Removal of a Model Protein Foulant from Metal Surfaces

Hong Xin, Xiao Dong Chen, and Necati Özkan
Dept. of Chemical and Materials Engineering, The University of Auckland, Auckland, New Zealand

DOI 10.1002/aic.10149

Using precoated whey protein concentrate gel films as a model milk protein fouling material, the removal kinetics from the inner surface of a stainless steel tube were determined continuously and noninvasively by a rapid UV spectrophotometric method. The influences of temperature, flow velocity, and the amount of fouling on the cleaning behavior were evaluated. For the first time, a cleaning model combining polymer dissolution and boundary layer mass transfer has been used to describe the cleaning process. It is suggested that the disengagement of protein molecules from the swollen gel–solution interface and the transfer of these disengaged protein molecules into the bulk cleaning solution are the rate-limiting steps in the range of conditions examined. The experimental results agree with the predictions of the cleaning model. The application of the current cleaning model to milk protein and whey protein fouling systems reported by various researchers is also discussed. © 2004 American Institute of Chemical Engineers *AIChE J,* 50: 1961–1973, 2004

Keywords: cleaning model, milk fouling, whey protein concentrate (WPC) gel, UV spectrophotometer

Introduction

Milk fouling on heat transfer surfaces is a serious problem in dairy processing plants, where frequent cleaning is required to meet the strict hygiene standards and to maintain normal production capability. Because of the high heat transfer resistance and difficulty of removal, proteinaceous fouling is a major concern for cleaning processes. In dairy processing plants, sodium hydroxide (NaOH)–based alkaline solutions are usually used to clean the proteinaceous fouling. Although the removal of milk protein fouling from stainless steel surfaces has been studied extensively (Fryer and Bird, 1994; Jeurnink and Brinkman, 1994; Lalande and Rene, 1988), the mechanisms of cleaning are still not fully understood. The understanding of the cleaning mechanisms and the development of a mathematical model are required to optimize the cleaning processes.

Cleaning is a multistage process constituting various steps that may be controlled by mechanical action, chemical reactions, and mass transfer. The cleaning process may be described as a three-stage process based on the cleaning rate periods: a swelling stage, a uniform stage, and a decay stage (Gillham et al., 1998). Studies have shown that the dissolution of deposits in the cleaning solution is an important factor controlling cleaning processes, especially in the uniform or plateau stage (Bird and Fryer, 1991; Gillham et al., 1999; Xin et al., 2002a,b). A chemical dissolution–based mass-transfer controlled cleaning mechanism has already been suggested by several researchers. Schlussler (1970) developed an empirical model based on the transportation of reaction products through a boundary layer to describe the removal of porous deposits from a heat transfer surface. In the study of the cleaning procedure of milk fouling, Gallot-Lavallée and Lalande (1985) found that the measured cleaning rate was proportional to the concentration gradient of the intermediate reaction products and the mass-transfer coefficient of the reaction products at the interface.

In a recent study, Xin et al. (2002 a,b) illustrated the simi-

---

Correspondence concerning this article should be addressed to X. D. Chen at d.chen@auckland.ac.nz.

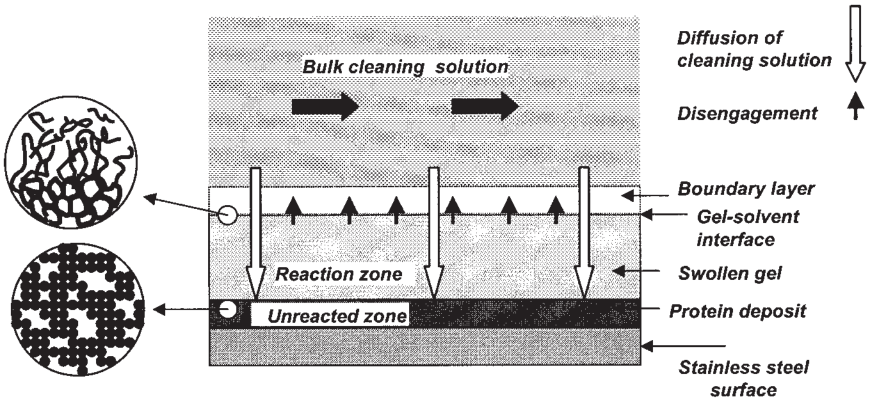

Figure 1. WPC gel film dissolution process.

larity of both the dissolution and mechanical properties between whey protein concentrate (WPC) gel and WPC fouling deposits. In this study, using a WPC gel as a model fouling material, the removal kinetics of precoated WPC gel films from the inner surface of a stainless steel tube were determined. A mathematical cleaning model, based on the polymer dissolution theory and the mass-transfer theory, was developed and compared with the experimental data. A discussion on the effects of experimental conditions on the validity of the cleaning model is also reported.

Mechanisms and Mathematical Models

Polymer dissolution–based cleaning mechanisms

The basic structure of milk protein deposits consists of aggregated milk protein molecules and voids. Milk protein molecules have long molecular chains, which have been defined as polymers of amino acids (Biddle, 1986). The swelling of the protein deposit and the final removal of this swollen layer are analogous to the dissolution process of polymers when they are treated with suitable solvents (Xin et al., 2002a). Therefore, the polymer dissolution concept can be applied to reveal the cleaning mechanism of the protein deposits.

In general, polymer dissolution behavior differs from that of a nonpolymeric material in two aspects. Polymers require an induction time before starting to dissolve, whereas nonpolymeric materials dissolve instantaneously. Polymer dissolution involves both the disentanglement of the polymer chains and the diffusion of the chains through the boundary layer adjacent to the solvent–polymer interface. The dissolution of the nonpolymeric material is generally controlled by the mass-transfer step (Narasimhan, 2001). The important parameters affecting the polymer dissolution process include the diffusion coefficient of solvent, the molecular weight (or the molecular size) of the polymer, the structure and the shape of the polymer chains, the thermodynamic compatibility of the solvent, agitation, and temperature (Devotta et al., 1993).

By adoption of the polymer dissolution concept, the essential physical features of the cleaning of milk protein deposit are depicted in Figure 1. First, the cleaning agent is transported from bulk solution to the surface of the deposit through a fluid boundary layer. Contact of the cleaning solution with the deposit will trigger a series of reactions, generating some intermediate reaction products (certain modified protein molecules and peptide chains). Further penetration of the cleaning solution into the deposit will build up a “reaction zone” and form a swollen gel layer. A disengagement process is needed before the intermediate reaction products can be transferred across the boundary layer into the bulk cleaning solution.

The disengagement process of the protein molecules from the gel–solution interface is complicated. In the swollen region, the cleaning agent concentration is high and the protein molecules have a high mobility, and the movement of the protein molecules essentially starts from this region. After a short reptation time, the protein molecular chains on the gel side of the gel–solution interface tend to disengage from the interface and move into the solution. The long and mutually entangled protein chains are inhibited from entering the liquid phase as a result of the friction between themselves. The disengagement rate of protein molecules is one of the factors controlling the dissolution process.

In addition to the disengagement, a mass-transfer resistance also exists at the gel–solution interface. If the disengagement rate is relatively small, the mass-transfer resistance through the external boundary layer may be ignored. However, if the magnitude of disengagement rate and mass-transfer rate are similar, then the dissolution process can be both disengagement and diffusion limited (Ranade and Mashelkar, 1995). With increasing cleaning time, the disengaged polymer chains will begin to accumulate on the gel–solution interface until a maximum volume fraction is reached. Then, the rate of disengagement from the interface would be constrained by the rate of mass transfer to the bulk solution. The concentration gradient between the interface of the swollen gel and the bulk cleaning solution provides a driving force for the movement of the disengaged protein molecules. The dissolution rate then reaches its highest value.

Mathematical model of cleaning

The dissolution of solid polymers in solvents is very important in microlithography photoresist development, controlled release systems, and polymerization processes. The study of the mechanism of polymer dissolution process has been a very active research area, and some mathematical models, involving some complicated moving boundary layer problems, have been published recently (Devotta and Mashelkar, 1996; Parker et al., 2000; Peppas and Wu, 1994; Ranade and Mashelkar, 1995).

