# Effect of Particle Size on Filtration Resistance in Crossflow Microfiltration

# Toshiro MIYAHARA, Yuji YAMADA and Yasuharu AKAGI

Department of Applied Chemistry
Faculty of Engineering
Okayama University of Science
1-1, Ridai-cho, Okayama 700-0005, Japan
(Received November 1, 2000)

Key Words: Solid Liquid Separation, Crossflow Microfiltration, Filtration Resistance, Submicron Particle,
Overmicron Particle

Experiments were carried out to investigate the effect of particle size on filtration resistance in crossflow microfiltration by varying the particle size from 0.19  $\mu$  m to 17.1  $\mu$  m which corresponded to so-called submicron and overmicron particles. Filtration resistance can be calculated from Darcy's law using time course results of filtration flux under various operating conditions. From these results, three empirical correlation equations for the filtration resistance of the cake layer are obtained depending upon the range of particle sizes. Steady and unsteady filtration flux states can be estimated using these three empirical correlation equations.

### Introduction

Crossflow microfiltration is a relatively new process of growing importance which allows separation of small particles at higher permeate fluxes than conventional dead-end filtration. In the process, the fluid stream flows tangentially to the membrane surface; the shearing action of the fluid prevents the development of thick filter cakes at the membrane surface. Primary uses for this technology include clarification of fruit juices, treatment of industrial wastewater, and concentration of materials such as fermenter broths, especially a solid-liquid separation technique in the downstream processing of bio-products from microbial sources (Nakanishi and Tanaka, 1994; Tanaka et al., 1994). Recently, this technology has also been applied to the separation of oily water (Mueller et al., 1997). While it is generally accepted that the shear stress associated with tangential flow is responsible for keeping cake growth to a minimum, the shear action is not clearly defined (Blake et al., 1992; Nassehi, 1998; Chellam and Wiesner, 1998; Hwang et al., 1996; Iritani et al., 1991; Stamatakis and Tien, 1993; Wang et al., 1995). In the present study, the effect of particle size on filtration resistance in crossflow microfiltration for separation of four polymethylmethacrylate (PMMA) particles and two kinds of polystyrene (PS) particles was investigated. The filtration resistance in crossflow microfiltration

was calculated from Darcy's law using time course results of filtration flux and three empirical correlation equations for the filtration resistance of the cake layer obtained according to the range of particle sizes. Furthermore, steady and unsteady filtration flux states were predicted using three empirical correlation equations for the tested samples.

### 1. Experimental

A schematic diagram of the crossflow microfiltration system is shown in Fig. 1. It is

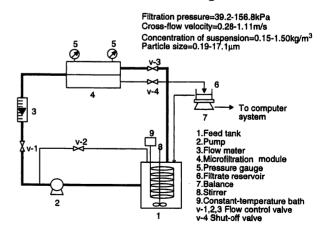


Fig. 1 Schematic diagram of experimental apparatus

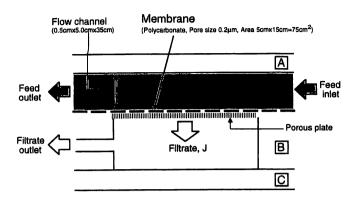


Fig. 2 Crossflow microfiltration module

comprised of a feed circulation system, a crossflow filtration module and a computer system to measure the filtrate. Figure 2 shows the assembly of the The module was constructed with three assemblies (A, B and C). The feed solution was pumped into the flow channel through the A and B assemblies. The dimensions of the flow channel were: depth 5 mm, width 5 cm and length 35 cm. A nuclepore membrane with a pore size of  $0.2 \mu m$ (Polycarbonate, Nomura Micro Science Co., Ltd.) was set on the bronze support of a porous plate. The effective membrane area for the filtrate was 75 cm<sup>2</sup>. The cross flow velocity, u, was varied from 0.28 to The filtration pressure,  $\Delta P$ , was varied in the range of 39.2 - 156.8 kPa. The concentration of the suspensions ranged from 0.15 kg/m<sup>3</sup> to 1.50 kg/m<sup>3</sup>. Four kinds of PMMA particles and two kinds of PS particles were used as shown in Table 1. The particle size in Table 1 was evaluated using a particle size instrument (model ELS-80R, Otsuka Electronics Co.) and an optical microscope.

