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Cross-Flow Microfiltration of Fine Particles Suspended in Polymeric Aqueous Solution

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Abstract: The filtration characteristics of cross-flow microfiltration of fine particles suspended in polymeric aqueous solution are studied. Polymethylmethacrylate (PMMA) submicron particles are suspended in polyacrylamide (PAA) aqueous solution to prepare the suspensions used in experiments. Effects of operating conditions, such as cross-flow velocity, filtration pressure, and PAA concentration, on the filtration flux and the cake properties are discussed. The results show that an increase in cross-flow velocity or filtration pressure causes the filtration flux to be higher, but the filtration flux decreases with an increase in PAA concentration. Since the flow behavior indices of three prepared suspensions are almost the same, the average specific filtration resistance of cakes under various cross-flow velocities and PAA concentrations remain almost constant; and then the cake mass plays a major role in determining the filtration resistance and the filtration flux. A force balance model is derived for particle deposition on the membrane surface. Once the empirical coefficients are obtained from experimental data, the filtration flux at pseudo-steady state can be predicted accurately.

Keywords: Cross-flow microfiltration, cake properties, particle deposition, polymeric solution, solid-liquid separation

INTRODUCTION

Cross-flow microfiltration is an efficient mode for recovering high valued products or removing fine particles from a liquid. This mode of filtration

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has attracted the attention of many plant engineers and researchers because of its advantages, e.g. thin cake layer, high filtration flux, and long operation time, etc. In recent years, many theoretical models have been proposed in order to estimate the filtration flux under various operating conditions, such as concentration polarization models (1, 2), hydrodynamic models (3, 4), etc. However, the dispersed media of suspensions in most researches are limited to Newtonian fluids.

The filtration of fine particles in polymeric solutions often occur in many industrial fields, such as biotechnology, chemical industries, food processing industries, petroleum, and polymer engineering, etc.; therefore, it is worth paying more attention to this subject in future research. In the past, Shirato et al. (5) and Kozicki (6, 7) derived a Ruth-type filtration equation based on the modified Rabinowitsch-Mooney equation. They concluded that a more compact cake would be formed in the filtration of power law slurry compared to that of Newtonian, and that the cake porosity decreased with the decrease of flow behavior index of the dispersed fluid (5). The value of the new-defining average specific filtration resistance of cake increased with the increase of filtration pressure or the flow behavior index of dispersed fluid (7). In 1997, Hwang and Lu (8) proposed a force balance model for estimating the critical friction angle of depositing particles in cross-flow filtration of power law slurry. The probability of particle deposition, the particle size distribution, and the growth rate of cake could then be calculated. A dynamic simulation method had also been proposed by the same authors (9) to estimate the variations of local cake properties during a course of cross-flow filtration. However, the particle sizes used in previous research ranged from several microns to hundred microns. In order to be applied to fine chemical processes, to study the filtration characteristics of cross-flow microfiltration of fine (especially submicron-sized) particles suspended in Non-Newtonian solutions will increase its importance very quickly.

In this article, the effects of operating conditions on the filtration flux and cake properties in cross-flow microfiltration of fine particles suspended in polymeric aqueous solution are studied and discussed. A force balance model for depositing submicron particles on the membrane surface is derived in order to relate filtration flux and operating conditions.

THEORY

Basic Filtration Equations

If the flow behavior of suspension follows the power law, the basic filtration equation can be written as (5, 6):

$$q^n = \frac{\Delta P}{K(R_c + R_m)} = \frac{\Delta P}{K(\gamma_{av} \cdot w_c + R_m)} \tag{1}$$

where q is the filtration flux, ΔP is the filtration pressure, γ_{av} is the average specific filtration resistance of cake, w_c is the cake mass, R_m is the medium filtration resistance, while n and K are the fluid consistency index and flow behavior index, respectively. In general, R_m is negligibly small compared to cake filtration resistance.

Based on a material balance of the formed cake, the cake mass can be calculated by the following equation:

$$w_c = \rho_s(1 - \epsilon_{av})L \tag{2}$$

where ρ_s is the particle density, ε_{av} is the average cake porosity and L is the cake thickness. Hence, in keeping with Eq. (1), the analyses on cake properties, such as γ_{av}, ε_{av}, L and w_c are essential to understand the filtration flux in a cross-flow microfiltration of power law slurry.

