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STEADY-STATE PERMEATE FLUX OF CROSSFLOW CERAMIC MICROFILTRATION

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This paper deals with both theoretical and experimental studies of the ceramic microfiltration of dilute model suspensions. Steady-state permeate fluxes were analyzed and a simple model based on the critical flow velocity theory has been developed and tested in this study. This model predicts satisfactorily the steady-state permeate flux of crossflow microfiltration under various operating conditions.

Introduction

The crossflow microfiltration (CFMF) technique, which can remove particles of 0.1 - 10 μm effectively, has been widely developed and utilized in water and waste water treatment processes. The major factor affecting the separation efficiency of CFMF is the formation of a cake layer on the surface of the membrane. The permeate flux decreases with filtration time due to increasing hydraulic resistance of the growing cake layer. However, for every operating condition, a steady-state will be reached where the cake thickness and permeate flux remain constant. This is in contrast to the conventional filtration process in which the cake keeps growing with filtration time.

The CFMF has become a more attractive process in recent years owing

to the advent of ceramic membranes. Ceramic membranes exhibit unique physical and chemical properties which are only partially shown up (or are completely absent) by polymeric membranes. For example, they can be used at significantly higher temperatures, have better structural stability without the problems of swelling or compaction, generally can withstand more harsh chemical environments, are not subjected to microbiological attack, and can be backflushed, steam sterilized or autoclaved. They are made of macroporous structure coated on the inside with a thin microporous layer having pore sizes in the range from submicron size to 10 μm[1-3]. At present they are used widely in the processing industry, e.g. in water and waste water treatment processes, dairy products and oil water emulsions.

In a recent paper[4], more specially concerned with crossflow microfiltration of latex waste water on alumina membranes, we have demonstrated that a limiting flux was observed which depended on the species of the latexes and on the flow velocity of the feed. The decrease in the flux of permeate was attributed to the resistance of the gel layer formed on the membrane or blocking of the pores in the membrane. By these means, low permeate fluxes were obtained at relatively high solution velocity.

The aim of this study is to gain an insight into the importance of some processes parameters in the crossflow microfiltration of suspensions using ceramic membranes. In addition, a simple model based on the critical flow velocity theory is proposed in this study to predict the steady-state flux of CFMF under various operating conditions.

Theory

Three assumptions were made in the development of the model:

1. Particles cannot penetrate into or through the membrane.
2. The cake formed is relatively incompressible.
3. A particle deposited beneath the membrane to form the cake during steady-state will not be resuspended.

When a stable cake layer has built up on the inside of a tubular membrane, the pressure drop across the cake layer is much greater than that across the membrane and its support. So the major resistance to flow is controlled by this cake which is produced by mechanisms dominated by the hydrodynamic processes in the tube. Both cake deposition and removal are possible. Figure 1 illustrates the process of filtration through a cake layer, which is formed on a membrane of inner radius R to a depth of $R - R_o$ on its surface. The permeate flux, J, is based on the inner radius of the membrane and is expressed in $l\,m^{-2}\,h^{-1}$ of inner surface. The flow of fluid or suspension through the tube in crossflow is based on the cross sectional area of the membrane πR^2

and has value u. The actual mean velocity through the fouled tube u_C, however, is given by

$$u_C = \frac{uR^2}{R_o^2} \tag{1}$$

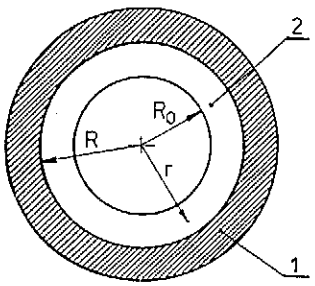

Fig. 1 Microfiltration through a cake layer on the tubular membrane: 1 membrane, 2 cake layer

The rate at which a cake will build up on the inside of the tube will increase with the concentration of solid in the feed and with the rate of permeate production. A cake will be removed by the cross-flowing fluid at a rate dependent on the wall shear stress and the inner surface area of the cake layer exposed to the crossflow fluid. Hence the rate of removal will be greater for smaller values of R_o. As the rate of build up of fouling is proportional to the permeate flux J, an equilibrium situation will be reached when the rates of build up and removal become equal.