 Table 1
 Characteristics of particles employed in this study

Particle	Volumetric mean diameter [µm]	Density [kg/m <sup>3</sup> ]	Remarks
MP-1451	0.19	1183	Polymetylmeth acrylate(PMMA)
MP-1000	0.48	1183	
MP-1400	0.86	1183	
MX-500	4.07	1183	
SGP-70C	12.0	1050	Polystyrene(PS)
SGP-100C	17.1	1050	

**Figure 3** shows particle size distribution on lognormal probability distribution plots. As shown in this figure, for all kinds of particles, the linearity suggests that the log-normal probability function

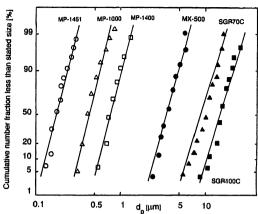


Fig. 3 Particle size distribution

provides a satisfactory representation of the measured distribution. Therefore, the particle size distribution data roughly fit a log-normal probability distribution. The number distribution function in this case is given as

$$\frac{dR}{d\left\{\ln\left(d_{p}/d_{pG}\right)\right\}} = \frac{1}{\sqrt{2\pi}\ln s_{G}} \exp\left[-\frac{\left\{\ln\left(d_{p}/d_{pG}\right)\right\}^{2}}{2\left(\ln s_{G}\right)^{2}}\right]$$
(1)

Two parameters are required to determine the mean particle size, that is,  $d_{pG}$  and  $s_G$ . They can be obtained from the data as

$$d_{pG} = \left(d_{p1} \cdot d_{p2} \cdot \cdots \cdot d_{pn}\right)^{1/n} \tag{2}$$

and

$$s_G = \exp\left[\sum \left\{\ln(d_p / d_{pG})\right\}^2 / n\right]^{1/2}$$
 (3)

Where  $d_{pG}$  is the geometric mean diameter and  $s_G$  is the geometric standard deviation. The relationship between  $d_{ji}$  and  $d_{pG}$  of a log-normal probability distribution is (Mugele and Evance, 1959)

$$d_{ji} = d_{pG} \exp\{(j+i)(\ln s_G)^2 / 2\}$$
 (4)

The volumetric mean diameter of each particle employed,  $d_{30}$ , can be calculated using Eqs. (2), (3) and (4). The results are also shown in Table 1. Distilled water was used to suspend these particles.

#### 2. Results and Discussion

### 2.1 Time course of filtration flux

Figure 4 shows the data for the time course of filtration flux under all conditions in this study. A steady state was reached when the filtration flux remained constant. As shown in this figure, the steady state for each operating condition was reached within around 4 hours. Therefore, the measuring time for filtration flux was set within 4 hours.

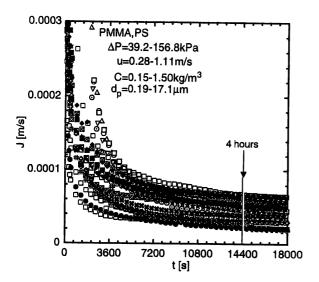


Fig. 4 Time course of filtration flux

Figure 5(a) and 5(b) show examples of the effect of cross flow velocity, u, on the filtration flux, J, for PMMA and PS particle suspensions. These figures show that there is a small effect of cross flow velocity in increasing the filtration flux, J. However, the effect of cross flow velocity for the PMMA suspension was large compared to that for the PS suspension. This indicates that the cake layer of the PS particles (large particle in size) is hard to remove by shear force induced by cross flow.

The effects of filtration pressure,  $\Delta P$ , on filtration flux are shown in **Fig. 6(a)** (for PMMA) and **6(b)** (for PS), respectively. As shown in these figures, increasing the filtration pressure enhances the filtration flux and the effect was large on the PMMA suspension. This was probably due to the same mechanism as the effect of cross flow velocity. As a matter of course, decreasing the concentration of the suspension enhanced the filtration flux.

## 2.2 Filtration resistance

Generally, the filtration flux, *J*, is given by Darcy's law as (Murkes, 1990)

$$J = \frac{\Delta P}{\mu(R_m + R_c)} \tag{5}$$

where  $\mu$  is the viscosity of liquid,  $R_m$  is the filtration resistance of the membrane and  $R_c$  is the

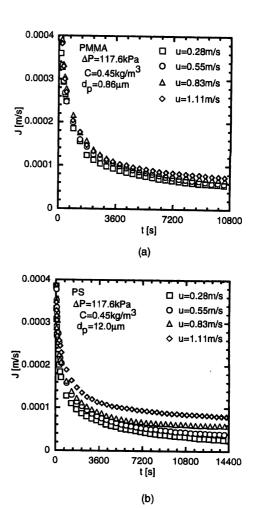


Fig. 5 Effect of crossflow velocity on filtration flux

filtration resistance of the cake layer.