Force Balance Model for Particle Deposition

Some particles in the suspension are carried by the filtrate to migrate toward the membrane surface during a filtration. However, a few of them have opportunities to deposit onto the filter membrane to form a filter cake. Figure 1 shows a schematic diagram of the region around the membrane surface in a cross-flow microfiltration system. *Particle A* is just arriving at the position

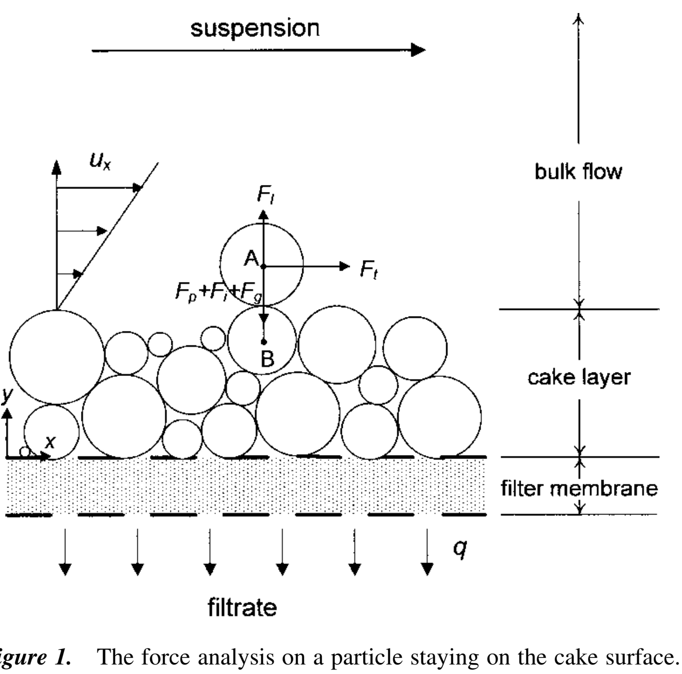

Figure 1. The force analysis on a particle staying on the cake surface.

on the surface of *particle B*. Whether *particle A* can deposit stably or not is determined from the external forces exerted on it. In general, the forces include the drag forces due to fluid flow, the interparticle forces, the lift force due to inertial effect, and the gravitational force. However, the inertial lift and the gravitational forces are negligible small for submicron particles in most operating conditions. The particle can deposit stably only if the frictional force between particles is larger than the net force in the opposite direction; otherwise, the particles will be swept away from the contact point. The force balance model at the critical condition (or pseudo-steady state) can be expressed as (4):

$$F_t = f_c(F_p + F_i) \qquad (3)$$

where f_c is the friction coefficient, F_i is the interparticle force, and F_t and F_p are the drag forces in the directions of cross-flow and filtration, respectively. These forces are analyzed as follows.

According to the analysis of Hwang and Lu (8) for a two-parallel-plate microfilter, the drag in cross-flow direction can be calculated by

$$F_t = 2\pi K a^{2-n} u_A^n \cdot C_1' \qquad (4\text{-}1)$$

or be written in the following form:

$$F_t = C_1 a^{2-n} \cdot \tau_w \qquad (4\text{-}2)$$

where a is the particle radius, u_A is the fluid velocity at the particle centre and τ_w is the shear stress on the membrane surface. The C_1' and C_1 are correction factors and are constants for a given fluid.

The drag force in the filtration direction can be given by (8)

$$F_p = 2\pi K a^{2-n} q_s^n \cdot C_2' \qquad (5\text{-}1)$$

or

$$F_p = C_2 a^{2-n} \cdot q_s^n \qquad (5\text{-}2)$$

where q_s is the filtration flux at pseudo-steady state.

The interparticle force in a polymeric solution is mainly due to the adsorption of polymer molecules on the cake surface. It is too difficult to obtain this force from a theoretical analysis because of the complicate phenomena. Consequently, it is better to estimate it by an experimental method, e.g. the methods described in Hwang and Lu (8, 9). According to the analysis of Hwang and Lu (8), this force is a function of polymer concentration, particle size and shear stress on the membrane surface.

For a uniform-sized particulate sample, a linear relationship among F_i, τ_w and C_o can be expected, that is (8),

$$F_i = C_3' f(\tau_w, C_o) \\ = (C_3 - C_4 \cdot C_o) \cdot (\tau_w - C_s) \tag{6}$$

where C_3',C_3, C_4, and C_5 are empirical coefficients.