Although these models have given the points for the development of a cleaning model, they are generally complex and need a numerical integration method to calculate the results.

In this study, we have attempted to capture the key mechanisms by using a simple mathematical model. In fouling and cleaning studies, it is the first time that a polymer dissolution concept was used to describe the complicated protein foulant cleaning process and predict the cleaning results. The approach used herein to model the dissolution process is different from approaches used in the polymer community, such as in microlithography photoresist development or the controlled release systems. First, the complicated moving boundary layer problem is not considered in this model. It makes this model straightforward and does not need a numerical integration method to predict the results, which is common in previous polymer dissolution models. Second, this cleaning model includes two stages, the swelling–uniform stage and the decay stage, which is different from other dissolution models. Finally, the most important parameter used in current model, R_m, has not been identified in previous polymer dissolution models.

For the cleaning process, we assumed that the disengagement of protein molecules from the swollen gel–solution interface and the transfer of these disengaged protein molecules into the bulk cleaning solution are the rate-limiting steps. Diffusion of cleaning solution and the chemical reactions take place very rapidly. During the swelling and uniform stage, the cleaning rate may be calculated in terms of the mass-transfer coefficient and the concentration gradient of the disengaged protein molecules in the boundary layer. In the decay stage, an effective surface area may be used to correlate the cleaning rate. For simplicity, the accumulation of the disengaged protein molecular chains in the boundary layer is neglected. The moving boundary layer problem and the difference protein volume fractions within the swollen gel are not considered.

Swelling and Uniform Stage. Based on the conventional concept of mass transfer, the cleaning rate (or mass flux) of the WPC gel molecules from the gel–solution interface may be written as

$$-R = \frac{1}{A}\frac{dm}{dt} = k_\phi(\phi - \phi_b) \tag{1}$$

where m is the mass removed, R is the cleaning rate (taken as the absolute value), k_ϕ is the mass-transfer coefficient based on volume fraction changes of materials, A is the surface area, ϕ is the volume fraction of the disengaged protein molecules at gel–solution interface, and ϕ_b is the volume fraction of the disengaged protein molecules in the bulk cleaning solution. Because the cleaning solution is not recirculated in this study, the concentration of the disengaged protein molecules outside the boundary layer ϕ_b is negligible and can be taken as zero, and thus Eq. 1 becomes

$$-R = \frac{1}{A}\frac{dm}{dt} = -k_\phi \phi \tag{2}$$

The change of the volume fraction of disengaged protein molecular chains accumulated at the gel–solution interface at any time has been assumed to change according to a first-order reaction mechanism

$$\frac{d\phi}{dt} = k_d \phi \tag{3}$$

where k_d is the disengagement rate constant. The physics of the disengagement process has been elaborated by Devotta et al (1993). The disengagement is recognized to be directly related to the mobility of polymer molecules, whereas the mobility of the polymer molecules will depend on their volume fraction. It has been assumed that the variation of the mobility of polymer chains is a product of a kinetic constant and the extent of the departure from the maximum mobility (Devotta et al., 1995). Based on this assumption, in the current study, it is assumed that the disengagement rate constant of the molecule chains (k_d) decreases with increasing ϕ, and approaches zero when the maximum value (ϕ_m) is reached. As a first estimation, k_d takes the following form

$$k_d = \xi\left(1 - \frac{\phi}{\phi_m}\right) \tag{4}$$

where ξ is a kinetic constant and ϕ_m is the maximum volume fraction taken up by the disengaged protein molecules. The change of the volume fraction of the disengaged protein molecules at the gel–solution interface is then expressed by the following equation

$$\frac{d\phi}{dt} = \xi\left(1 - \frac{\phi}{\phi_m}\right)\phi \tag{5}$$

Given that a polymer chain requires a finite induction time to disengage from the gel–solution interface, the disengagement rate is initially zero. This minimum induction time required for the "first few" chains to disengage is equivalent to the reptation time (t_r). Thus, it is assumed that the following initial condition exists at the gel–solution interface

$$\frac{d\phi}{dt} = 0 \qquad t < t_r \tag{6}$$

After the reptation process, from $t = t_r$ to $t > t_r$, the volume fraction of the disengaged protein molecules at the gel–solution interface can be calculated by integrating Eq. 5

$$\phi = \frac{\phi_m e^{\xi(t-t_r)}}{\dfrac{\phi_m}{\phi_0} - 1 + e^{\xi(t-t_r)}} \tag{7}$$

where ϕ_0 is the volume fraction of the tangling protein chains at the solution side of the gel–solution interface at the time $t = t_r$ ($\phi_0 \neq 0$). The above equations represent a self-limiting process when the concentration of the protein molecules become high (Murray, 2002).

We now define a dimensionless parameter ψ as

$$\psi = \frac{\phi_m}{\phi_0} - 1 \tag{8}$$

Combining Eqs. 2 and 7, the cleaning rate can be rewritten as follows

$$-R = \frac{1}{A}\frac{dm}{dt} = -\frac{R_m e^{\xi(t-t_r)}}{\psi + e^{\xi(t-t_r)}} \tag{9}$$

where R_m (the constant cleaning rate during the uniform cleaning stage) is defined as

$$-R_m = -k_\phi \phi_m \tag{10}$$

The amount of mass removed (under a certain constant cleaning condition with a known temperature, velocity, and concentration of cleaning solution, etc.) from the deposit as a function of time can then be calculated by integrating Eq. 9 from time $t \geq t_r$ to $t \leq t_{su}$ (t_{su} is the total cleaning time during the swelling and uniform stages)

$$m = \frac{AR_m}{\xi}\left(\ln\frac{(\psi + e^{\xi(t-t_r)})}{(\psi + 1)}\right) \tag{11}$$

Decay Stage. At the end of the uniform stage, the continuous film of WPC deposit is broken up and only the patches of the deposit film are left on the stainless steel surface. When the experiments were stopped during the decay stage, such residual patches can be observed. Because the film is no longer continuous and uniform, it is expected that the cleaning rate will depend on the amount of the gel film remaining on the surface, whereas the cleaning rate will still be controlled by dissolution and mass-transfer processes. In the study of the removal of organic films in the decay stage, the change of the surface area of the remaining film has been modeled as a first-order process (Beaudoin et al., 1995), and adopting this approach, the protein gel removal in the decay stage is given as

$$\frac{dA_L}{dt} = -k_A A_L \tag{12}$$

where A_L is the surface area covered by the protein film in the decay stage and k_A is the first-order rate constant for the surface area reduction. This rate constant is expected to be dependent on temperature, mechanical properties of deposit, cleaning solution concentration, and flow velocity. The initial condition for Eq. 12 is

$$A_L = A_{L,0} \qquad \text{when } t \leq t_{su}$$

where $A_{L,0}$ is the total surface area covered by the protein film. Integrating Eq. 12 from $t \geq t_{su}$ to $t \leq t_t$ (the total cleaning time) yields

$$\frac{A_L}{A_{L,0}} = e^{[-k_A(t-t_{su})]} \tag{13}$$

Assuming that the cleaning rate during the decay stage depends on the remaining protein film area A_L, the cleaning rate during this stage can be expressed as

$$R = R_m \frac{A_L}{A_{L,0}} \tag{14}$$

Combining Eqs. 13 and 14 gives

$$R = R_m e^{[-k_A(t-t_{su})]} \tag{15}$$

To calculate t_{su} and the total cleaning time t_t, a critical protein mass remaining (m_c) at the start of the decay stage is defined as

$$m_c = m_0 - m_{su} \tag{16}$$

where m_0 is the original mass of the deposit and m_{su} is the total mass removed during the swelling and uniform stages. m_{su} can be calculated from Eq. 11 with the boundary condition at $t = t_{su}$.

