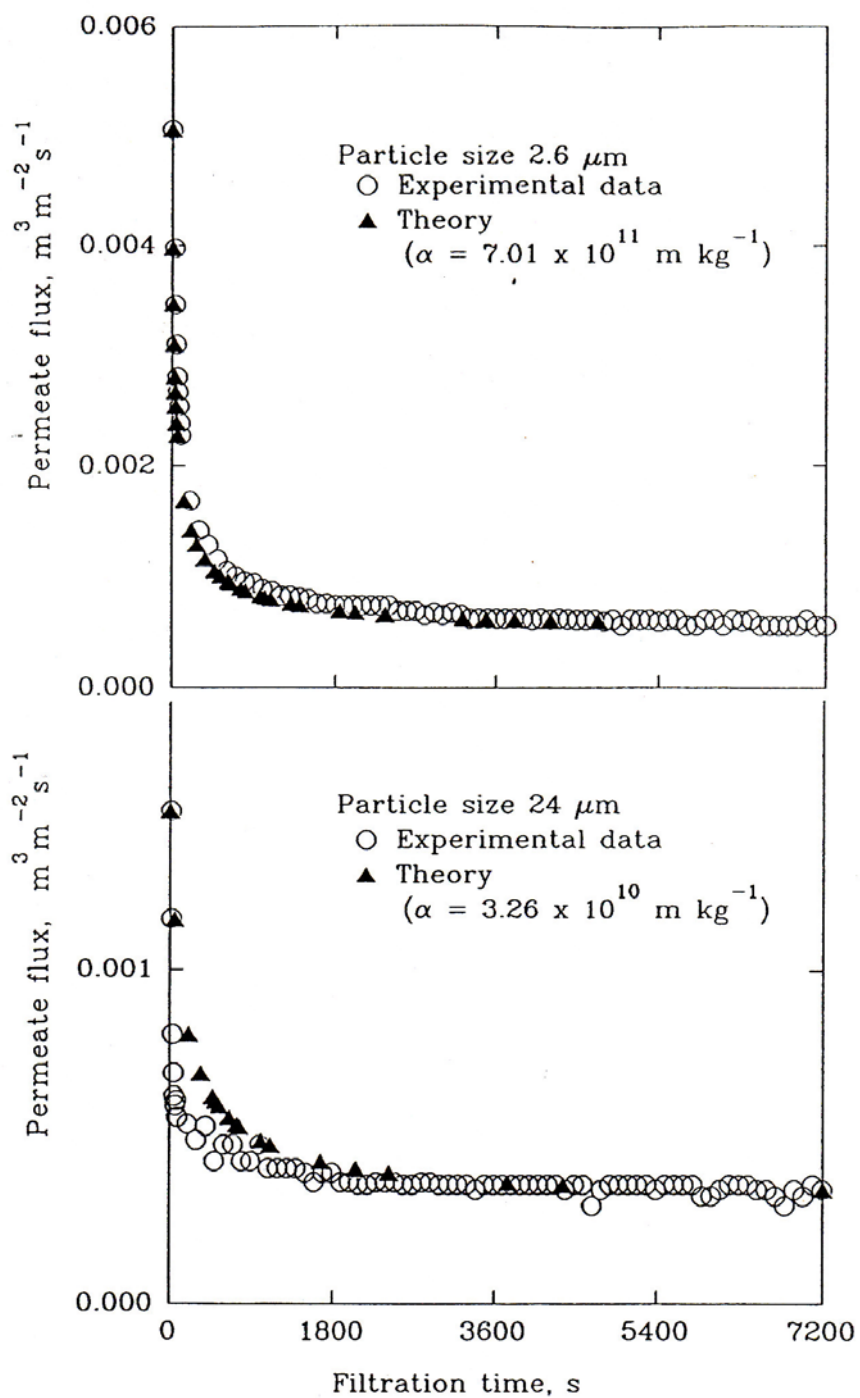


Figure 2: Fitting Mikhlin's theory to the experimental data.

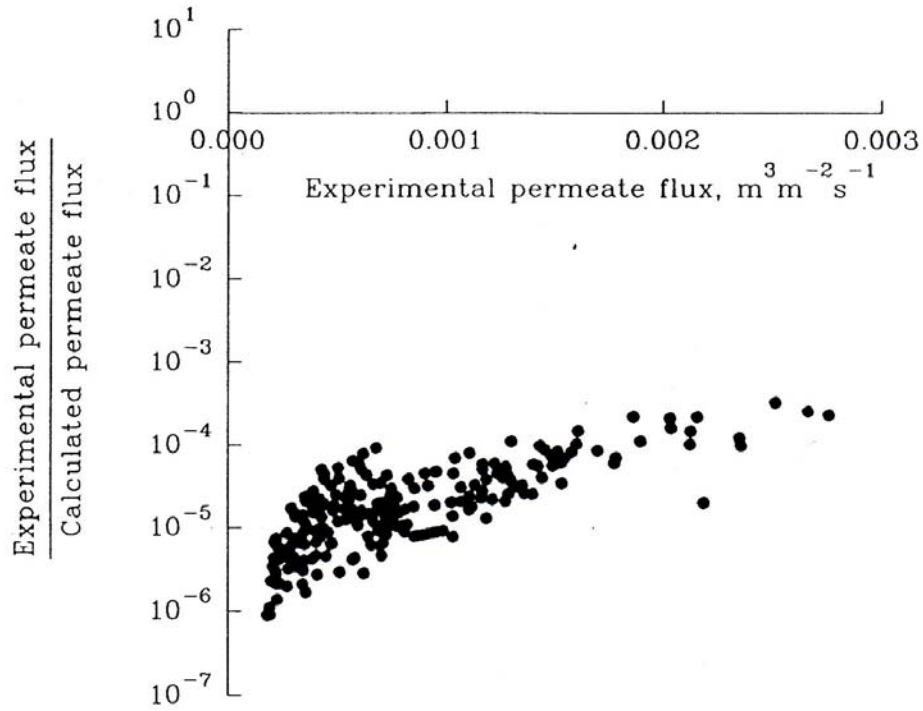


Figure 3: Predictions from the Zhevnovatyi model compared with experimental data.