If P is the pressure in the cake layer at distance r from the centre of the tube then

$$\frac{dP}{dr} = J\frac{\mu}{k}\frac{R}{r} \tag{2}$$

where k is the permeability of the cake layer. Integration of Eq. (2) from $r = R_o$ to $r = R$ and with ΔP_C equal to the pressure drop in the cake layer gives

$$\frac{\Delta P_C}{J} = \frac{\mu}{k} R \ln\frac{R}{R_o} = \frac{\mu R}{2k} \ln\left(\frac{R}{R_o}\right)^2 \tag{3}$$

The relationship between the transmembrane pressure and pressure drop in the cake layer can be expressed as follows

$$\Delta P_C = \Delta P - \Delta P_M = \Delta P - \mu R_M J \tag{4}$$

where ΔP is transmembrane pressure, ΔP_M is pressure drop in the membrane, and R_M is hydraulic resistance of the membrane.

There are several models[5] which can be used to determine the ultimate

value of R/R_o; probably the simplest is to consider that equilibrium between cake build up and removal occurs at fixed critical velocity in the tube, thus

$$u_C = u_{C,cr} = u\left(\frac{R}{R_o}\right)^2 \tag{5}$$

where $u_{C,cr}$ is the critical velocity at which the cake layer disappears. So substitution back into Eq. (3) gives

$$\frac{\Delta P - \mu R_M J}{J} = \frac{\mu}{k} R \ln \frac{u_{C,cr}}{u} \tag{6}$$

Equation (6) can then be rearranged to calculate the steady-state permeate flux J_{SS} as

$$J_{SS} = \frac{\Delta P}{\mu\left[\frac{R}{2k}\ln\left(\frac{u_{C,cr}}{u}\right) + R_M\right]} \tag{7}$$

The validity of the steady-state flux prediction equation (7) for the crossflow microfiltration of diluted suspensions is discussed using experimental data obtained with alumina/water, PVC/water and previously reported data for magnesia/water suspension[6].

Experimental

Membranes

The ceramic membranes studied in this work were asymmetric, double-layered, alumina membranes (TERRONIC, Czech Republic). They were configured as single cylindrical tubes either 0.1 m long, 6mm ID and 10mm OD, or 0.2 m long, 12 mm ID and 17 mm OD, consisting of a thin α-alumina layer deposited at the internal surface of the tubular alumina support. In our experiments, microfiltration membranes were used with the mean pore diameter equal to 0.2 μm. The pore size distribution of the membrane used (see Fig. 2) was determined by the liquid displacement method[7]. The data presented in Fig. 2 indicate that the double-layered ceramic membrane used in these studies has a very narrow pore size distribution. This narrow distribution is extremely useful for accurately predicting the performance of tubular ceramic membranes in different applications.

Feeds

Two kinds of feed were used in the crossflow apparatus, i.e. commercial suspensions of poly(vinylchloride) (PVC), and an alumina (Al_2O_3) powder. These were suspended in deionized water. The particle size distribution of the dispersion used, shown in Fig. 2, was determined by a particle sizer SediGraph 5100 (Micromeritics). Concentration of solids in the suspensions was constant (1 % w/w).

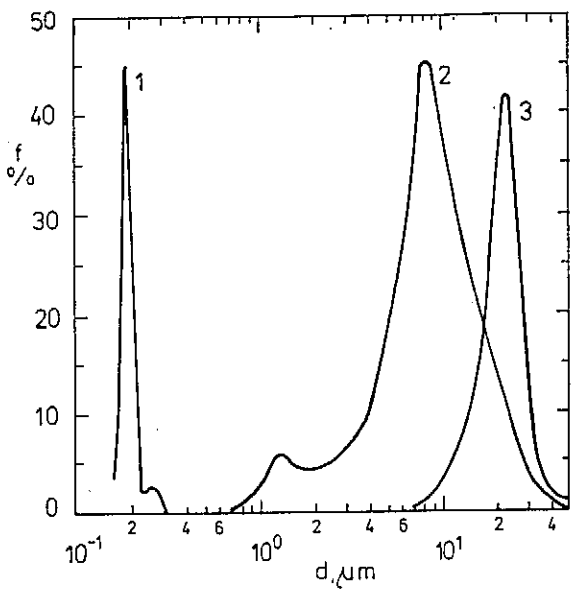

Fig. 2 Pore size distribution of 0.2 µm membrane (1) and particle size distribution of PVC (2) and Al_2O_3 (3) suspension used

Equipment

The microfiltration studies were carried out in a membrane filtration unit equipped with membrane module formed from membrane placed in an acrylic casing. The suspensions were circulated through the module by a centrifugal pump with the feed flowing in parallel direction to the membrane surface and perpendicularly to the permeate. A schematic diagram of the experimental apparatus is shown in Fig. 3. It consisted mainly of a microfiltration module, a pump, a storage tank equipped with a thermal regulation system, and a temperature and pressure control system. The unit allowed studies in which the transmembrane pressure and the crossflow velocity were independently varied.