Combining Eqs. 11 and 16, the mass removed during the decay stage can be determined as

$$m_c = m_0 - \frac{AR_m}{\xi}\ln\frac{[\psi + e^{\xi(t_{su}-t_r)}]}{(\psi + 1)} \tag{17}$$

Rearranging Eq. 17, t_{su} is given by the following equation

$$t_{su} = \frac{1}{\xi}\ln[(\psi + 1)e^{(m_0-m_c)\xi/R_mA} - \psi] + t_r \tag{18}$$

The mass loss of the deposit during the decay stage can also be expressed as

$$\frac{1}{A_L}\frac{dm}{dt} = \frac{dm}{Ae^{[-k_A(t-t_{su})]}dt} = -R_m \tag{19}$$

Integration with the boundary conditions

$$m = 0 \qquad \text{and} \qquad A_L = A_{L,0} \qquad \text{when } t = t_{su} \tag{19a}$$

$$m = m_c \qquad \text{and} \qquad A_L = 0 \qquad \text{when } t = t_t \tag{19b}$$

Then m_c can be expressed as

$$m_c = \frac{R_m A}{k_A}\{1 - e^{[-k_A(t_t-t_{su})]}\} \tag{20}$$

By rearranging the above equation, t_t is given by the following equation:

$$t_t = -\frac{1}{k_A}\ln\left(1 - \frac{m_c k_A}{R_m A}\right) + t_{su} \tag{21}$$

By combining Eqs. 18 and 21, then the total cleaning time t_t can be determined as

$$t_t = \frac{1}{\xi}\ln[(\psi + 1)e^{(m_0 - m_c)\xi/R_mA} - \psi] - \frac{1}{k_A}\ln\left(1 - \frac{m_ck_A}{R_mA}\right) + t_r \tag{22}$$

Experimental Procedures

The cleaning system designed to determine the cleaning kinetics of WPC gel deposits from a stainless steel tube is illustrated schematically in Figure 2. The stainless steel tube (AISI 316, ID = 16 mm and length = 150 mm) was precoated with a WPC gel film using a rotation device (Xin et al., 2002b). A cleaning solution reservoir containing heating coils and an agitator was used to prepare the alkaline cleaning solutions with a given concentration and temperature. The flow velocity of the cleaning solution is controlled by a rotary flow meter. In the once-through cleaning loop, the cleaning solution, containing the removed deposits, was continuously transported to the UV spectrophotometer (HP 8312 model) by a sample pump. The UV absorption data (248 and 256 nm) were recorded at 10-s intervals to monitor the whole cleaning process. The protein concentration in the cleaning solutions is estimated by the following equation (Xin et al., 2002b)

$$\text{WPC concentration (g/L)} = 1.0368(A_{248} - A_{256}) + 0.0005 \tag{23}$$

where A_{248} and A_{256} are the values of UV absorbance at the wavelengths of 248 and 256 nm, respectively. Because the WPC gel prepared for the cleaning study contain only 25 wt. % WPC solid, the cleaning rate is expressed as four times the whey protein removal rate in all the following calculations.

Results and Discussion

The cleaning experiments were carried out under various experimental conditions with flow velocities ranging from 0.07 m/s (Re = 2400) to 0.62 m/s (Re = 21,000) and temperatures ranging from 35 to 85°C using 0.5 wt. % NaOH cleaning solutions. The amount of WPC gel layer on the stainless steel surface ranges from 205 to 732 g/m^2 with gel film thickness of 0.2 to 0.7 mm. A typical cleaning rate against time curve with the descriptions for the three cleaning stages, the reptation time (t_r), the cleaning time during the swelling and the uniform stage (t_{su}), the constant cleaning rate (R_m), and the critical mass remaining (m_c) are shown in Figure 3. The slight decrease of the cleaning rate in the uniform stage might be atributable to the faster removing of the inlet region of the gel layer caused by the hydrodynamic disturbance. The swell of the WPC gel film during the cleaning process could also have some effects on the calculation of cleaning rate. A repeatability study of the experiments was conducted and no significant differences were observed between the cleaning rate curves.

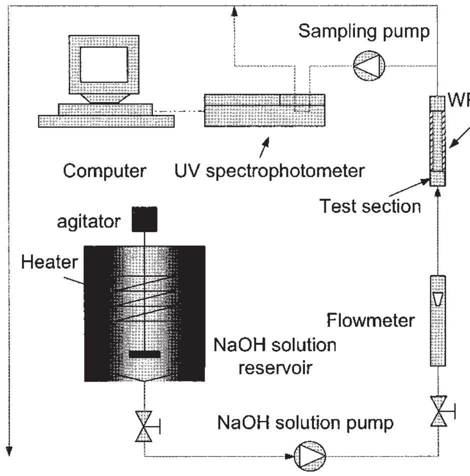

Figure 2. Cleaning apparatus.

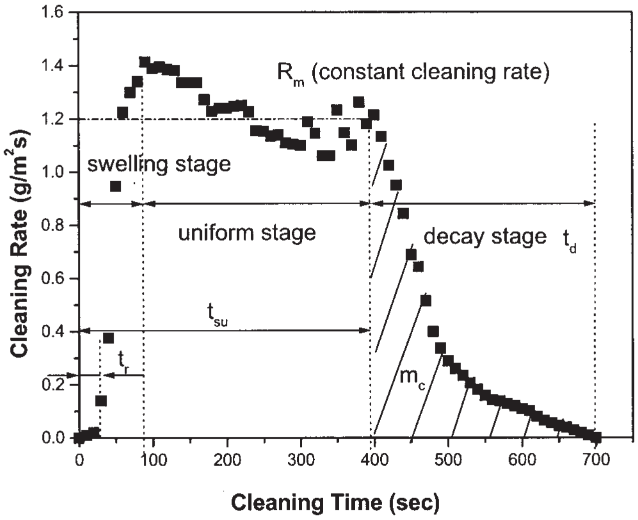

Figure 3. A typical cleaning rate against cleaning time curve.

Conditions: 65°C, 0.25 m/s, and 0.5 wt % NaOH.

The accuracy of the cleaning experiments is dependent on the agreement between the total dissolved mass of the WPC gel calculated using the UV absorbance data (m_{uv}) and the original mass of the WPC gel coated on the inside of the tubes (m_o). A ratio between 0.92 and 1.04 was achieved when the Reynolds number (Re) was below 15,000. If the WPC gel layer is completely dissolved by the cleaning solution, the value should be equal to 1. The possible reason for the deviations from this value might be (1) air bubbles and some impurities in the cleaning solution affected the UV absorbance measurements; (2) the WPC gel layer might not have completely dissolved in the cleaning solution, and some small amount of the WPC gel layer may be removed as lumps when the gel layer was broken. In particular, when the Reynolds number was high, the effect of the shear force would be very significant. Thus the ratio of m_{uv} to m_o was found to be only 0.75 for Re > 20,000. In this case, the polymer dissolution–based cleaning model may no longer be applicable.

Identification of model parameters

A cleaning model combining the polymer dissolution concept and the boundary layer mass-transfer theory has been used to describe the cleaning process. During the swelling and uniform stage, the cleaning rate is given by Eq. 9. Four parameters (R_m, ξ, t_r, and ψ) are used to characterize the cleaning process.

The constant cleaning rate R_m is the most important parameter. Because the removal of the film was not very uniform along the tube length, selecting the highest value to be R_m is misleading. Therefore, an average cleaning rate value was determined from the experimental results using the following equation

$$R_m = \frac{m_u}{t_u} \tag{24}$$

where m_u and t_u are the mass removed and the cleaning time during the uniform stage, respectively.

During the swelling stage, after the cleaning solution contacts with the deposit, a certain delay time, referred to as the "reptation time," exists before a finite amount of dissolved WPC protein becomes measurable in the cleaning solution. With an accurate on-line measurement of the cleaning kinetics, Gallot-Lavallée et al. (1982) showed that the initial cleaning rate is zero. In the study of whey protein deposits cleaning, a delay before the start of the cleaning process was also observed (Bird and Fryer, 1991). The theoretical basis of the reptation time as a function of flow velocity and temperature is not available. However, at low temperature and low flow velocity, the reptation time can be roughly observed directly from the experimental results (as shown in Figure 4). The intercept values of the lines from the swelling curves with the x-axis may be recognized as the reptation times.

