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On the Relation between Filtrate Flux and Particle Concentration in Batch Crossflow Microfiltration

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ABSTRACT

The relation between the filtrate flux and particle concentration in batch crossflow microfiltration is investigated using a model based on classical filtration theory and the Kern–Seaton theory of surface fouling. The model, which includes the effects of cake compressibility but not of membrane fouling, is solved for both laminar and turbulent tangential flows. It is found that the sole effect of cake compressibility is to reduce the flux without altering the general shape of the flux versus concentration curve. Fluxes which increase with increasing concentration are shown to be a result of enhanced cake removal due to the increased wall shear stress brought about by increased suspension viscosity. A sigmoidal relation between flux and concentration is reproduced by the model only if there is a reduction in the cake removal rate as the tangential flow regime changes from turbulent to laminar.

INTRODUCTION

The relation between filtrate flux and particle concentration during thickening of a suspension by batch crossflow microfiltration is a subject that has provoked considerable comment (1). A simplified diagram of such an operation is given in Fig. 1. As filtration proceeds, the volume of suspension declines, with a parallel increase in particle concentration. Most authors have found that, initially, there is a very rapid decline in flux with increasing concentration (2–5). This is followed by a plateau region in

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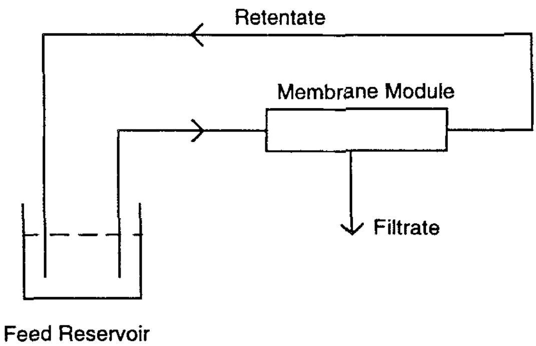

FIG. 1 Batch crossflow microfiltration.

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which the flux is relatively insensitive to changes in concentration. After the plateau region, two types of behavior have been observed. The first is characterized by a flux which increases with particle concentration (2, 4), while the second is distinguished by a sudden drop in flux leading to the classic sigmoidal curve (4–6). While it has been suggested that data of the latter type can be extrapolated to zero flux (5), experimental evidence has been obtained which suggests that the flux does not decline to zero, but reaches another plateau region at very high concentrations (2, 4).

While it is generally accepted that many, if not all, of the above phenomena arise as a result of changes in suspension rheology (4), no attempt has been made to incorporate these ideas into a single mathematical model. In addition, the effect (if any) of cake compressibility on the shape of the flux versus concentration curve remains uncertain. The purpose of this paper is to clarify the roles of rheology and compressibility by developing a simple mathematical model of batch crossflow microfiltration.

MODEL DEVELOPMENT

The basic model consists of a particle balance, a liquid balance, an equation describing the dynamics of cake formation in crossflow systems, and the classical filtration equation relating filtrate flux to pressure driving force. The particle balance can be written

$$\frac{d(Vc)}{dt} = -A\frac{dM}{dt} \tag{1}$$

where A is the membrane area and V, c, and M are the suspension volume, particle concentration, and cake mass per unit area, respectively, at time t. The liquid balance for the system takes the form

$$dV/dt = -AJ \tag{2}$$

where J is the filtrate flux.

The following equation is used in this study to describe cake formation;

$$dM/dt = cJ - k\tau M \tag{3}$$

where k is a constant, τ is the shear stress at the surface of the filter cake, cJ is the particle deposition rate, and $k\tau M$ is the particle removal rate.

This expression owes its origins to the Kern–Seaton model of heat exchanger fouling (7). Those authors assumed that fouling of heat exchangers was the net result of the simultaneous deposition on to, and removal of particulates from, the heat exchanger surface. The particle removal term, $k\tau M$, is based on the assumption that randomly sized clumps of particles are removed by the "scouring" action of the wall shear stress. While a more rigorous derivation of the first-order dependence on M can be formulated using the concept of turbulent bursts (8), the lack of experimental evidence for particle reentrainment by such bursting phenomena (9), means that Eq. (3) must be considered as a purely empirical expression for cake formation.

