An Improved Model for Cross-Flow Microfiltration Properties of Lactic Acid Fermentation Broth

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An unsteady-state model for cross-flow microfiltration of lactic acid fermentation broth is presented in this manuscript. Compared with earlier models, the improved model can calculate specific cake resistance and membrane fouling resistance simultaneously. Filtration properties are obtained by solving the model with numerical integration and optimization. The model has been used for the analysis of experimental data and the results show good agreement with the experimental permeate flux. The model predicts smaller specific cake resistance by taking into account the membrane fouling resistance.

Introduction

In a previous study (Fitriani and Kokugan, 2010), the cake weight accumulated on the surface of a membrane (W_c^*) in cross-flow filtration was calculated by integration of the unsteady-state permeate flux from set-up to pseudo-steady state of the run under a cake filtration model. The numerical integration-trapezium method was used in the calculation. Calculation of the specific cake resistances (α) under specific operating conditions was simple, and the results were comparable with those of other researchers. However, the calculated permeate flux could not be fitted accurately to the experimental flux at the initial stage of the filtration process, possibly due to the membrane fouling resistance not being considered. To overcome this problem, we propose an improved model that is capable of simultaneously determining both membrane fouling resistance and specific cake resistance. The effects of operating conditions (transmembrane pressure, cross-flow velocity and cell concentration) on the cake resistance, membrane fouling resistance and specific cake resistance are discussed.

1. An Improved Model for Cross-flow Filtration Properties

Microfiltration is generally used at steady state under certain specific membrane and cake resistances, which are formed at the initial stage of the run through a short unsteady-state period. The unsteady state of the permeate flux in cross-flow filtration is defined as follows:

$$I_{V}(t) = \frac{\Delta P}{\mu(R_{\rm m} + R_{\rm f} + R_{\rm c}(t))}$$
(1)

where ΔP is the transmembrane pressure, $R_{\rm m}$ is the membrane resistance, $R_{\rm f}$ is the membrane fouling resistance, $R_{\rm c}$ is the cake resistance and μ is the viscosity of the permeate.

In the present study, cell debris and denatured proteins other than enzymes cause the membrane fouling for a short time at the beginning of the filtration process, and it was independent of time.

The cake filtration model (Zeman and Zydney, 1996) assumes that the rate of particle accumulation on a membrane (W_c) is proportional to the particle concentration (C) and the difference between the permeate flux (Jv) and the hydraulic lift velocity of the particles. In this case, the lift velocity is equal to the pseudo-steady state flux (Jv^*) (Shimizu *et al.*, 1993; Furukawa *et al.*, 2008).

$$\frac{1}{A}\frac{dW_{c}(t)}{dt} = (Jv(t) - Jv^{*})C$$
(2)

The specific cake resistance (α) in cross-flow filtration is defined as follows:

$$\alpha = \frac{R_{\rm c}(t)}{W_{\rm c}(t)/A} \tag{3}$$

where A is the membrane surface area.

Combining Eqs. (1) and (3) results in the following relationship:

$$J_{\mathcal{V}}(t) = \frac{\Delta P}{\mu \left(R_{\rm m} + R_{\rm f} + \alpha \frac{W_{\rm c}(t)}{A} \right)} \tag{4}$$

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Combining Eqs. (2) and (4) results in the following relationship:

$$\frac{1}{A}\frac{\mathrm{d}W_{\mathrm{c}}(t)}{\mathrm{d}t} = C \left[\frac{\Delta P}{\mu \left(R_{\mathrm{m}} + R_{\mathrm{f}} + \alpha \frac{W_{\mathrm{c}}(t)}{A} \right)} - Jv^{*} \right]$$
(5)

In this equation, the values of A, ΔP , μ , $R_{\rm m}$ and Jv^* are constant for a run. $W_{\rm c}(t)$ is numerically calculated from experimental data using the fourth order Runge– Kutta method, if the values of $R_{\rm f}$ and α are assumed. Finally, the two parameters $R_{\rm f}$ and α are simultaneously estimated by solving the following optimization problem:

$$\hat{R}_{\rm f}, \,\hat{\alpha} = \arg \min \sum_{i=1}^{N} (J v_{\rm exp} - \hat{J} v)^2 \tag{6}$$

where Jv_{exp} is the experimental data, Jv is calculated from Eqs. (4) and (5) and N is the number of samples in a run.

The model is applied to the experimental results of cross-flow filtration of *Streptococcus bovis* (Fitriani and Kokugan, 2010).

2. Results and Discussion

2.1 Permeate flux and cake weight

Figure 1 shows the permeate flux and cake weight under the following conditions: transmembrane pressure (ΔP) of 200 kPa, cross-flow velocity (u) of 0.45 m/s and cell concentration (C) of 7.5 g/L until filtration time (t) of 60 min. It is evident that the permeate flux line calculated by the present model shows good agreement with the experimental results. On the other hand, the previous model cannot exactly predict the temporal decline in the permeate flux for a period less than 15 min. The discrepancy between the experimental and calculated results at the initial stage of the run in the previous model has also been observed in other experimental runs. The difference between the model proposed here and the previous model is the introduction of the membrane fouling resistance $(R_{\rm f})$. Without membrane fouling being considered, the calculated initial flux is higher due to lower values of resistance and the model behaves similarly as the previous model.