For a given polymer–solution pair and polymer molecular weight, it is reasonable to assume a unique value of interfacial polymer volume fraction at the gel–solution interface (Papanu and Soane, 1989; Tu and Ouano, 1977). A recent study showed that the interfacial polymer concentration did not vary significantly with time during the dissolution process (Devotta et al., 1995). The product of the molecular weight and the maximum volume fraction has an approximately constant value and it has been reported to be about 27,000 daltons for the polystyrene–MEK system (Graessley, 1974). Using these data as a guideline, Tu and Ouano (1977) successfully modeled the polystyrene dissolution kinetics. During the fouling and cleaning process, the molecular weight of the disengaged protein is difficult to estimate. For simplicity, it is assumed that the dimensionless value ψ is a constant. By rearranging Eq. 9, a relationship between ψ and the reptation time can be expressed by the following equation

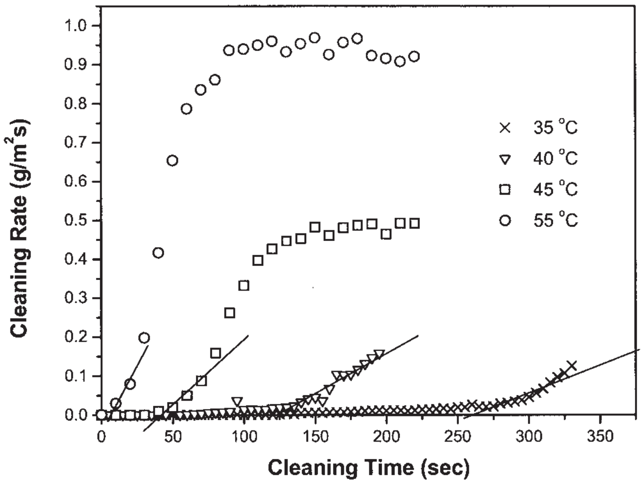

Figure 4. Estimation of reptation time at various temperatures from cleaning rate against time plots.

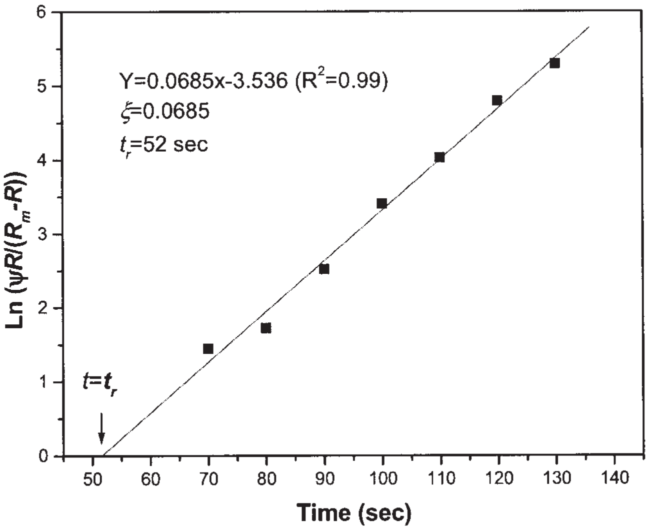

Figure 5. A typical plot of ln[$\psi R/(R_m - R)$] against time used to identify the values of ξ and t_r from the experimental results.

$$\ln\left(\frac{\psi R}{R_m - R}\right) = \xi(t - t_r) \tag{25}$$

Using the measured reptation times at various low temperatures (see Figure 4), a simple regression method was used to identify the value of ψ. For example, using the cleaning rate data at 45°C, as shown in Figure 4, a plot of ln[$\psi R/(R_m - R)$] against time should give a nearly straight line passing through the point where $t = t_r$ when ln[$\psi R/(R_m - R)$], providing that an appropriate value for ψ was selected (see Figure 5). The minimized sum of the square deviations was obtained when an average ψ value of 25 ± 5 was chosen. The value of ψ is independent of ξ, and subsequently is used in all the other calculations. After obtaining the value of ψ, it is also possible to determine ξ and t_r from the same plot, especially when t_r is too small to be directly observed from the experimental results (see Figure 5).

The time at which the decay stage commences is given by Eq. 18 and the cleaning rate during this stage is given by Eq. 15. The other two parameters m_c and k_A were used to model the cleaning progress during the decay cleaning stage. The mass of the gel layer left at the start of the decay stage is referred to as the critical mass (m_c), and can be used to determine the swelling–uniform cleaning time t_{su}. The critical mass value can be calculated by the area integration of the cleaning rate curve in the decay stage, as shown in Figure 3. k_A is the rate constant for the surface area reduction in the decay stage. By rearranging Eq. 15, the following equation is obtained

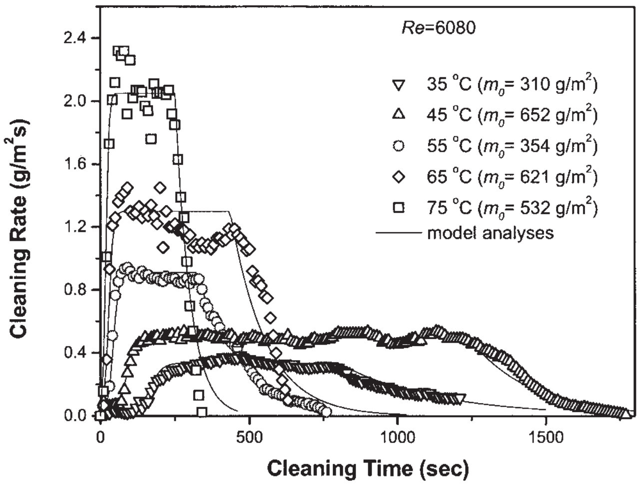

Figure 6. Comparison of the experimental and predicted cleaning results at different temperatures at a constant Reynolds number (Re = 6080) using the 0.5 wt % NaOH.

$$\ln\left(\frac{R}{R_m}\right) = k_A(t - t_{su}) \tag{26}$$

The value of k_A can be determined from the slope of the plot of $\ln(R/R_m)$ against $(t - t_{su})$.

Effects of temperature and velocity on cleaning

The effect of temperature on the cleaning rate has been studied at a constant Reynolds number (Re = 6080) at temperatures ranging from 35 to 75°C. The experimental results are illustrated in Figure 6. The effect of flow velocity (0.09–0.46 m/s) on the cleaning rate at 65°C is shown in Figure 7. The model parameters obtained from the analysis of experimental results are summarized in Tables 1 and 2. The reptation time (t_r) was calculated using the method described in Figure 5. In the analysis of the model parameters, it was found that the critical mass (m_c) was only slightly dependent on temperature and flow velocity; as a result, the critical mass (m_c) may be taken as a characteristic parameter for a given fouling and cleaning system. An average critical mass value of 100 g/m^2 was determined from the experimental data for the WPC gel deposits used in this study.

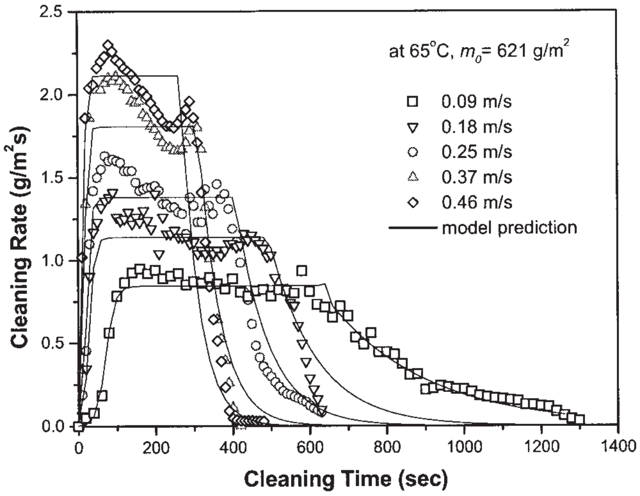

Figure 7. Comparison of the experimental and predicted cleaning results at different flow velocities at 65°C using the 0.5 wt % NaOH.