The experiments were carried out at 20 °C. New membrane was used in each experiment and the pure water flux was measured with prefiltered

deionized water before the run. A suspension was then introduced to the unit, the pump was powered on and operating pressure and superficial velocity (it is worth recalling that superficial velocity, u, is defined in the system as the ratio of volumetric flow rate to membrane tube cross-sectional area) adjusted by the regulating system. The flux through the membrane was measured by collecting the permeate into graduated cylinder and timing the collection period. Both permeate and retentate were recycled to the storage tank to maintain a relatively constant suspension concentration. Every experiment was carried out until the flux became virtually constant. Duplicate microfiltration experiments for selected conditions showed good reproducibility of the data measured.

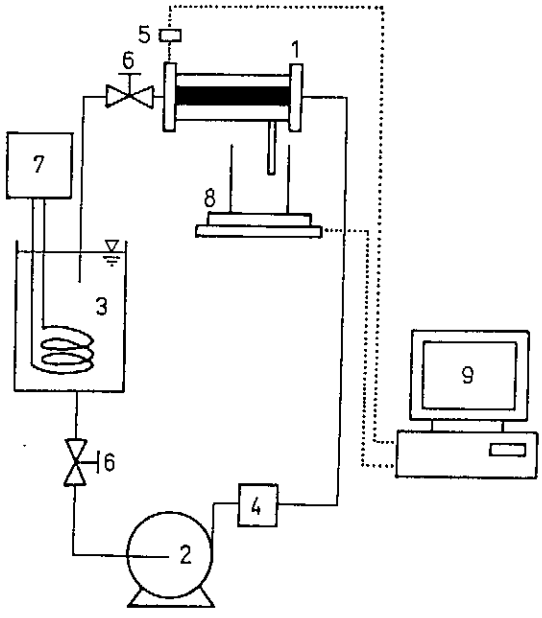

Fig. 3 Scheme of experimental apparatus: 1 membrane module, 2 pump, 3 feed tank, 4 damper, 5 pressure transducer, 6 regulating valve, 7 thermostat, 8 electric balance, 9 computer

Results and Discussion

Analysis of the permeate composition showed complete retention of the alumina particles in the feed. Note that the diameter of the dispersion particles was always much larger than the membrane pore size (see Fig. 2) in order to prevent particle penetration into or through the membrane.

Values of flux versus time for various mean feed velocities are plotted in Fig. 4 for alumina/water suspension and in Fig. 5 for the PVC/water suspension used. As a general rule, the steady-state flux during crossflow microfiltration

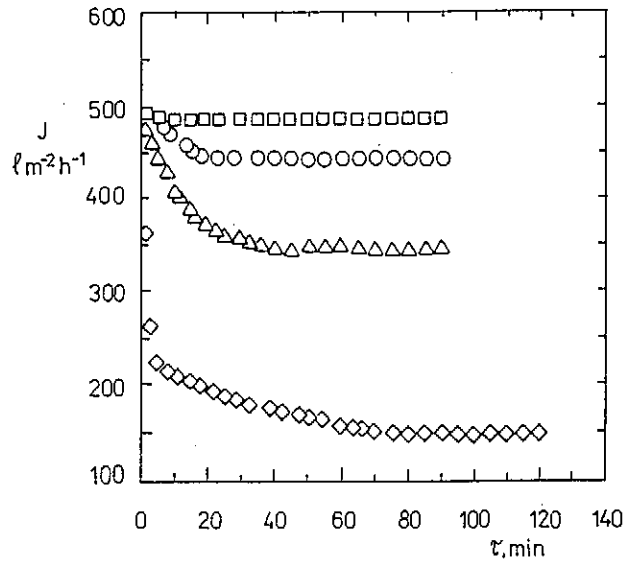

Fig. 4 Variation of the permeate flux with time during microfiltration of alumina/water suspension: ◇ u = 0.05 m s^{-1}; △ u = 0.10 m s^{-1}; ○ u = 0.20 m s^{-1}; □ u = 0.28 m s^{-1} (concentration 1% solids, transmembrane pressure 50 kPa)