Substituting Eqs.(4-2), (5-2) and (6) into Eq.(6), the force balance equation for a uniform-sized submicron particulate sample at the critical condition becomes

$$q_s^n = (C_6 - C_7 \cdot C_0)\tau_w + (C_8 - C_9 \cdot C_0) \tag{7}$$

in which C_6–C_9 are empirical coefficients and can be obtained in experiments. Therefore, a linear relationship between q_s^n and τ_w can be expected for a given polymer concentration.

Based on the analysis of Hwang and Lu (8), the shear stress acting on the membrane surface in a two-parallel-plate cross-flow microfilter can be given by

$$\tau_w = K\left[\left(\frac{u_s}{H - L}\right)\left(\frac{4n + 2}{n}\right)\right]^n \tag{8}$$

If the cake thickness, L, is far smaller than the filter clearance, H, the value of τ_w can be obtained directly from the original average cross-flow velocity and the flow behavior indices. If the cake thickness cannot be neglected compared to H, it can be calculated by Eq.(2) once the mass and porosity of cake are measured in experiments. Consequently, the filtration flux at pseudo-steady state can be estimated by Eq.(7) from operating conditions once the empirical constants are known.

EXPERIMENTAL

Polyacrylamide (PAA), a kind of polymer, with a mean molecular weight of 200,000 Da, was dissolved in a de-ionized water to prepare the dispersed medium. Three concentrations of the aqueous polymer solutions, 0.5, 1.0 and 1.5 kg/m^3, were prepared in this study. The densities of these solutions were almost equal to that of pure water. The rheology of PAA solutions were measured by a *Brookfield DV-II+* viscometer, the elastic behavior of fluid could be neglected within the operating conditions of this study due to the low shear rates ($<200\,s^{-1}$) and dilute PAA solutions ($<1.5\,kg/m^3$). In other words, the polymer solution exhibits a pure viscous behavior, which could be reasonably modeled to a power low fluid. The measured values of fluid consistency index and the flow behavior index for three aqueous polymer solutions used in this study were listed in Table 1. Although the values of n were almost the same, the apparent viscosities of these solutions were increased with PAA concentration due to the increase of K. The

Table 1. The fluid consistency index and flow behavior index of PAA solutions with various concentrations

PAA concentration (kg/m^3)	K ($N s^n/m^2$)	n (−)
0.5	0.059	0.638
1.0	0.097	0.633
1.5	0.128	0.628

spherical particles made of polymethyl methacrylate (PMMA) manufactured by Soken Co. of Japan, were used as the fine particulate sample in the experiments. The density of the particles was $1210\,kg/m^3$. The particle size distribution was very narrow and its mean diameter was $0.4\,\mu m$. The pH of the suspension was kept at 10.8 by adding NaOH to avoid the occurrence of particle flocculation. The zeta potential of PMMA particles in such a condition was measured as $-28\,mV$. The filter membrane made of mixed cellulose ester with a mean pore size of $0.1\,\mu m$ was used as the filter medium in filtration experiments.

A schematic diagram of the two-parallel-plate cross-flow filtration system used in this study was shown in Fig. 2. The filtration area in the filter was $1.4 \times 10^{-3}\,m^2$, while the clearance between two plates was $0.0035\,m$. PMMA particles were added slowly into PAA solutions to prepare 0.1wt% suspensions. The suspensions were well mixed by a stirrer and pumped into the filter by a circulation pump. The concentrate was recycled back into the slurry tank, while the filtrate was collected into a receiver. The weight of the filtrate was *in situ* detected by a load cell and recorded in a personal computer. The formed filter cake was sent to determine its wet and dry

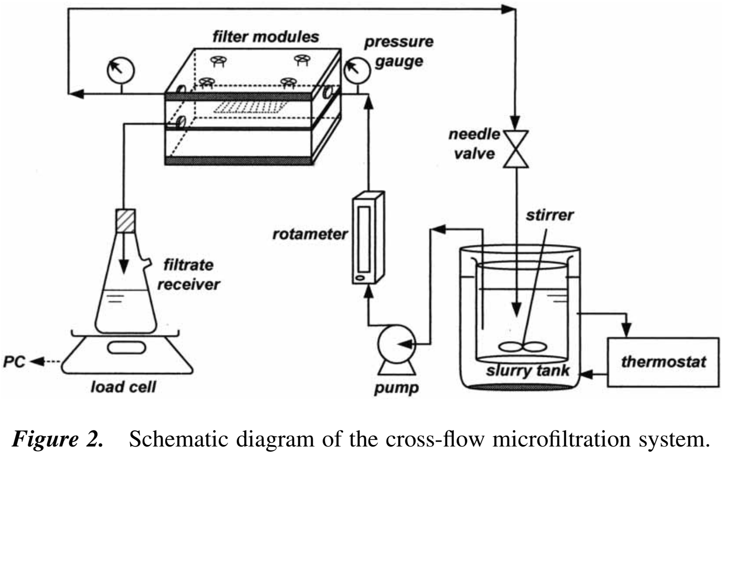

Figure 2. Schematic diagram of the cross-flow microfiltration system.