Table 1. Model Parameters Obtained from the Analyses of Experimental Results Given in Figure 6

Temperature (°C)	35	45	55	65	75
Velocity (m/s)	0.29	0.24	0.21	0.18	0.16
t_r (s)	100	40	14	7	2
ξ (s^{-1})	0.05	0.07	0.11	0.17	0.20
k_A (s^{-1})	0.003	0.005	0.007	0.009	0.020
R_m (g/m^{2}s)	0.33	0.48	0.84	1.12	2.07

Figures 6 and 7 show that with increasing of the flow velocity and temperature of the cleaning solution, the cleaning rate during the swelling and uniform stages was increased and the cleaning time in the decay stage was reduced. As a result, the whole cleaning time is reduced. These observations are consistent with previous cleaning studies (Gillham et al., 1999).

The relationship between temperature and t_r (reptation time) can be expressed as (Peppas and Wu, 1994)

$$t_r \propto (\eta/k_B T) M^3 \phi^{1.5} \tag{27}$$

where k_B is the Boltzmann constant, ϕ is the polymer volume fraction, η is viscosity, and M is the polymer molecular weight of polymer. Equation 27 suggests that the reptation time gets shorter with increasing temperature. As can be seen from Table 2, the t_r values decreased with increasing velocities, which could be related with the influence of the velocity on the detachment of the disengaged protein chains from the gel surface at the gel–solution interface.

It is expected that the values of ξ increase with increasing temperature because the protein chains have higher mobility and larger disengagement rates at higher temperatures. Both the increased maximum volume fraction of disengaged protein molecules and mass-transfer coefficient should contribute to a larger R_m at higher temperatures. The changes of k_A with temperature are possibly attributable to the changes in the microstructure and the mechanical properties of the fouling layer with temperature.

The influence of the flow velocity on the cleaning process was previously noted (Plett, 1985), and the current results confirm the same trend. If the shear force does not cause the detachment of the WPC film, the enhancement of the cleaning with increasing flow velocity would be mainly attributed to the increase of mass-transfer coefficient in the boundary layer. As a result, the transport of the disengaged protein molecules back

Table 2. Model Parameters Obtained from the Analyses of Experimental Results Given in Figure 7

Velocity (m/s)	0.09	0.18	0.25	0.37	0.46
t_r (s)	25	7	2.4	0	0
ξ (s^{-1})	0.08	0.15	0.16	0.20	0.26
k_A (s^{-1})	0.004	0.009	0.012	0.020	0.023
R_m (g/m^{2}s)	0.88	1.21	1.39	1.85	2.07

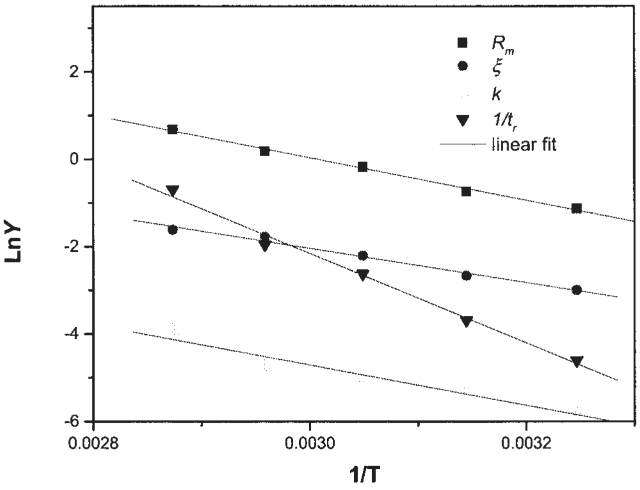

Figure 8. Arrhenius plots [ln ξ, ln R_m, ln k_A, and ln($1/t_r$) against $1/T$] for the WPC gel films treated with the 0.5 wt % NaOH.

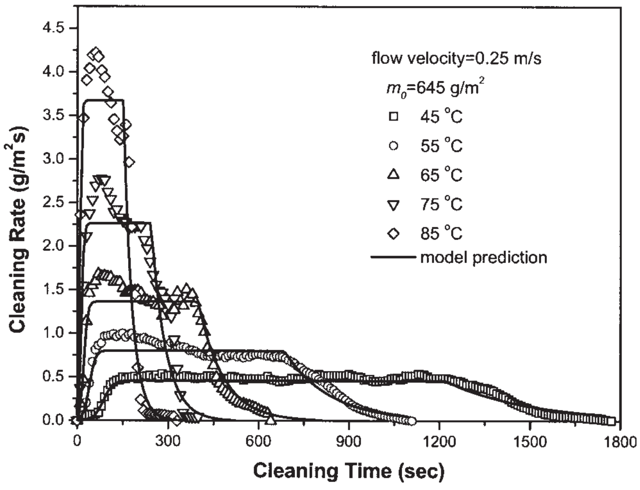

Figure 9. Comparison of the experimental and predicted cleaning results at various temperatures at a flow velocity of 0.25 m/s.

into the bulk cleaning solution would be more efficient at higher flow velocities, thus giving higher values of R_m and k_A.

The disengagement rate would be influenced by the action of the hydrodynamic forces on the polymer chains dangling into the liquid, so it is expected that ξ increases with increasing flow velocity as well. Faster disengagement of polymer chains could contribute a shorter reptation time.

To evaluate the role of temperature in each cleaning stage, the temperature-dependent cleaning model parameters [R_m, ξ, k_A, and ($1/t_r$)] are described using the Arrhenius relationship. Using the experimental data provided in Table 1, ln ξ, ln R_m, ln k_A, and ln($1/t_r$) against $1/T$ curves were plotted in Figure 8. The slope of the curves was then used to calculate the apparent activation energies for the various cleaning stages. The results are presented in Table 3. In this study, we have assumed the same controlling mechanism for both uniform and decay stage cleaning, and that the similar value of apparent activation energy for both stages is in agreement with our model assumption. The significant larger activation energy value for reptation time than other parameters indicates a different control mechanism.

Although the effects of Reynolds number (or flow velocity) on the model parameters could be very complex, it was possible to describe the influence of Reynolds number on the model parameters with a simple equation using the data provided in Table 2 because the effects of Reynolds number on the apparent activation energies of the cleaning rate during the whole cleaning stage are not significant (Gillham et al., 1999; Tuladhar et al., 2001). In this study, it is assumed that the apparent activation energy is independent of Reynolds number. Thus, the dependency of the model parameters on temperature and Reynolds number can be defined using the following semiempirical equation

$$Y = f(\mathrm{Re})\exp\left(-\frac{E_a}{R_g T}\right) \tag{28}$$

where Y represents the model parameters R_m, ξ, k_A, and $1/t_r$; E_a is the apparent activation energy (J/mol); R_g is the molar gas constant, and it is assumed that this is a linear relationship between the Reynolds number and Y within the Re number range used in this study, which is independent of temperature and defined as

$$f(\mathrm{Re}) = \alpha + \beta\,\mathrm{Re} \tag{29}$$

where α and β are the constants.

The comparison between experimental results and model predictions are provided in Figures 6 and 7. The parameters used in the model predictions were given in Table 3. To confirm the validity of the model presented here, a second set of cleaning experiments were performed at different temperatures ranging from 45 to 85°C, at a constant flow velocity of 0.25 m/s. Using the parameters in Table 3, the cleaning rates are predicted and compared with the experimental results in Figure 9. As can be seen from Figure 9, good agreement was observed.