Furukawa *et al.* (2000) studied the cross-flow filtration of soy-sauce lees in a flat-type membrane module. They reported a model to predict the permeate flux decline and determine specific cake resistance using an analytical solution of the cake filtration model (Zeman and Zydney, 1996). They found that the model could not accurately predict the decline in permeate flux, especially at the initial stages of filtration, possibly due to plugging or constriction occurring inside the pore, which led to an increase in the membrane resistance. They modified the analytical model of cake filtration in a later study



Fig. 1 The comparison between permeate flux calculated by improved model and the previous model at $\Delta P =$ 200 kPa, u = 0.45 m/s and C = 7.5 g/L

(Furukawa *et al.*, 2008), in which they determined the initial flux (J_o) using the membrane resistance value for the plugged membrane resistance observed during the early period in the cross-flow filtration instead of the clean membrane hydraulic resistance value determined using pure water. They obtained a more accurate prediction of permeate flux through the modified analysis. However, their method to determine the initial flux (J_o) was unclear.

It is unnecessary to determine the period of fouling in the model proposed here, where both membrane fouling resistance (R_f) and specific cake resistance (α) can be calculated simultaneously. We observed that the introduction of R_f facilitated the best fit of the permeate flux curve with the experimental data at the initial period. Therefore, the proposed model indicates that membrane fouling occurs at the initial period.

Huang and Morrissey (1998) studied the membrane fouling that occurs during microfiltration of surimi wash water in a plate and frame membrane module. They found that the pore blocking resistance of membrane fouling was the dominant resistance during the initial filtration period, and the cake resistance began to dominate after the initial pore blocking.

The model proposed in the present study also allows the dynamic observation of cake weight (W_c) during cross-flow filtration. Accumulation of the cake on the membrane surface occurs simultaneously with membrane fouling. **Figure 2** shows the behaviors of Jv and W_c over a larger range of filtration time (t), taking into account the transmembrane pressure (ΔP) as a parameter. Figure 1 is identical to Figure 2(d) ($\Delta P = 200$ kPa), but it is clearer at the initial stage of filtration. From Figure 2, we see that the considerations used for $\Delta P =$ 200 kPa in Figure 1 are valid for $\Delta P = 20$ kPa, $\Delta P =$ 100 kPa and $\Delta P = 150$ kPa, and W_c increases with increasing transmembrane pressure.

Figure 3 shows the effect of cross-flow velocity (a)





Fig. 3 The effect of (a) cross-flow velocity and (b) cell concentration on permeate flux and cake weight at $\Delta P = 100$ kPa

and cell concentration (b) on permeate flux and cake weight at $\Delta P = 100$ kPa. The result shows a decrease in cake weight with an increase in the cross-flow velocity (Figure 3(a)). This is due to the higher shear stress at high cross-flow velocity, which diminishes the formation of the cake layer on the membrane surface. The cake weight is also observed to increase with increasing cell concentration (Figure 3(b)). Our results are in agreement with the results reported by Tanaka *et al.* (1994).

2.2 Cake resistance and membrane fouling resistance

Figure 4 shows the effect of operating conditions $(\Delta P, u \text{ and } C)$ on the cake resistance $(R_c^*_{app}, R_c^*_{true})$, membrane fouling resistance (R_f) and cake weight on the membrane surface at pseudo steady state (W_c^*) . $R_c^*_{true}$ and $R_c^*_{app}$ are the cake resistances calculated by the proposed model and the previous model at the pseudo-steady state, respectively. R_m is the membrane resistance calculated using pure water. The figure shows



Fig. 4 The effect of operating conditions on resistance: (a) transmembrane pressure, (b) cross-flow velocity, and (c) cell concentration

that both $R_c^*_{true}$ and $R_c^*_{app}$ increase with increasing transmembrane pressure (ΔP) and cell concentration (C) and decrease with increasing cross-flow velocity (u). The figure also shows that $R_c^*_{true}$ is lower than $R_c^*_{app}$. This is because the current model considers the membrane fouling resistance in cake filtration. $R_c^*_{true}$ is the result of the reduction of R_f from $R_c^*_{app}$.

Figure 4 also shows that the membrane fouling resistance (R_f) increases with increasing transmembrane pressure (ΔP) . However, it is not affected by cross-flow velocity and cell concentration above 4.5 g/L.