masses using a thermal gravimetric analyzer (TGA) after filtration. The mass, the average porosity and the average specific filtration resistance of cake could then be calculated accordingly.

RESULTS AND DISCUSSION

Figure 3 shows the attenuations of filtration fluxes during cross-flow microfiltration of PMMA particles suspended in three different concentrations of PAA aqueous solutions. The filtration fluxes decay very quickly at the beginning of filtration and then approach pseudo-steady values as the filtration time exceeds 1000 seconds. This is because the cake formation is limited by the shear stress acting on the membrane surface, and this trend is the same as those in most cross-flow filtration. This is the reason why this mode of filtration can be operated for a longer time than those "dead-end" modes. Furthermore, it can be seen that the filtration flux decreases with increasing PAA concentration. This is because of the increase of fluid apparent viscosity.

Figure 4 shows the effects of operating conditions on the pseudo-steady filtration flux. The filtration flux decreases with the increase of PAA concentration. The filtration flux in the condition of $C_o = 1.5\,\mathrm{kg/m^3}$ is about a half value of that in $C_o = 0.5\,\mathrm{kg/m^3}$ under a given cross-flow velocity and filtration pressure. Although the flow behavior indices of these two solutions are almost the same, the difference of the fluid consistency index between

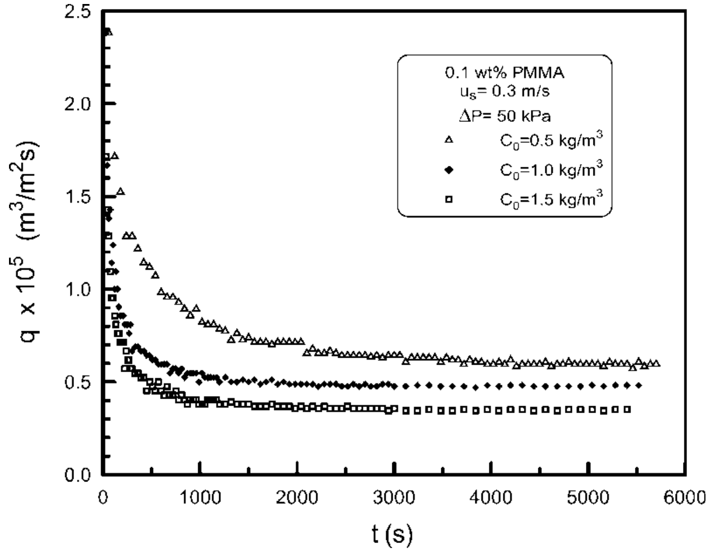

Figure 3. The time courses of filtration fluxes during cross-flow microfiltration for three suspensions with different PAA concentrations.

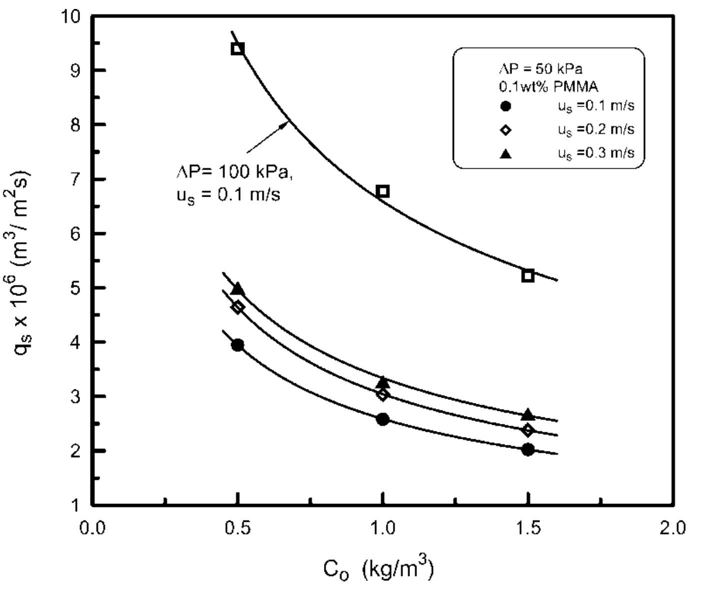

Figure 4. Effects of operating conditions on the pseudo-steady filtration flux.