In this paper, Eq. (3) is assumed to be applicable to both laminar and turbulent tangential flows. This is probably a simplification since one might expect different mechanisms of particle reentrainment for different flow regimes, with a parallel difference in the form of the basic equation for cake formation. In order to distinguish between the two flow regimes, it is assumed later that turbulent-to-laminar transitions can be modeled as involving changes in the cake removal constant, k.

The balance Eqs. (1) to (3) are accompanied by the filtration equation (10)

$$J = \frac{\Delta P}{\mu[R_M + \alpha M]} \tag{4}$$

where ΔP is the transmembrane pressure, μ is the filtrate viscosity, R_M is the membrane resistance, and α is the specific cake resistance.

Inverting Eq. (4) and differentiating with respect to time gives

$$-\frac{1}{J^2}\frac{dJ}{dt} = \frac{\mu}{\Delta P}\left[\alpha\frac{dM}{dt} + M\frac{d\alpha}{d\Delta P_c}\frac{d\Delta P_c}{dt}\right] \tag{5}$$

where it is assumed that the membrane resistance is constant and ΔP_c is the cake pressure drop defined by

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$$\Delta P_c = \Delta P - \mu R_M J \tag{6}$$

Differentiating Eq. (6) with respect to time and combining with Eq. (5) gives

$$\frac{dJ}{dt} = \frac{\alpha(cJ - k_T M)}{\left(-\frac{\Delta P}{\mu J^2} + \mu R_M M \frac{d\alpha}{d\Delta P_c}\right)} \tag{7}$$

Therefore, Eqs. (1), (2), and (7) form the basis of the model, with Eqs. (3) and (4) being employed to substitute for M and dM/dt where required.

Now, assuming that the pressure dependence of the crossflow specific resistance takes the same form as its dead-end counterpart, we can use the empirical expression (11)

$$\alpha = \frac{\alpha_0(1 - n)\left(\frac{\Delta P_c}{P_a}\right)}{\left(1 + \frac{\Delta P_c}{P_a}\right)^{1-n} - 1} \tag{8}$$

where α_0, n, and P_a are constants. The use of this equation clearly introduces another simplification into the model. Previous work has shown that the pressure dependence of the crossflow specific cake resistance may be influenced by particle polydispersity (12, 13). In addition, recent work by Tanaka et al. (14) indicates that the orientation of deposited nonspherical particles is a function of the relative magnitudes of the transmembrane flux and crossflow velocity. It is likely that this effect would also alter the pressure dependence of the crossflow specific resistance. Therefore, the analysis presented in this paper is only applicable to monodisperse suspensions of spherical particles.

It is convenient to write the differential equations in nondimensional form using the following transformations:

$$J^* = J/J_0 \tag{9}$$

$$c^* = c/c_0 \tag{10}$$

$$V^* = V/V_0 \tag{11}$$

$$t^* = \frac{t}{V_0/AJ_0} \tag{12}$$

$$\alpha^* = \alpha/\alpha_0 \tag{13}$$

$$\Delta P^* = \Delta P/P_a \tag{14}$$

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$$\Delta P_c^* = \Delta P_c/P_a = \Delta P^*(1 - J^*) \tag{15}$$

where c_0, V_0, and J_0 refer to initial values of c, V, and J.

Equations (1), (2), and (7), written in dimensionless form and having substituted for all terms involving M, thus become

$$\frac{dJ^*}{dt^*} = -\left[\frac{\varphi_1\alpha^*c^*J^{*3} + \varphi_2J^{*2} - \varphi_2J^*}{1 - \dfrac{\Delta P^*}{\alpha^*}(J^* - J^{*2})\dfrac{d\alpha^*}{d\Delta P_c^*}}\right] \tag{16}$$

$$dV^*/dt^* = -J^* \tag{17}$$

$$\frac{dc^*}{dt^*} = \frac{\varphi_2}{\varphi_1\alpha^*V^*}\left(\frac{1}{J^*} - 1\right) \tag{18}$$

where J^*, V^*, and c^* are all equal to unity at $t = 0$.