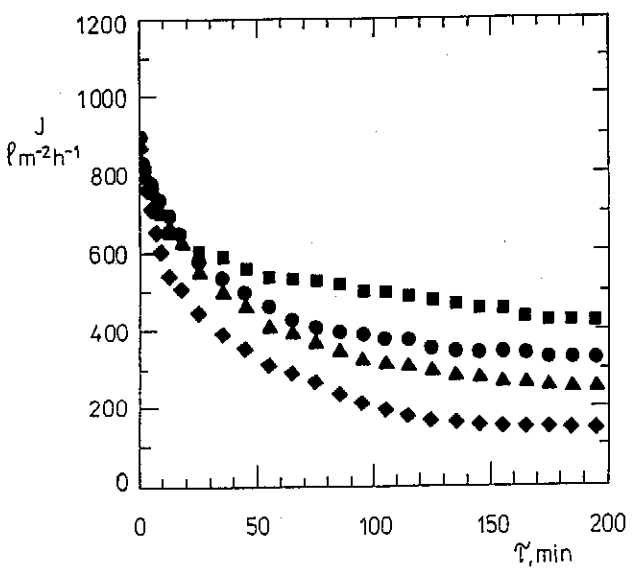

Fig. 5 Variation of the permeate flux with time during microfiltration of PVC/water suspension: ♦ u = 0.08 m s^{-1}; ▲ u = 0.94 m s^{-1}; ● u = 2.21 m s^{-1}; ■ u = 3.00 m s^{-1} (concentration 1% solids, transmembrane pressure 100 kPa)

was substantially lower than the pure water flux - ranging from 10% to 95% of pure water values. Besides the fact that high levels of permeate flux are

achievable (for comparison see Ref.[3]), the most noticeable feature in Fig. 4 is the strong dependence of the flux on the superficial velocity of the feed. The following trends are evident:

(i) Significant flux decay was observed mainly in the initial periods of the process, and
(ii) the flux decline shows significant dependence on operating conditions such as superficial velocity.

As the microfiltration proceeds, the filtration rate reaches the limiting or steady-state value. The limiting value is approached when the further growth of the filter cake is hindered by the applied axial fluid shear upon the system. Depending on the shear forces developed by the feed flow, the cake thickness is decreased and in the limiting case the cake cannot be created any more under given hydrodynamic conditions. It is evident from Figs 4 and 5 that this steady-state flux increases (the thickness of the cake layer and its hydraulic resistance decreases) with the superficial velocity.

The ceramic membrane data were obtained with tubes of a different inner radius so that the variation of J with R as shown in Eq. (3) could be proved in this set of experiments. When $\Delta P_C / J$ was plotted against the natural logarithm of $1/u$, straight line plots were obtained as shown in Fig. 6. Thus, the permeability of the cake layer k was obtained from the slope of the straight line plots in Fig. 6.

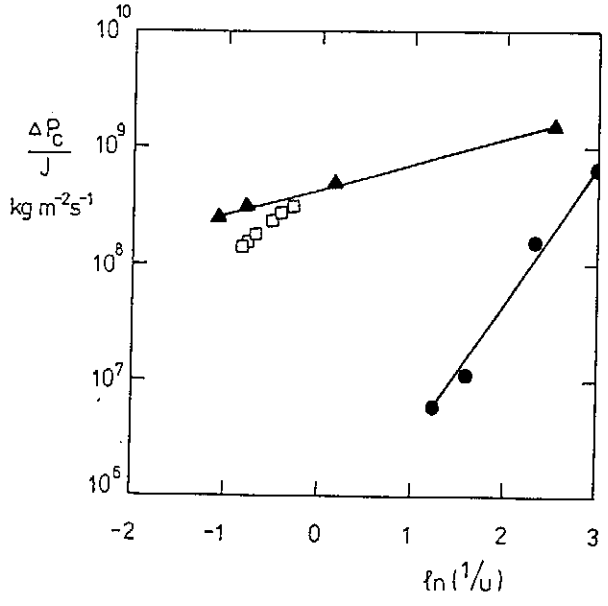

Fig. 6 Plot of experimental values of pressure drop in cake layer and flux ratio vs. ln(1/u) for different suspensions: ● Al_2O_3/water; ▲ PVC/water; □ $Mg(OH)_2$/water[6]

The model outlined in this paper permits the correlation of microfiltration data to be carried out by determining the critical feed velocity $u_{C,cr}$. If the flow velocity is large enough, the model predicts that the resistance of the cake layer is minimal. Therefore, the experimental results were then recalculated and a satisfactory plot was obtained as shown in Fig. 7. This Figure shows that the plots of $(\Delta P_C - \Delta P_M)$ against u for different dilute suspensions are straight lines. It is interesting to note that the data for feeds of low concentration shown that $(\Delta P_C - \Delta P_M)$ tends to zero as u increases. Referring back to Eq. (3) when $(\Delta P_C - \Delta P_M) = 0$, we find the critical feed velocity at which the cake layer disappears.