them results in different flow resistances of the fluids. The other possible reasons are due to the effects of cake properties. These effects will be discussed later. It can be also noticed in this figure that an increase in cross-flow velocity leads to a higher pseudo-steady filtration flux for a given PAA concentration. This is because a thinner cake may be formed under a higher cross-flow velocity. The lighter the cake mass, the lower the filtration resistance will be. Moreover, compared with the results under different filtration pressures, a double increase in filtration flux can be obtained as filtration pressure increases twofold. This implies that the cake filtration resistance varies trivially with filtration pressure; and the filtration flux is mainly determined by the filtration pressure (the filtration driving force). Comparing the results shown in this figure with those in previous research on Newtonian suspensions (10), the filtration fluxes of the same PMMA particles suspended in water are at least one order of magnitude higher than those suspended in polymer solutions.

Figure 5 illustrates the relationships among the filtration resistance of cake, R_c, the average specific filtration resistance of cake, γ_{av}, and the cake mass, w_c. Although these data are measured under various cross-flow velocities and PAA concentrations, they can be regressed as straight lines. The values of γ_{av} remain almost constant under various operating conditions. Since the flow behavior indices of those suspensions are almost the same, this result implies that γ_{av} is only a function of n under a given filtration pressure. In other words, the same flow behavior index causes a similar filtration characteristic of filter

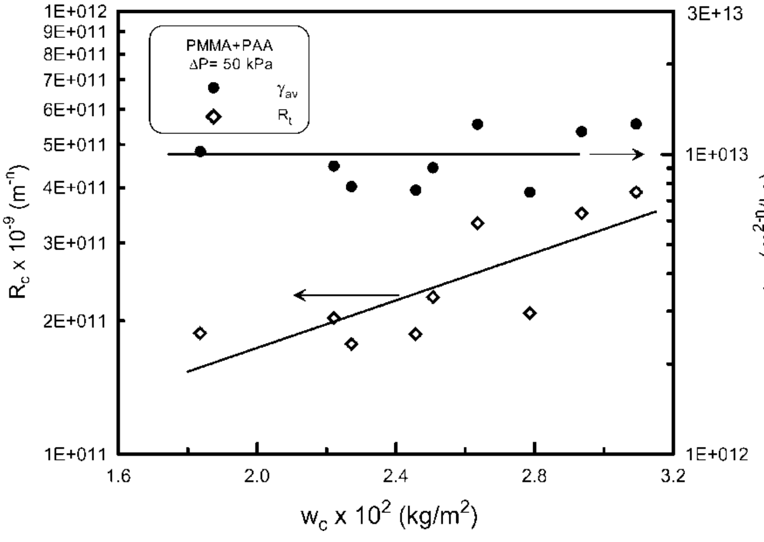

Figure 5. The relationships among the filtration resistance of cake, the average specific filtration resistance of cake, and the cake mass under various conditions.

cakes. Since γ_{av} remains constant, the filtration resistance of cake will be in proportion to the cake mass. This trend can be easily seen in Fig. 5. Thus, under the same filtration pressure, the cake mass plays the major role in determining the filtration resistance as well as the filtration flux.

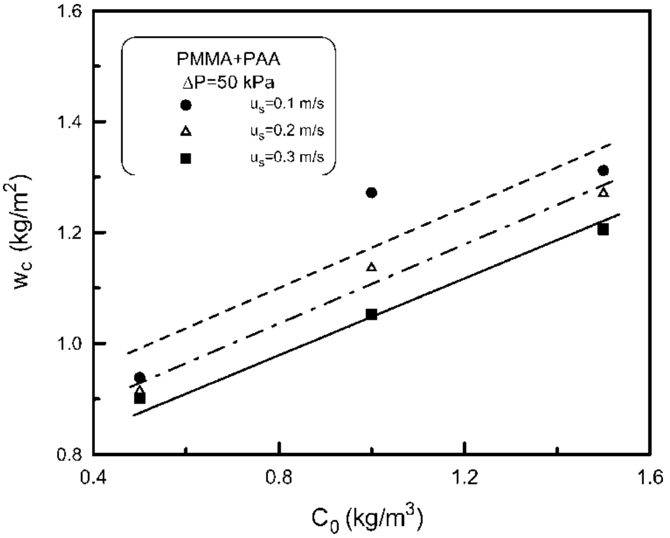

Figure 6. The cake masses formed under various cross-flow velocities and PAA concentrations.