The constant cleaning rate during the uniform cleaning stage is one of the most important parameters determining the overall efficiency of a cleaning process. As shown in Eq. 10, the constant cleaning rate has been defined as a product of mass-transfer coefficient and maximum volume fraction. Because the fluid flow during the whole cleaning process is carried out at a well-defined condition, identification of the mass-transfer coefficient is possible. The maximum volume fraction of disengaged protein molecules cannot be easily estimated because of

Table 3. Model Parameters Used in the Model Predictions for the Removal of WPC Gel Fouling Deposits

Cleaning Stage	Parameter (units)	α	β	E_a (kJ/mol)
Reptation	$1/t_r$ (s^{-1})	−2.7E+12	1.0E+09	85
Swelling	ξ (s^{-1})	6.5E+03	1.4E+00	33
Uniform	R_m (g/m^{2}s)	1.0E+06	1.8E+02	41
Decay	k_A (s^{-1})	−5.6E+02	1.2E+00	38

Note: The units of α and β are the same as the parameters of R_m, ξ, k_A, and $1/t_r$.

the lack of the information about the molecular weight of the disengaged protein chains during the cleaning process. However, it is reasonable to assume that when the maximum volume fraction of polymer in the gel–solution interface is reached, a saturation concentration is also obtained. Thus, the maximum volume fraction of disengaged protein molecules can be substituted by the saturation concentration of the WPC gel in the dissolution-based model. In the case of turbulent flow through a long, cylindrical pipe with a smooth wall, the constant cleaning rate for a WPC gel was predicted accurately in our previous cleaning study (Xin et al., 2002b)

$$R_m = 0.0263\left(\frac{\rho du}{\eta}\right)^{0.8}\left(\frac{\eta}{\rho D}\right)^{1/3}\frac{DC_s}{d} \tag{30}$$

where D is the mass diffusivity of the disengaged protein molecules in the boundary layer (m/s^2), d is the diameter of the tube (m), u is flow velocity (m/s), η is the viscosity of the cleaning solution (Pa s^{-1}), ρ is the density of the cleaning solution (kg/m^3), and C_s is the saturation concentration (g/m^3).

In Figure 10, the experimental constant cleaning rates observed from Figures 6, 7, and 9 were compared with that of the mass-transfer predictions (Eq. 30) and semiempirical model predictions (Eq. 28). The good agreement between experimental and predictive results further confirmed the validity of cleaning model provided in this study.

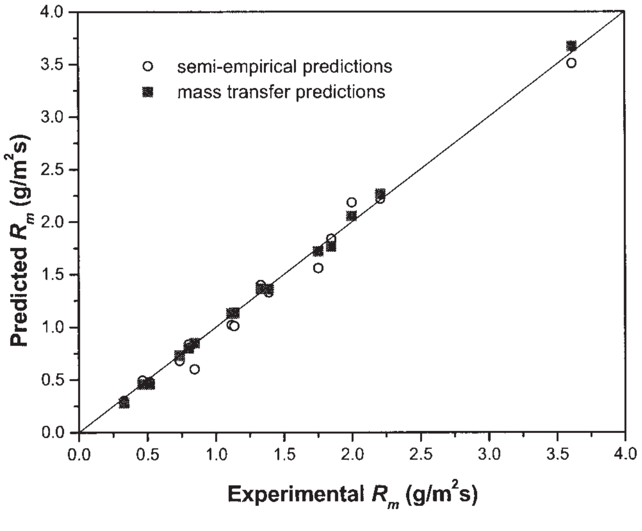

Figure 10. Comparison of the experimentally measured constant cleaning rates with the predicted ones calculated using the semiempirical equation and the mass-transfer theory (Eq. 29).

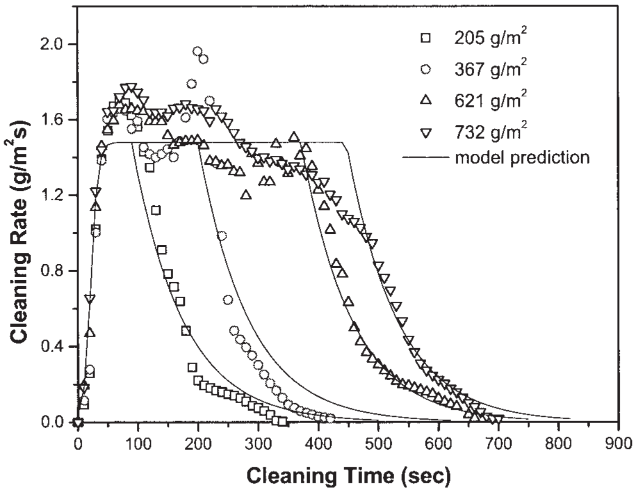

Figure 11. Comparison of the experimental and predicted cleaning results with various masses at a flow velocity of 0.25 m/s and 65°C using the 0.5 wt. % NaOH.

Various investigators have reported the apparent activation energy values ranging from 23 to 80 kJ/mol for the cleaning of whey protein fouling deposits using alkaline solutions (Bird and Bartlett, 1995; Gillham et al., 1999; Nagata et al., 1995). In a recent study, apparent activation energies of 46–50 kJ/mol for the swelling stage, 43–45 kJ/mol for the decay stage, and 41–42 kJ/mol for the overall cleaning rate have been reported (Tuladhar, 2001). The agreement between the values of apparent activation energy for the WPC gels and the actual fouling deposits indicates the similarity of the physical and chemical properties, as well as the controlling cleaning mechanisms between the thermally induced WPC gel and whey protein fouling. In the polymer dissolution process, an activation energy of 25 kJ/mol has been reported for the dissolution of poly(methyl methacrylate) films in mixed solvents (Cooper et al., 1986). The similar activation energy observed in both polymer dissolution and cleaning study has further confirmed our cleaning model assumption that disengagement and mass transfer control the cleaning process when a mechanical removal is not significant.

Effects of the amount of initial deposits on cleaning

Gallot-Lavallée et al. (1982) suggested that the cleaning rate is a zero-order reaction with respect to the initial mass of the deposit. Gillham et al. (1999) also showed that during the uniform stage, the cleaning rate is independent of the amount of the deposit present as long as swelling of the deposit does not cause significant duct reduction. Experimental results for the WPC gel films with different masses (thickness) at a flow velocity of 0.25 m/s and at 65°C are shown in Figure 11.

It is obvious that the cleaning time became longer with increasing of the deposits amount. For the WPC gel with the lowest mass (212 g/m^2), the uniform stage does not appear in the cleaning rate curve attributed to the early onset of the decay stage. However, the cleaning profiles are almost unaffected by the mass of the WPC gel films and follow the same trend. The less-evident starting of the decay stage for 732 g/m^2 gel film could be caused by the removal of a piece of gel at the end of the uniform cleaning stage. Using the parameters in Table 3, the cleaning rates are predicted and compared with the experimental results in Figure 11. Reasonably good agreement was obtained. The overprediction of the cleaning times are mainly caused by use of a smaller value of constant cleaning rate in the prediction.

Table 4. Formation Conditions and Properties of the Proteinaceous Fouling Used in Different Cleaning Studies

	Authors		
Condition	Gillham et al.	Fryer and Bird	Gallot-Lavallée and Lalande
Fluid used for fouling	3.5 wt. % WPC	3 wt % WPC	Whole milk
Formation conditions	80–87°C at 4600 Re	75–95°C at 6000 Re	85°C at 0.6 m/s
Deposit mass	70–170 g/m^2 (protein)	100 g/m^2	0.05–1.2 kg/m^2
Water content of fouling	58–66 wt %	30 wt %	—
Composition of fouling	Mainly protein	90% protein	60% protein, 30% fat, and 10% minerals
Monitoring method	Protein assay	Protein assay	Turbidity measurement
Expression for cleaning rate	Protein	Protein	Total mass (protein + water)
Cleaning tube	Length 250 mm ID 16 mm	Length 100 mm ID 6 mm	Length 100 mm ID 6 mm

Previous Cleaning Results

The cleaning model proposed here is obtained from the investigation of a model system based on WPC gel films. Comparing this model system with more realistic experimental systems reported previously by other researchers would not only confirm the validity of the cleaning model proposed in this study, but also make it possible to apply this cleaning model to realistic protein deposits.

Systematic studies on the cleaning kinetics of whole milk fouling (Gallot-Lavallée and Lalande, 1985) and whey protein fouling (Fryer and Bird, 1994; Gillham et al., 1999) have been presented using the cleaning rate curves. The experimental methods and properties of proteinaceous fouling in these studies are summarized in Table 4. Similar to the WPC gel removal, all the cleaning results from previous studies have shown a typical cleaning rate curve with three well-defined stages. Using the current model, the data from these previous studies were analyzed (see Table 5). From Table 5, it can be seen that the influence of temperature and flow velocity on the model parameters shows a similar trend as that of WPC gel, confirming the cleaning mechanism developed to be valid in more complex and realistic proteinaceous fouling and cleaning systems.