 $W_{\rm c}*_{\rm true}$, which is the value at $t = t^*$ (pseudo-steady state), agrees with $W_{\rm c}*_{\rm app}$. Although $W_{\rm c}*$ and $R_{\rm c}*$ increase with increasing ΔP , the increment in $W_{\rm c}*$ is smaller than in $R_{\rm c}*$ (refer to Figure 4(a)). Therefore, in

Fig. 5 Comparison of α_{app} and α_{true} for various operating conditions: (a) transmembrane pressure, (b) cross-flow velocity, and (c) cell concentration

accordance with Eq. (3), the specific cake resistance (α) is not constant for ΔP .

2.3 Specific cake resistance

The specific cake resistance was determined using the proposed model (α_{true}) and the previous model (α_{app}), and the results of comparison are shown in **Figure 5**. The figure shows the effects of three variables on the specific cake resistance with the operating conditions considered as a parameter: (a) transmembrane pressure (ΔP), (b) cross-flow velocity (*u*) and (c) cell concentration (*C*). α_{true} was found to be lower than α_{app} for all operating conditions. This is recognized from Eq. (3) that $R_c^*_{true}$ is smaller than $R_c^*_{app}$ whereas $W_c^*_{true}$ is equal to $W_c^*_{app}$. Furthermore, α_{true} and α_{app} show similar tendencies under all operating conditions. Both α_{true} and α_{app}

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increase with increasing transmembrane pressure and cross-flow velocity, decrease with increasing cell concentration up to 4.5 g/L and then change minimally at concentration above 4.5 g/L. The value of cake compressibility (*n*) from Figure 5(a) is 0.92 compared to 1.0 calculated in a previous study.

The possible mechanism of the increase of the specific cake resistance of the filter cake in the increase of cross-flow velocity (Figure 5(b)) can be considered that the chain-like cells of *Streptococcus bovis* tended to be better arranged parallel to the circulation flow at the higher cross-flow velocity which results in the lower cake porosity and higher specific cake resistance. The increase of the specific cake resistance in the decrease of cell concentration (Figure 5(c)) because the mobility of cells is high at the low cell concentration which results in a lower cake porosity comparing to that at the high cell concentration (Ho and Sirkar, 1992).

McCarthy *et al.* (2002) studied the separation characteristics of *Kluyveromyces marxianus* using a tubular ceramic membrane by quantifying the cake weight and specific cake resistance experimentally. They also found that α_{true} was lower than α_{app} due to the significance of membrane fouling in the system.

Conclusions

An improved model to determine cross-flow filtration properties of S. bovis in the lactic acid fermentation broth has been investigated in this manuscript. The proposed model allows simultaneous determination of specific cake resistance (α) and membrane fouling resistance (R_f) through numerical integration and optimization of the unsteady-state cake filtration model. We found that introducing the membrane fouling resistance in the cake filtration model provided the best match between the calculated permeate flux and the experimental data for the observed period, especially at the initial permeate flux. The cake resistance calculated by the proposed model was found to be lower than the calculation of the previous model. The cake compressibility (n) was calculated as 0.92 compared to 1.0 in a previous study. Moreover, the membrane fouling resistance was found to increase with increasing transmembrane pressure but was unaffected by increases in cross-flow velocity and cell concentration. The specific cake resistance obtained using the proposed model was lower than the resistance predicted by the previous model, although it showed similar behavior at different operating conditions (transmembrane pressure, cross-flow velocity and cell concentration). Compared with the previous model, more accurate microfiltration properties can be obtained by including membrane fouling resistance and simultaneously es-

timation of $R_{\rm f}$ and α .

Nomenclature

4	=	membrane surface area	[m ²]
С	=	concentration of cell	[g/L]
Iv	=	permeate flux [L/(mi	$n \cdot m^2)$]
Jv*	=	permeate flux at pseudo-steady state [L/(mi	$n \cdot m^2)$]
n	=	cake compressibility	[—]
R	=	resistance	$[m^{-1}]$
R _c	=	cake resistance	$[m^{-1}]$
R [*] _{app}	=	pseudo-steady-state cake resistance in the p	revious
		model	$[m^{-1}]$
$R_{c}^{*}_{true}$	=	pseudo-steady-state cake resistance in the pr	roposed
		model	$[m^{-1}]$
R _f	=	membrane fouling resistance	$[m^{-1}]$
R _m	=	membrane resistance	$[m^{-1}]$
•	=	filtration time	[min]
*	=	pseudo-steady state filtration time	[min]
u	=	cross-flow velocity	[m/s]
W _c	=	cake weight	[g]
W [*] _c app	=	cake weight in the previous model	[g]
W _c * _{true}	=	cake weight in the proposed model	[g]
α	=	specific cake resistance	[m/kg]
α_{app}	-	specific cake resistance in the previous model	[m/kg]
α _{true}	=	specific cake resistance in the proposed model	[m/kg]
ΔP	=	transmembr <mark>ane pressure</mark>	[kPa]
и	=	viscosity of the permeate	$[Pa \cdot s]$
		a second a second as a second	

<Subscripts>

alc	=	calculation
хр	=	experiment

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