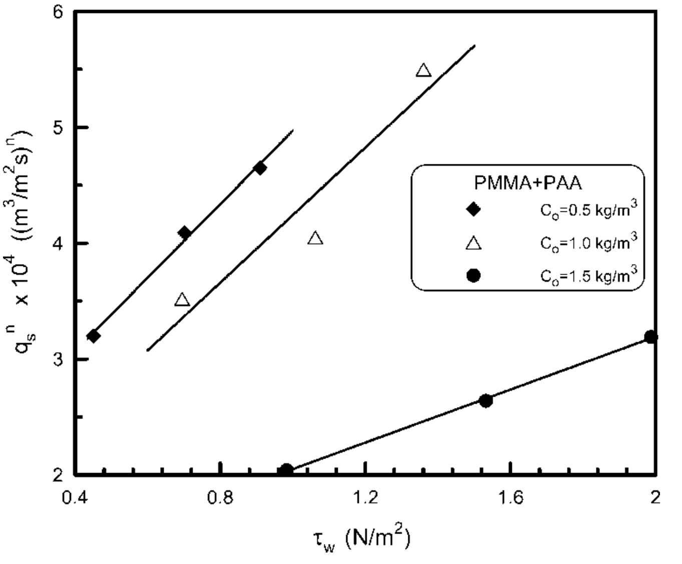

Figure 7. Effect of shear stress on the membrane surface on the filtration flux for three PAA concentrations.

The cake masses formed under various cross-flow velocities and PAA concentrations are shown in Fig. 6. The filtration pressure is fixed at 50 kPa in this figure. It can be found that the cake mass increases linearly with PAA concentration while decreases with cross-flow velocity. The relationship between cake mass and cross-flow velocity can be easily attributed to the effect of shear stress acting on the membrane surface. A higher cross-flow velocity causes the shear stress to be higher; as a result, fewer particles can be deposited stably on the membrane surface. On the other hand, since the probability of particle deposition on the membrane surface increases with the increase of fluid apparent viscosity (in the condition of higher K or lower n) (8), this effect leads more particles to deposit to form the filter cake under a higher PAA concentration. This is also one of the reasons for explaining why the filtration flux decreases with an increase in PAA concentration.

In order to demonstrate the correction of the force balance model proposed in this study, the values of q_s^n are plotted against τ_w for three different PAA concentrations in Fig. 7. The values of q_s are measured in experiments, while those of τ_w are calculated by Eq.(8). According to the

Table 2. The values of empirical coefficients in Eq.(7) (in SI system)

$C_6 \times 10^4$	$C_7 \times 10^4$	$C_8 \times 10^4$	$C_9 \times 10^4$
4.45	2.04	2.22	0.88

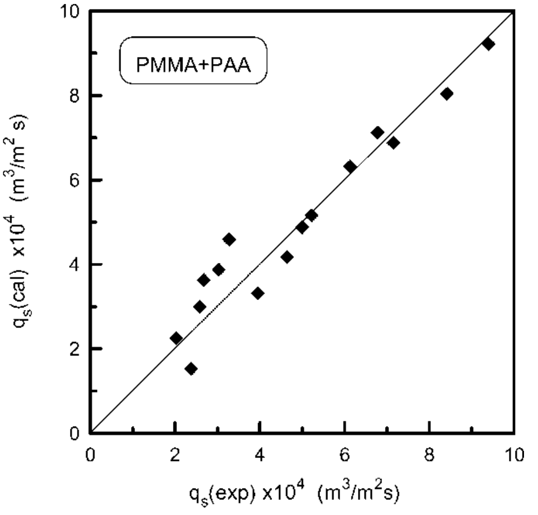

Figure 8. A comparison of filtration flux at pseudo-steady state between calculated results and experimental data.

force balance model, Eq.(7), a linear relationship between q_s^n and τ_w can be obtained for a given PAA concentration. It can be expected that the filtration flux increases linearly with shear stress acting on the membrane surface. From the slopes and intercepts of the lines shown in this figure, the coefficients in Eq.(7) can be regressed. These coefficients are listed in Table 2 and used for further calculation.