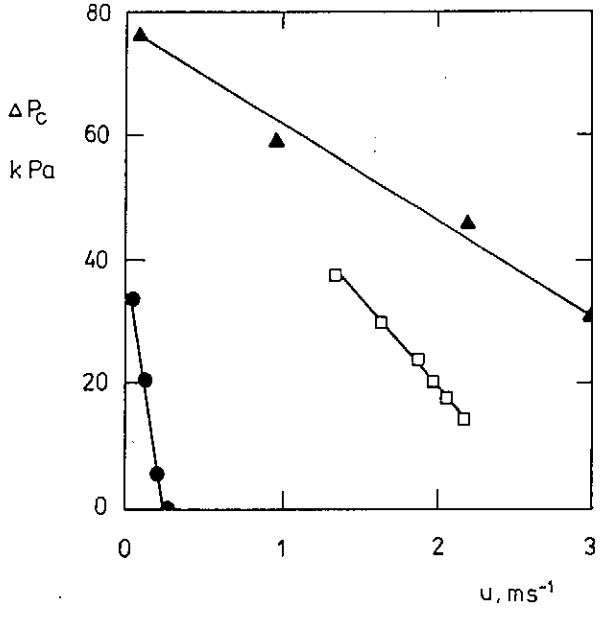

Fig. 7 Estimation of the critical superficial velocity during the ceramic crossflow microfiltration of different suspensions: ● Al_2O_3/water; ▲ PVC/water; □ $Mg(OH)_2$/water[6]

Finally, to illustrate the model predictions, Table 1 gives an example calculated for the case of magnesia [$Mg(OH)_2$]/water suspension[6]. The physical properties and geometric dimensions valid for the steady-state permeate flux experiments were:

$R_M = 2.73 \times 10^{11}$ m^{-1}, $\Delta P = 64$ kPa, $\mu = 1.002 \times 10^{-3}$ Pa s,
$R = 5 \times 10^{-3}$ m, $u_{C,crit} = 2.67$ m s^{-1}.

It should be noted that there is good agreement between model predictions and experimental results. This shows that the steady-state permeate flux of dilute

suspensions has been correlated by the proposed simple model.

Table I Comparison of the steady-state flux experimental results[6] with predictions according to Eq. (7)

Exp. No.	u m s^{-1}	$J_{SS,exp}$ l m^{-2} h^{-1}	$J_{SS,cal}$ l m^{-2} h^{-1}	δ %
1	1.34	229	236	-3.1
2	1.66	288	301	-4.5
3	1.88	342	370	-8.2
4	1.95	367	393	-7.1
5	2.02	383	416	-8.6
6	2.17	425	463	-8.9

$$\delta = \frac{J_{SS,exp} - J_{SS,cal}}{J_{SS,exp}} \times 100$$

Conclusions

Experiments were conducted to study crossflow microfiltration of dilute suspensions of PVC and alumina particles in water using ceramic membranes. It was found that the steady-state permeate flux increased with increasing superficial velocity. However, it decreased with decreasing particle size.

Attempts have also been made to explain the process behaviour in terms of the simple model of the operative mechanisms. The model was examined with particular attention to the critical feed velocity at which the cake layer disappears. It was found that the application of the model outlined in this paper shows good agreement between model predictions and experimental results.

The performance of this system, at the moment, cannot be taken to be a general case but rather system specific. A deeper understanding of the mechanism of particle deposition on the surface of the membrane and more detailed information between crossflow velocity, transmembrane pressure, concentration and permeate flux performance is required.

Acknowledgements

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Symbols

J	permeate flux, $1\,m^{-2}\,h^{-1}$
J_{SS}	steady-state permeate flux, $1\,m^{-2}\,h^{-1}$
k	permeability of cake layer, m^2
ΔP	transmembrane pressure, Pa
ΔP_C	pressure drop in the cake layer, Pa
ΔP_M	pressure drop in the membrane layer, Pa
r	coordinate, m
R	radius of membrane tube, m
R_o	radius of cake layer, m
R_M	hydraulic membrane resistance, m^{-1}
u	crossflow velocity, $m\,s^{-1}$
u_C	crossflow velocity in the fouled tube, $m\,s^{-1}$
$u_{C,cr}$	critical crossflow velocity in the fouled tube, $m\,s^{-1}$
δ	relative deviation
μ	dynamic viscosity, Pa s
τ	time, s

Indexes

exp	refers to experimental value
cal	refers to calculated value

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