The dimensionless groups in the above equations are defined as follows:

$$\varphi_1 = \frac{\alpha_0c_0V_0}{AR_M} \tag{19}$$

$$\varphi_2 = \frac{k_TV_0}{AJ_0} \tag{20}$$

Physically, φ_1 is the ratio of cake resistance to membrane resistance if all the particles in the feed reservoir were placed on the membrane with $\Delta P_c = 0$. Likewise, φ_2 can be viewed as the ratio of the particle removal rate when all the particles in the feed have deposited, to the initial particle deposition rate.

Case 1. Laminar Flow in a Rectangular Module

In this case it is assumed that filtration is taking place in a module with rectangular geometry, in which the tangential flow is laminar and Newtonian. Therefore, the shear stress at the cake surface can be approximated by

$$\tau = \frac{\tau_{w0}}{(1 - \delta/H)^2}\frac{\mu_s}{\mu} \tag{21}$$

where τ_{w0} is the wall shear stress for flow of pure filtrate through a clean channel, δ is the cake thickness at time t, H is the channel half-height, and μ_s is the viscosity of the suspension at time t. While Eq. (21) is strictly true only when the membrane has zero permeability, it is a good assumption when the ratio of flux to crossflow velocity is significantly less than

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unity. This condition is satisfied in most microfiltration operations of practical interest.

Assuming that the particles have a density ρ_s, and that the average particle volume fraction in the cake is ϕ, the cake thickness can be related to the cake mass per unit area through the expression

$$M = \rho_s \phi \delta \tag{22}$$

For compressible cakes, the average particle volume fraction in the cake can be computed using the expression (11)

$$\phi = \phi_0 \frac{1 - n - \beta}{1 - n} \left[\frac{(1 + \Delta P_c^*)^{1-n} - 1}{(1 + \Delta P_c^*)^{1-n-\beta} - 1} \right] \tag{23}$$

where ϕ_0 is the particle volume fraction in the unstressed cake (i.e., at $\Delta P_c = 0$), and β is another empirical constant.

Equations (22) and (4) can be combined to relate the cake thickness to the flux, and Eq. (22) can then be rewritten in terms of the dimensionless flux as

$$\tau = \frac{\tau_{w0} \mu_s^*}{\left(1 - \frac{\varphi_3}{\alpha^* \phi^*} \left(\frac{1}{J^*} - 1\right)\right)^2} \tag{24}$$

where

$$\varphi_3 = \frac{R_M}{\alpha_0 \rho_s \phi_0 H} \tag{25}$$

$$\phi^* = \phi / \phi_0 \tag{26}$$

and

$$\mu_s^* = \mu_s / \mu \tag{27}$$

The suspension viscosity is assumed to be related to the pure filtrate viscosity by the equation

$$\mu_s^* = \left(1 - \frac{1 + \rho_s^*/c_m^*}{1 + \rho_s^*/c^*}\right)^{-2} \tag{28}$$

where c_m is the maximum allowable particle concentration and both c_m and ρ_s have been nondimensionalized with respect to c_0. This expression is the well-known Krieger–Doherty equation written in terms of mass concentrations rather than volume fractions, and with the product of the intrinsic viscosity and maximum packing fraction taken to be equal to 2

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(15). In summary, the dimensionless group, φ_2, can be written in full as

$$\varphi_2 = \frac{\varphi_{20}\mu_s^*}{\left(1 - \frac{\varphi_3}{\alpha^*\phi^*}\left(\frac{1}{J^*} - 1\right)\right)^2} \tag{29}$$

where

$$\varphi_{20} = \frac{k\tau_{w0}V_0}{AJ_0} \tag{30}$$

Equations (16) to (18) can now be solved numerically once the necessary dimensionless groups are specified.