Once the empirical coefficients are obtained, the filtration flux at pseudo-steady state can be estimated by Eq.(7). Figure 8 shows a comparison of filtration flux between calculated results and experimental data under various conditions. The shear stress is calculated by Eq.(8), in which the cake thickness is calculated by Eq.(2). The diagonal line in this figure indicates the agreement between calculated results and experimental data. Since the deviations between these data are reasonable and acceptable, it can be said that the proposed model can be used to predict the pseudo-steady filtration flux.

CONCLUSION

The filtration flux and the cake properties in cross-flow microfiltration of fine particles suspended in polymeric aqueous solution have been studied. An increase in cross-flow velocity or filtration pressure caused the filtration flux to be higher; however, an increase in PAA concentration resulted in a lower filtration flux. The average specific filtration resistances of cakes remained almost constant under various cross-flow velocities and PAA concentrations; therefore, the cake mass played a major role in determining the filtration resistance and the filtration flux under a given filtration pressure. A force balance model was derived for particle deposition on the membrane surface. Once the empirical coefficients were obtained from experimental data, the filtration flux at pseudo-steady state could be estimated accurately.

NOMENCLATURE

a	particle radius (m)
C_o	polymer concentration in suspension (kg/m^3)
C_1–C_9	empirical coefficients
F_t	tangential drag force due to suspension flow (N)
F_p	normal drag force due to filtrate flow (N)
F_i	interparticle force (N)
f_c	friction coefficient (−)
H	clearness between two parallel plates of the filter (m)
K	fluid consistency index (N s^n/m^2)
L	cake thickness (m)
n	flow behavior of fluid (−)
q	filtration flux (m^3/m^2 s)
q_s	filtration flux at pseudo-steady state (m^3/m^2 s)
R_c	filtration resistance of cake (m^{-n})
R_m	filtration resistance of membrane (m^{-n})
t	filtration time (s)
u_A	the cross-flow velocity at the location of particle centre (m/s)
u_s	cross-flow velocity (m/s)
w_c	mass of dry cake per unit area (kg/m^2)

Greek Letters

γ_{av}	average specific filtration resistance of cake (m^{2-n}/kg)
ΔP	filtration pressure (N/m^2)
ε_{av}	average cake porosity (−)
τ_w	shear rate at the membrane surface (N/m^2)
ρ_s	density of particles (kg/m^3)

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REFERENCES

1. Cheryan, M. (1998) *Ultrafiltration and Microfiltration Handbook*; Technomic Publishing Co: Pennsylvania, USA, 113–130.
2. Romero, C.A. and Davis, R.H. (1990) Transient model of crossflow microfiltration. *Chem. Eng. Sci.*, 45: 13–25.
3. Belfort, G., Davis, R.H., and Zydney, A.L. (1994) The behavior of suspensions and macromolecular solutions in crossflow microfiltration. *J. Membr. Sci.*, 96: 1–58.
4. Lu, W.M. and Hwang, K.J. (1995) Cake formation in 2-D cross-flow filtration. *AIChE J.*, 41: 1443–1455.
5. Shirato, M., Aragaki, T., Iritani, E., Wakimoto, M., Fujiyoshi, S., and Nanda, S. (1977) Constant pressure filtration of power-law non-Newtonian fluids. *J. Chem. Eng. Jpn.*, 10: 54–60.
6. Kozicki, W. (1986) Recent developments in filtration of non-Newtonian fluids. *Proc. Filtration Symposium Nagoya′86*, Nagoya, Japan, 121–130.
7. Kozicki, W. (1990) Factors affecting cake resistance in non-Newtonian filtration. *Can. J. Chem. Eng.*, 68: 69–80.
8. Hwang, K.J. and Lu, W.M. (1997) Particle deposition in laminar cross-flow filtration of power law slurry. *Sep. Sci. Technol.*, 32: 1315–1334.
9. Hwang, K.J. and Lu, W.M. (1997) Hydrodynamic analysis on the mechanism of cross-flow filtration of power law slurry. *J. Chem. Eng. Jpn.*, 30: 698–705.
10. Hwang, K.J., Yu, M.C., and Lu, W.M. (1997) Migration and deposition of submicron particles in cross-flow microfiltration. *Sep. Sci. Technol.*, 32: 2723–2747.