Although the conditions of fouling and cleaning are quite different between real milk protein fouling and WPC gel model fouling, it was found that the apparent activation energies obtained from this study are still valid for the protein deposits. The effects of Reynolds number on model parameters were then calculated according to the results obtained from the analyses of the experimental curves using Eqs. 28 and 29. An average critical mass of 34 g/m^2 was estimated for the experimental results of Gillham et al. and Fryer and Bird; and an average critical mass of 160 g/m^2 was estimated for the experimental results of Gallot-Lavallée, respectively. Because of the shortage of the data at the beginning period of cleaning process, the reptation time was taken as zero for all the following predictions. The literature experimental results and model predictions are compared in Figure 12 (Gallot-Lavallée and Lalande, 1985), Figure 13 (Gillham et al., 1999), and Figure 14 (Fryer and Bird, 1994). The parameters used for the model predictions are summarized in Table 6.

The same value of the critical mass identified from each real fouling and cleaning system suggests that the critical mass may be taken as a system constant. Because of the lack of reliable data, especially at the swelling cleaning period, the prediction could not be very accurate, which may explain the different α-value obtained for a similar fouling and cleaning system (Fryer and Bird, 1994; Gillham et al., 1999). The model parameters used here may be valid only within the experimental conditions as a result of choosing a simple linear relationship. More systematic and repeatable data are needed to establish a more accurate relationship to evaluate the effect of Reynolds number on model parameters within a wide range of experimental conditions.

The comparison of the constant cleaning rate (R_m) between cleaning results reported in the literature with that of WPC gels performed under the same operation conditions could provide valuable information about the cleaning properties of different proteinaceous fouling (see Table 5). The similar trend of R_m values observed from the experimental results and the WPC gel model predictions in all the tested conditions suggest that the cleaning properties of whey protein fouling and whole milk fouling formed in their experiments behaved similarly to the WPC gel used in this study. The larger R_m values in Gillham

Table 5. Model Parameters Obtained from the Analyses of Experimental Results (Figures 12–14) and the Comparison of the Constant Cleaning Rates

	Authors								
Parameter	Gallot-Lavallée and Lalande			Gillham et al.			Fryer and Bird		
Temperature (°C)	55	75	95	30	50	70	70	70	70
Velocity (m/s)	1.1	1.1	1.1	0.464	0.464	0.464	0.175	0.35	0.525
ξ (s^{-1})	0.20	0.40	4.00	0.02	0.10	0.17	0.15	0.20	0.30
k_A (s^{-1})	0.011	0.041	0.130	0.001	0.011	0.021	0.015	0.018	0.025
m_c (g/m^2)	160	160	160	34	34	34	34	34	34
m_0 (g/m^2)	290	360	240	103	98	90	94	80	110
R_m (g/m^{2}s)*	2.50	8.00	21.00	0.07	0.22	0.49	0.48	0.76	0.85
R_m (g/m^{2}s)**	2.50	6.73	16.25	0.04	0.13	0.36	0.25	0.32	0.40

*The experimental values.
**Predicted results using the parameters obtained from the WPC gel system.

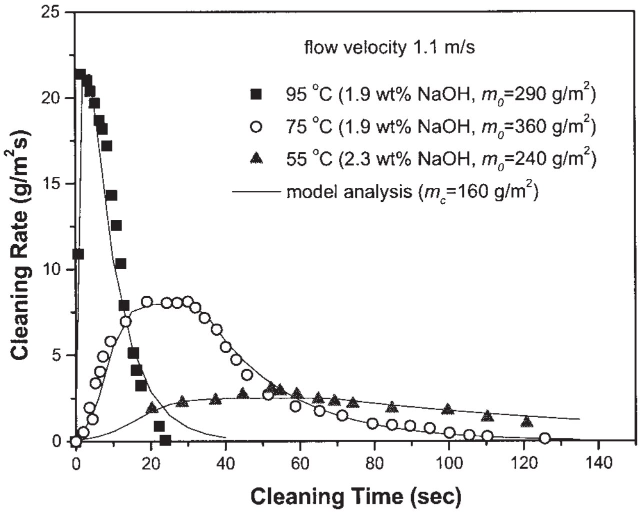

Figure 12. Model predictions together with the experimental results reported by Gallot-Lavallée and Lalande (1985).

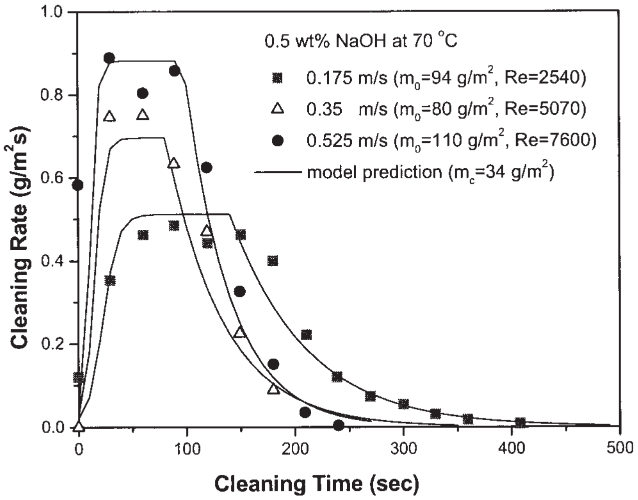

Figure 14. Model predictions together with the experimental results reported by Fryer and Bird (1994).

and Bird's experiments indicate that the whey protein fouling could be more easily removed than could WPC gel fouling during the uniform cleaning stage. Certainly, in Gallot-Lavallée's experiment, using 2 wt % NaOH instead of 0.5 wt % NaOH as cleaning solutions, and using the turbidity method instead of the UV absorption method to monitor the cleaning kinetics may have some effects on the accuracy of interpreting the cleaning properties of the whole milk fouling deposits.

Conclusions

Understanding the chemical reaction, mass transfer, and mechanical forces involved in the cleaning process is important in optimizing a CIP system. Using the WPC gel as a model material and a rapid continuous UV assay method, a comprehensive study of the cleaning of the WPC gel deposits by NaOH cleaning solutions was conducted. The kinetics of cleaning have been established and expressed in mathematical terms. This research has provided insights into the removal mechanism of milk protein deposits from the metal surface under various experimental conditions.

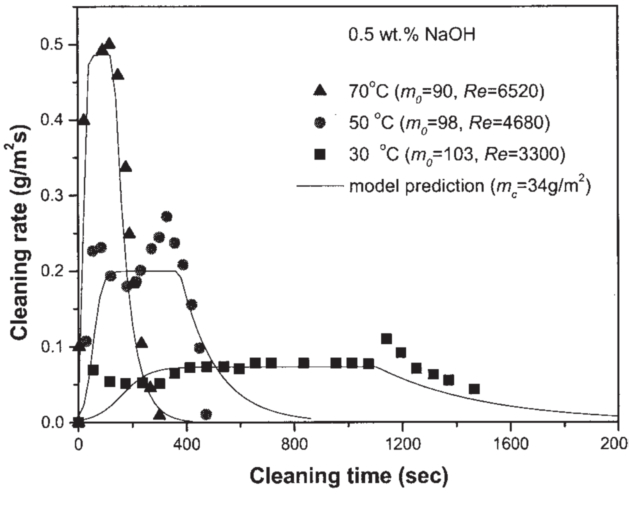

Figure 13. Model predictions together with the experimental results reported by Gillham et al. (1999).

Dissolution of the protein fouling deposits in alkaline solutions plays the major role in the removal of WPC gel deposits. The governing factor is the balance between the rate of generating the dissolvable protein molecules at the gel–solution interface and the rate of removing these molecules from this interface. This study suggests that the disengagement of protein molecules from the swollen gel–solution interface and the transfer of these disengaged protein molecules back into the bulk cleaning solutions are the controlling cleaning mechanisms. It has also introduced a useful model material and a monitoring method for studying cleaning kinetics.