Figure 7 shows the filtration resistance of a membrane in the absence of particles, that is, in the case of feeding distilled water only. From this figure, the filtration resistance of the membrane lies in order of  $10^8 \, \text{m}^{-1}$ .

The filtration resistance of the cake layer,  $R_c$ , can be calculated from Eq.(5) using the time course of filtration flux and the results for the membrane filtration resistance shown in Fig.7. Figure 8(a) and 8(b) show examples of the time course of the cake layer filtration resistance. In these figures,  $R_c$  lies in order of around  $10^{12}$  m<sup>-1</sup>. This implies that the crossflow microfiltration is controlling the filtration resistance of the cake layer.

# 2.3 Correlation of the cake layer filtration resistance

As mentioned above, as the crossflow microfiltration in this study was controlling the filtration resistance of the cake layer. Thus, we obtained the following correlation equations for  $R_c$  which are shown in **Fig.9**.

For  $d_p$ =0.19-0.86  $\mu$  m

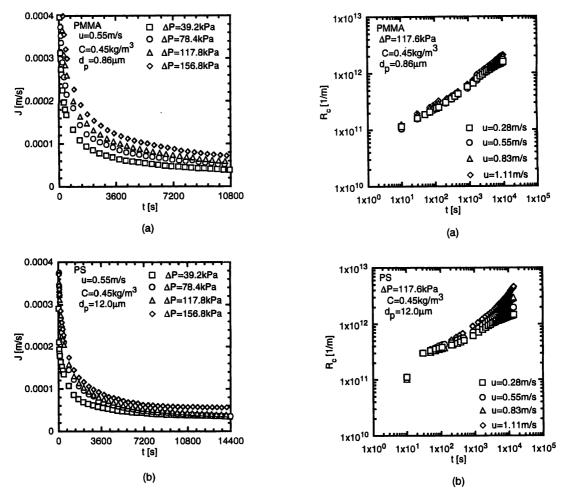


Fig. 6 Effect of filtration pressure on filtration flux

Fig. 8 Effect of crossflow velocity on the filtration resistance of the cake layer

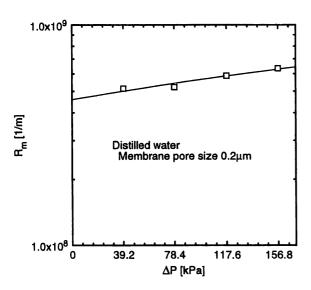


Fig. 7 Filtration resistance of the membrane

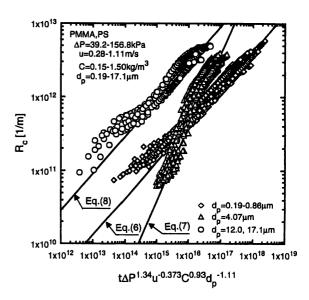


Fig. 9 Correlation of the filtration resistance of the cake layer

$$R_c = 4.14 \times 10^3 t^{0.5} \Delta P^{0.67} u^{-0.187} C^{0.465} d_p^{-0.555}$$
 (6)

for  $d_{n}$ =4.07  $\mu$  m

$$R_c = 3.83 \times 10^{-5} t \Delta P^{1.34} u^{-0.373} C^{0.93} d_p^{-1.11}$$
 (7)

and for  $d_p = 12.0-17.1 \ \mu \text{ m}$ 

$$R_c = 2.87 \times 10^4 t^{0.5} \Delta P^{0.67} u^{-0.187} C^{0.465} d_p^{-0.555}$$
 (8)

It is inferred that these three equations depend on the range of suspended particle sizes. Usually, particles less than 1  $\mu$  m show colloidal behavior and particles greater than 1  $\mu$  m have sedimentation behavior. Eq.(7) for  $d_p$ =4.07  $\mu$  m shows the probable transition between the colloidal and sedimentation behaviors.

# 2.4 Prediction of filtration flux in the unsteady state

**Figure 10** shows the comparison of measured filtration fluxes in the unsteady state with the calculated results using Eqs. (5), (6), (7) and (8). The calculated curves fit well with the measurements.

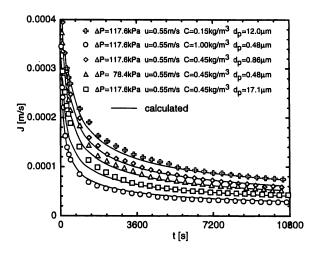


Fig. 10 Comparison of the calculated filtration flux in the unsteady state with measurements

# 2.5 Prediction of filtration flux in the steady state

As mentioned before, the steady state for each operating condition was reached within around 4 hours. **Figure 11** shows the parity plot for the filtration flux in the steady state using Eq.(5) with  $R_c$  values after 4 hours. It shows good agreement with the measurements.