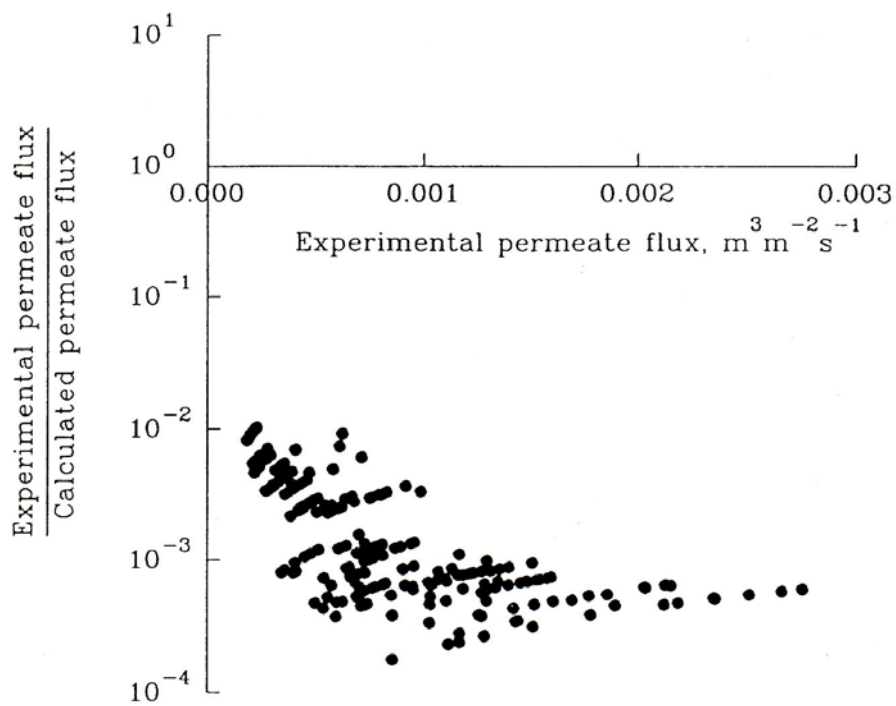


Figure 4: Predictions from the Hoogland model compared with experimental data.

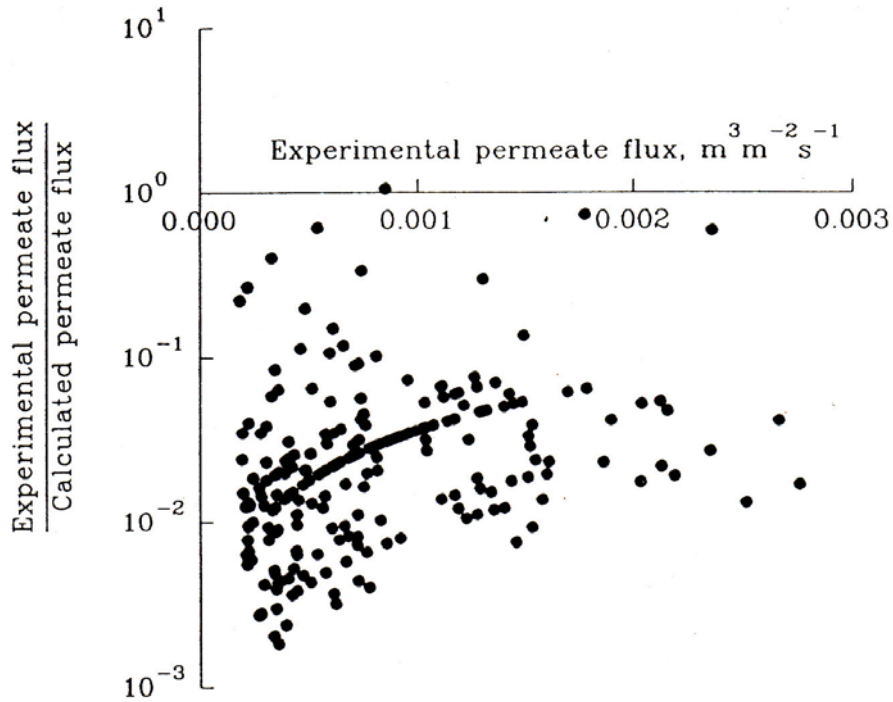


Figure 5: Predictions from the shear induced hydrodynamic diffusion model compared with experimental data.

Filter area	2400 mm^2
Particle size	2.6 - 24 μm
Solids volume fraction in feed	0.033 - 1.8 %
Crossflow velocity	0.8 - 2.3 m s^{-1}
Trans-membrane pressure	69 - 345 kN m^{-2}
Membrane permeability	2.7×10^{-16} - $9.4 \times 10^{-14} \text{ m}^2$
Membrane thickness	10 - 175 μm

Table 1: Range of parameters studied in the experiments.