Case 2. Initially Turbulent Flow in a Tubular Module

Here it is assumed that filtration occurs in a tubular module in which the flow is initially turbulent but may become laminar as a result of increasing suspension viscosity. It should be noted that in the analysis presented for tubular systems in this paper, the cake thickness is assumed to be small relative to the tube diameter, and thus the curvature of the cake can be neglected.

The shear stress at the cake surface can be approximated by

$$\tau = \frac{\tau_{w0}}{(1 - \delta/R_0)^4}\frac{f}{f_{w0}} \tag{31}$$

where R_0 is the tube radius, f is the friction factor when the cake has reached a thickness δ, and f_{w0} is the friction factor based on pure filtrate flow through a clean channel. Thus, φ_2 can be expanded as before to give

$$\varphi_2 = \frac{\varphi_{20}}{\left(1 - \frac{\varphi_3}{\alpha^*\phi^*}\left(\frac{1}{J^*} - 1\right)\right)^4}\left(\frac{f}{f_{w0}}\right) \tag{32}$$

where, in this case, φ_3 is defined in terms of the tube radius R_0 rather than the channel half-height H. It is now assumed that the module, both when the membrane is clean and when cake has deposited, exhibits similar friction characteristics to a smooth nonporous pipe. While this assumption obviously neglects the surface roughness of the filter cake and the nonzero wall permeability, it should serve as a useful starting point, particularly when one is only concerned with the qualitative behavior of batch microfiltration.

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In the case of the smooth nonporous pipe, the friction factor can be approximated using the following equation (16):

$$\sqrt{\frac{f}{f_{wo}}} = \frac{\log(6.9/\mathrm{Re}_{wo})}{\log(6.9/\mathrm{Re})} \tag{33}$$

where Re_{wo} is the Reynolds number for flow of pure filtrate through a clean tube. The Reynolds number, Re, for the general case of suspension flow with cake formation, is given by the expression

$$\mathrm{Re} = \frac{\mathrm{Re}_{wo}/\mu_s^*}{\left(1 - \frac{\varphi_3}{\alpha^*\phi^*}\left(\frac{1}{J^*} - 1\right)\right)} \tag{34}$$

where it is assumed that the liquid and solid densities are equal.

Equations (16) to (18) can now be solved numerically for set values of φ_1, φ_{20}, φ_{30}, and Re_{wo}.

SOLUTION METHOD

Since the primary goal of this work is to investigate the relationship between flux and particle concentration, the time variable was eliminated by dividing each of Eqs. (16) and (17) by Eq. (18) and solved with c^* as the independent variable. However, this created the problem of a singularity when $J^* = 1$. In order to overcome this difficulty, Eqs. (16) to (18) were first solved numerically with respect to time in order to obtain a finite initial condition for the equations written with c^* as the independent variable. All equations were solved using the NAG FORTRAN routine D02BBF (NAG Ltd., Oxford, UK).

RESULTS AND DISCUSSION

In this paper we do not present a comprehensive investigation of the model, but focus on its main predictions as to the general shape of the flux versus concentration curve. The lack of experimental data in the literature made it somewhat difficult to assign values to the model parameters. For example, it is clear from its definition that a wide range of values are possible for φ_1, depending on the nature of the suspension, the type of membrane used, the initial concentration, and the initial volume of the feed. Examination of the crossflow filtration literature previously discussed and using the dead-end specific resistance data of Nakanishi et al. (17) indicates that typical values of φ_1 range from 0.001 to 1,000. In this paper we chose an intermediate value of 1. Once a value of φ_1 is

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chosen, the value of φ_{20} can be chosen to give realistic values of dimensionless flux in the plateau region. For typical crossflow filtration equipment, φ_3 will tend to be in the range 1 to 50. Data on P_a is very scarce, but given that P_a is typically considerably smaller than normal operating pressures, our choice of 100 for ΔP^* is reasonable (11). For n, we have chosen values ranging from zero to 0.9, thus spanning a wide range of cake compressibilities. Our choice of 0.1 for β is arbitrary, but calculations have shown that changing β has no impact on the conclusions of this paper. The values chosen for c_m^* and ρ_m^* are appropriate for a range of microbial suspensions (18). The magnitudes of all model parameters used in the calculations are summarized in Table 1.