### Conclusion

To clarify the influence of particle size on

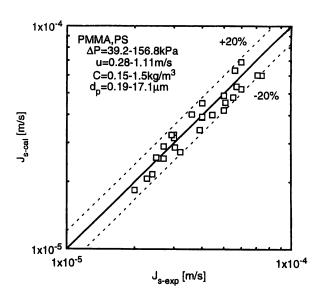


Fig. 11 Parity plot for filtration flux in the steady state

filtration resistance in crossflow microfiltration, experiments were conducted varying the particle size from 0.19 to 17.1  $\mu$  m. As a result, the following conclusions were drawn:

- 1) The steady state in filtration flux is reached within 4 hours.
- 2) Increasing the cross flow velocity and the filtration pressure, and decreasing the concentration of the suspension enhance the filtration flux.
- 3) The crossflow microfiltration in this study controlled the filtration resistance of the cake layer.
- 4) Three empirical correlation equations for the filtration resistance of the cake layer are obtained depending on the range of particle sizes.
- 5) Both steady and unsteady filtration flux states can be estimated using these empirical correlation equations.

### Acknowledgment

This work was supported by the Yakumo Foundation for Environmental Science.

#### Nomenclature

C = concentration of suspension, kg/m<sup>3</sup>  $d_p$  = particle diameter, m  $d_{ji}$  = general mean particle diameter, m  $d_{pG}$  = geometric mean particle diameter, m  $d_{30}$  = volumetric mean particle diameter, m  $d_{30}$  = filtration flux, m/s

 $J_{s-cal}$  = calculated value of filtration flux at steady state, m/s

 $J_{s-exp}$  = experimental value of filtration flux at steady state, m/s

 $\Delta P$  = filtration pressure, Pa

$R_c$	= filtration resistance of the cake layer,
	1/m

 $R_m$  = filtration resistance of the membrane, 1/m

 $s_G$  = geometric standard deviation

t = filtration time, s

u = cross flow velocity, m/s

 $\mu$  = viscosity of liquid, Pa.s

### References

- Blake, N. J., I. W. Cumming and M. Streat; "Prediction of steady state crossflow filtration using a force balance model," *J. Membrane Sci.*, **68**, 205-216 (1992)
- Chellam, S. and M. R. Wiesner; "Evaluation of crossflow filtration models based on sheared-induced diffusion and particle adhesion: Complications induced by feed suspension polydispersivity," J. Membrane Sci., 138, 83-97 (1998)
- Hwang, J., D.-J. Chang and C.-H. Chen; "Steady state permeate flux for particle cross-flow filtration," *Chem. Eng. J.*, **61**, 171-178 (1997)
- Iritani, E., T. Hayashi and T. Murase; "Analysis of filtration mechanism of crossflow upward and downward ultrafiltration," *J. Chem. Eng. Japan*, **24**, 39-44 (1991)

- Mueller, J., Y.-W. Cen and R. H. Davies; "Crossflow microfiltration of oily water," *J. Membrane Sci.*, **129**, 221-235 (1997)
- Mugele, R. A. and H. D. Evance; "Droplet size distribution in sprays," *Ind. Eng. Chem.*, **43**, 1317-1324 (1951)
- Murkes, J.; "Fundamentals of crossflow filtration," Separation and Purification Methods, 19, 1-29 (1990)
- Nakanishi, K. and T. Tanaka; "Crossflow filtration of microbial suspension," *Nippon ShokuhinKogyo Gakkaishi*, **41**, 578-584 (1994)
- Nassehi, V.; "Modeling of combined Navier-Stokes and Darcy flows in crossflow membrane filtration," *Chem. Eng. Sci.*, **53**, 1253-1265 (1998)
- Stamatakis, K. and C. Tien; "A simple model of cross-flow filtration based on particle adhesion," *AIChEJ*, **39**, 1292-1302 (1993)
- Tanaka, T., K. Abe, H. Asakawa, H. Yoshida and K. Nakanishi; "Filtration characteristics and structure of cake in crossflow filtration of bacterial suspension," J. Fermentation and Bioengineering, 78, 455-461 (1994)
- Wang, W., X. Jia and G. A. Davies; "A theoretical study of transient crossflow filtration using force balance analysis," *Chem. Eng. J.*, **60**, 55-62 (1995)