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Modeling of permeation process in cross-flow ultrafiltration

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Abstract
In cross-flow ultrafiltration (UF), it is considered that the surface part of fouling layer is removed by effect of shear stress exerted by that the feed liquid flow. As a result, decline of permeate flux is suppressed. However, the relationship between the feed flow and the permeate process have never been clarified. In this study, separation of protein from whey protein concentrate (WPC) suspension by cross-flow UF in dairy industry is taken up as a subject. The compressive yield stress model for the permeation through fouling layer on membrane was applied for the cross-flow UF taking the removal rate of fouling layer into account. The concentration distribution in the fouling layer should be estimated by the model. The concentration at the top surface of fouling layer was determined based on the relationship between yield stress varied by the concentration of WPC suspension and shear stress on the surface of the fouling layer. The validity of the model was discussed by comparing the results of estimation by the model and the experiments. As the result, it appeared that the permeate flux in steady state was approximately estimated by mean of the model.

Keywords: Ultrafiltration, Whey protein concentrate, Cross-flow, Compressive yield stress, Rheology

1. Introduction
UF process has been utilized widely in dairy industry. It is often used in many processes such as separation and concentration of protein from whey suspension. In such process, cake forms fouling layer on the surface of membrane and shows compressibility (Iritani et al., 1993). For compressible fouling layer, an increase of filtration pressure or flow rate toward the membrane results in more concentrated cake, and then the resistance against permeate flow becomes higher. Therefore, a model taking compressibility of cake into account is desirable in order to predict the filtration process. The compressive yield stress model was proposed by Buscall and White (1987) and developed by Landman et al. (1995). In the model, they considered permeate flux and concentration distribution of the inside of fouling layer based on pressure distribution which appeared from pressure loss when the permeate fluid flowed in the cake. The theory was subsequently applied to optimize pressure filtration in the case of no membrane resistance (Landman and White, 1997). In those studies, the theory was extended to UF of proteins where there is significant membrane resistance and fouling layer is compressible.

In this study, operation to separate protein from whey suspension by UF in the manufacturing process of cheese is taken up as a subject. In the dead-end UF of WPC suspension, which was used as a model fluid of whey suspension, it was enable to predict the permeate process by the compressive yield stress model (Landman et al., 1995). On the other hand, the theories of cross-flow UF based on the model was studied (Landman et al., 2004 and Lawrence et al., 2008). However, the relationship between change in permeate flux with time and the operational conditions has never been made clear because the behavior of the filtration process of the filtration process of cross-flow UF is quite complicated. The purpose of this study is that the theory of cross-flow UF of WPC suspension based on the compressive yield stress model is developed by predicting the behavior of the fouling layer based on rheology of WPC suspension.

2. A model on a permeate process
2.1 Compressive yield stress
When particle suspension is concentrated over gel point fraction, ϕ_{gel}, particles in the suspension forms connected aggregate structures. Once the network forms, the suspension has solid normal stress resisting against compression. This stress is called the compressive yield stress, P_y, and it is represented by the following equation:

$$P_y(\phi) = p\left[\left(\frac{\phi}{\phi_{gel}}\right)^q - 1\right] \qquad (1)$$

where ϕ is the volume fraction of suspension, p and q are the constant values determined by an intended particle. In WPC suspension, it was reported that $p = 220.39$ and $q = 4.0689$ (Lawrence, 1998), and $\phi_{gel} = 0.142$ (Buscall and White, 1987). P_y appears when ϕ is over ϕ_{gel}. P_y is 0 when ϕ is less than ϕ_{gel}.

2.2 Compressive yield stress model
In dead-end filtration, the volume fraction at the surface of membrane varies from ϕ_{gel} to ϕ_∞ with time. P_y is equal to

trans-membrane pressure (TMP), ΔP, under the condition that $\phi=\phi_\infty$. The period from ϕ_{gel} to ϕ_∞ is divided into 100 equal steps. The pressure and volume fraction distributions in the fouling layer in the normal direction to the membrane and permeate flux at each step are calculated using the values of volume fraction on the surface of the membrane and Eq.(1). The following mass balance is derived by the relationship between volumes of the fouling layer of i-th and $(i+1)$-th step.

$$\int_0^{l_i} \phi dz + \frac{\phi_0}{1-\phi_0} \Delta V_i = \int_0^{l_{i+1}} \phi dz \qquad (2)$$

where the l_i is the height of the fouling layer at i-th step and ΔV_i is permeate liquid volume per unit membrane area during i-th step. In Eq.(2), the first term of left side represents the volume of the fouling layer at i-th step and the right side is the volume at $(i+1)$-th step. The difference of the volume between each step is represented by the volume of particles transferred to the fouling layer with permeate flow, as shown in the second term of the left side. Filtration time, t, and permeate volume, V, are estimated by using this equation.

2.3 Application for cross-flow UF
In cross-flow UF, the equation of material balance in the fouling layer is represented by the following equation:

$$\int_0^{l_i} \phi dz + \frac{\phi_0}{1-\phi_0} \Delta V_i - v\Delta t_i = \int_0^{l_{i+1}} \phi dz \qquad (3)$$

where the third term of the left side is the volume removed from the fouling layer by the effect of shear stress exerted by the feed flow and v is the removal rate of the fouling layer per unit time and unit membrane area.

When the process of cross-flow UF is estimated based on the model, it is necessary to determine the volume fraction at the top of the fouling layer, ϕ_t. In dead-end UF, ϕ_t is equal to ϕ_{gel}. In cross-flow UF, however, ϕ_t is not equal to ϕ_{gel} because the upper part of the fouling layer is removed by the effect of shear stress.
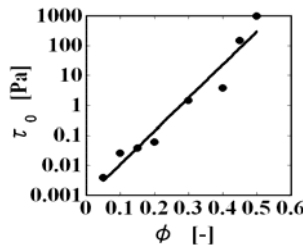

Figure 1 Relationship between yield stress and volume fraction

Rheology measurement of concentrated suspension which was regarded as the fouling layer was conducted to expect the behavior of the fouling layer and determine ϕ_t. As a result, it was clarified that concentrated suspension of WPC had yield stress and behaved as Bingham fluid. The relationship between yield stress and volume fraction is shown in Figure 1.

It appeared that the range of the yield stress in Fig. 1 corresponded to that of shear stress calculated from feed flow rate. Therefore, ϕ_t is determined as volume fraction where yield stress becomes equal to shear stress.

3. Simulation of permeate process
The results estimated by using the model stated in previous section were tried to be fitted to experimental data. Fitting parameter is removal rate, v, and the fitting was made so that the estimation result agree with experimental data in steady state. The result is shown in Fig. 2.
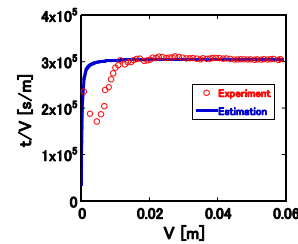

Figure 2 Fitting result (TMP = 200kPa, Q = 0.5L/min)

The estimation result was fitted well in steady state. In the initial period, however, the result was not fitted well. It is considered that the condition of the fouling layer is lack of stability or the behavior of developing the fouling layer in the initial period was not expected enough.

4. Conclusion
In this study, the compressive yield stress model in dead-end UF was applied for cross-flow UF taking the removal rate of the fouling layer into account. The volume fraction at top of the fouling layer was determined by rheology data of concentrated suspension. The estimation results could predict the permeate process in steady state.

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