Case 1

Cake Compressibility

It has been suggested that a possible reason for the sigmoidal relation between flux and concentration is that toward the end of a batch microfiltration run, compression of the filter cake produces a rapid decline in flux (5). This explanation is contradicted by Fig. 2 where it is shown that the sole predicted effect of cake compressibility is not to change the shape of the flux versus concentration curve, but simply to lower the flux. Of interest too is the prediction that the flux is a very strong function of concentration at low concentrations, but very weakly dependent on concentration at high concentrations. This prediction, which is in agreement with experiment (2–5), is due to the fact that at short times dJ/dt is large and dc/dt is small, while the converse is true at large times. Thus the tendency for the flux to vary slowly with time (at large times) is exagger-

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TABLE 1
Numerical Values of Model Parameters

	Figure 1	Figure 2	Figure 3
φ_1	1	1	1
φ_{20}	1	1	1
φ_3	1	1	1
Re_{w0}	—	—	7000
ΔP^*	100	100	100
n	Variable	0.2	0.2
β	0.1	0.1	0.1
c_m^*	25	Variable	12
ρ_s^*	100	100	100

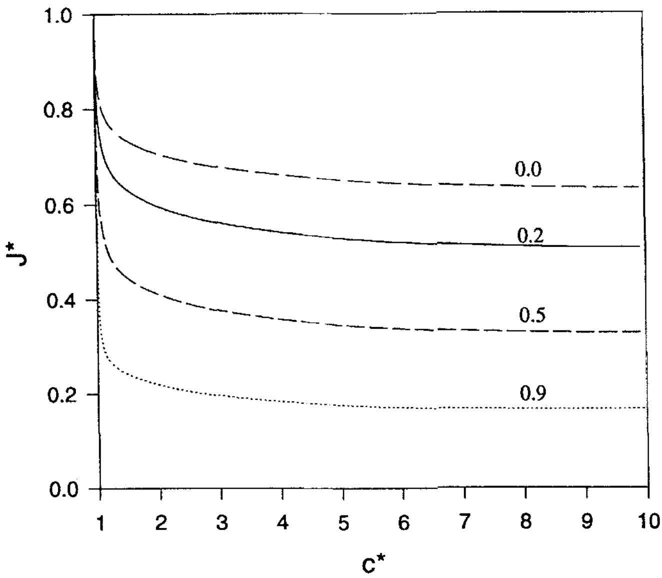

FIG. 2 J^* versus c^* for various values of n.

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ated by plotting it against the rapidly-varying concentration. This is a contributing factor in the occurrence of the so-called plateau region mentioned in the Introduction.

Suspension Viscosity

Figure 3 indicates that increasing suspension viscosity can produce regions of constant flux, and, if the concentration becomes high enough, lead to a situation where the flux actually increases with increasing concentration. This is in agreement with experiment (2, 4) and is consistent with the qualitative explanation of Pritchard et al. (4) who asserted that increasing fluxes were due to enhanced wall shear stress brought about by increased suspension viscosity. It should be noted, at this point, that increasing suspension viscosity is the only mechanism by which the flux can increase with increasing concentration, at least according to the predictions of this model. Extensive calculations have shown that increasing wall shear stress brought about solely by the reduction of the effective channel height does not produce such an effect.

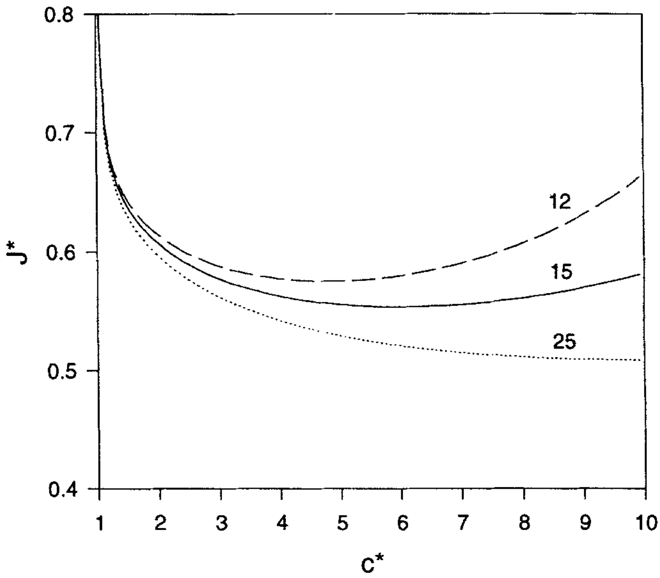

FIG. 3 J^* versus c^* for various values of c_m^*.

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Case 2

Turbulent-to-Laminar Transition

In order to investigate whether a sigmoidal curve could be predicted by the model, it was assumed that such behavior is due to a change in the cake removal rate as a result of the transition from turbulent to laminar flow. This pheneomenon was modeled by assuming that over an appropriate range of Reynolds numbers, the cake removal constant, k, decreases. Previous workers have suggested that the change in cake removal rate occurs at a Reynolds number of about 2300 (4). In the calculations reported in this paper, it is assumed that k (and hence φ_{20}) undergoes a linear drop from its initial value in the turbulent flow region to its final value in the laminar flow region. This drop was assumed to occur between Reynolds numbers of 3000 and 2000. Typical calculations for tubular systems are given in Fig. 4. Curve A demonstrates that if no change in φ_{20} occurs, then a plateau region is observed as before. However, in Curve B it is shown that when, for example, a tenfold drop in φ_{20} occurs, sigmoidal-like behavior occurs, but this is followed by a new plateau region. This

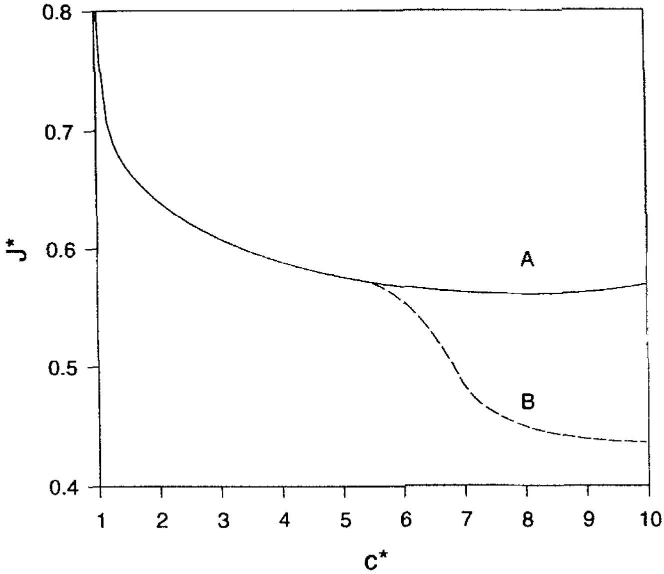

FIG. 4 J^* versus c^*. Curve A: no change in cake removal constant, k. Curve B: 10-fold linear drop in k between Re = 3000 and Re = 2000.

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is in agreement with the data of Mourot et al. (2) and Pritchard et al. (4). It should be noted, also, that if the concentration is allowed to increase sufficiently, then the flux will eventually increase with concentration due to increasing suspension viscosity. However such a region of increasing flux is unlikely to be accessible experimentally.

CONCLUSIONS

The model presented in this paper provides confirmation that the distinguishing features of the flux versus concentration curve in batch microfiltration are not indicative of cake properties, but of suspension rheology. Future work will involve modifying the model to take account of membrane fouling and performing experiments to test the model in a more quantitative way.

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