Based on polymer dissolution theory and the concept of fundamental mass transfer, a cleaning model was developed for estimating the cleaning rate and cleaning time for proteinaceous fouling. Various processes, such as reptation, disengagement, mass transfer through the boundary layer, and surface area changes were taken into account in the modeling process. The experimental results and model predictions support the modeling concepts used. The successful use of this model in literature cleaning results shows that this new mathematical model can be applied in a real fouling and cleaning process.

Several researchers have reported that both mechanical and dissolution mechanisms are involved in the removal of contaminant films (Harriott and Hamilton, 1965; Jennings, 1965). The current dissolution model does not account for the role of shear in removing large aggregates. However, the model provides a good foundation for further studies on the cleaning mechanisms of protein-based milk fouling.

Acknowledgments

This work was partially sponsored by the Foundation for Research, Science and Technology Program of New Zealand; Fonterra Co-operative Group Ltd.; and Orica (NZ) Ltd.

Table 6. Model Parameters Used in Prediction for the Literature Cleaning Results Provided in Figures 12, 13, and 14

	Authors					
	Gallot-Lavallée and Lalande		Gillham et al.		Fryer and Bird	
Parameter	α	β	α	β	α	β
ξ (s^{-1})	−1.5E+05	5.6	7.7E+02	2.3	6.3E+03	2.8
R_m (g/m^2s)	1.0E+06	187	7.0E+05	3.4	4.8E+05	108
k_A (s^{-1})	−1.5E+04	0.88	6.5E+03	1.1	6.4E+03	1.4
Re range	32,000–58,000		3300–6520		2540–7600	

Note: The same apparent activation energies as that of WPC gels have been used; the unit of α and β are the same as the parameters of R_m, ξ, k_A, and $1/t_r$.

Notation

A = surface area of deposits, m^2
A_L = deposit surface area left in decay stage, m^2
$A_{L,}$ = total surface area covered by the protein film, m^2
C_s = the saturation concentration, g/m^3
d = diameter of a stainless steel tube, m
D = mass diffusivity of the disengaged protein molecules, m/s^2
E_a = the apparent activation energy, kJ/mol
$f(\mathrm{Re})$ = linear function between Reynolds number and model parameters (Eq. 28)
k = a reaction rate
k_A = the first-order rate constant for the surface area reduction (Eq. 12), s^{-1}
k_B = Boltzmann constant
k_d = disengagement rate constant (Eq. 3), s^{-1}
k_ϕ = mass-transfer coefficient (Eq. 1), g/m^2s
m = mass removed, g
m_c = critical mass, g/m^2
m_o = original deposit mass, g/m^2
m_u = total mass removed during uniform stage, g/m^2
m_{su} = total mass removed during swelling and uniform stage, g/m^2
m_{uv} = dissolved mass calculated using the UV absorbance data, g/m^2
M = molecular weight, dalton
R = cleaning rate, g/m^2s
R_g = ideal gas constant, J/mol K
Re = Reynolds number
R_m = constant cleaning rate, g/m^2s
T = absolute temperature, K
t_d = cleaning time in decay stage, s
t_r = reptation time, s
t_{su} = sum of cleaning times in swelling and uniform stage, s
t_t = total cleaning time, s
t_u = cleaning time in uniform stage, s
u = flow velocity, m/s
Y = symbol of model parameters (Eq. 28)

Greek letters

α = a constant (Eq. 29)
β = a constant (Eq. 29)
ρ = density of the cleaning solution, kg/m^3
ϕ = volume fraction of the disengaged protein molecules gel–solution interface (Eq. 1)
ϕ_b = volume fraction of the disengaged protein molecules in the bulk cleaning solution (Eq. 1)
ϕ_m = maximum volume fraction of the disengaged protein molecules (Eq. 4)
ϕ_0 = volume fraction of the tangling protein chains at the solution side of gel–solution interface at the time $t = t_r$ (Eq. 7)
η = viscosity, Pa s^{-1}
ξ = kinetic constant (Eq. 4), s^{-1}
ψ = dimensionless parameter (Eq. 8)

Literature Cited

Beaudoin, S. P., C. S. Grant, and R. G. Carbonell, "Removal of Organic Films from Solid Surface Using Aqueous Solutions of Non-ionic Surfactants," *Ind. Eng. Chem. Res.*, **34**, 3307 (1995).

Biddle, V., "Chemistry Precision and Design," *Beka Book Science Series*, Beka Book Publications, Pensacola, FL (1986).

Bird, M. R., and M. Bartlett, "CIP Optimization for the Food Industry: Relationship Between Detergent Concentration, Temperature and Cleaning Time," *Trans. IChemE*, **73**(Part C), 63 (1995).

Bird, M. R., and P. J. Fryer, "An Experimental Study of the Cleaning of Surfaces Fouled by Whey Proteins," *Trans. IChemE*, **69**(Part C), 13 (1991).

Cooper, W. J., P. D. Krasicky, and F. Rodriguez, "Dissolution Rates of Poly(methyl methacrylate) Films in Mixed Solvents," *J. Appl. Polym. Sci.*, **31**, 65 (1986).

Devotta, I., V. D. Amebeskar, A. B. Mandhare, and R. A. Mashelkar, "The Life Time of a Dissolving Polymeric Particle," *Chem. Eng. Sci.*, **49**(5), 645 (1993).

Devotta, I., M. V. Badiger, P. R. Rajamohanan, S. Ganapathy, and R. A. Mashelkar, "Unusual Retardation and Enhancement in Polymer Dissolution: Role of Disengagement Dynamics," *Chem. Eng. Sci.*, **50**(16), 2557 (1995).

Devotta, I., and R. A. Mashelkar, "On Optimal Temperature for Dissolution of Polymers in Hydrogen Bonding Solvents," *Chem. Eng. Sci.*, **51**(15), 3881 (1996).

Fryer, P. J., and M. R. Bird, "Factors Which Affect the Kinetics of Cleaning Dairy Soils," *Food Sci. Technol. Today*, **8**(1), 36 (1994).

Gallot-Lavallée, T., and M. Lalande, "A Mechanistic Approach of Pasteurised Milk Deposit Cleaning," *Fouling and Cleaning in Food Processing,* Lund University, Sweden, 374 (1985).

Gallot-Lavallée, T., M. Lalande, and G. Corrieu, "An Optical Method to Study the Kinetics of Cleaning Milk Deposits by Sodium Hydroxide," *J. Food Proc. Eng.*, **5**, 131 (1982).

Gillham, C. R., P. J. Fryer, A. P. M. Hasting, and D. I. Wilson, "Cleaning-in-Place of Whey Protein Fouling Deposits: Mechanisms Controlling Cleaning," *Food Bioprod. Proc.*, **77**(C2), 127 (1999).

Gillham, C. R., D. I. Wilson, P. J. Fryer, and A. P. M. Hasting, "Studies in Cleaning of Whey Protein Deposits," *Fouling and Cleaning in Food Processing,* Jesus College, Cambridge University, Cambridge, UK, 271 (1998).

Graessley, W. W., "The Entanglement Concept in Polymer Rheology," *Adv. Polym. Sci.*, **16**, 1 (1974).

Harriott, P., and R. M. Hamilton, "Solid–Liquid Mass Transfer in Turbulent Pipe Flow," *Chem. Eng. Sci.*, **20**, 1073 (1965).

Jennings, W., "Theory and Practice of Hard Surface Cleaning," *Adv. Food Res.*, **14**, 325 (1965).

Jeurnink, T. J. M., and D. W. Brinkman, "The Cleaning of Heat Exchangers and Evaporators After Processing Milk or Whey," *Int. Dairy J.*, **4**, 347 (1994).

Lalande, M., and F. Rene, "Fouling by Milk and Dairy Product and Cleaning of Heat Exchange Surfaces," *Fouling Science and Technology*, L. F. Melo, T. R. Bott, and C. A. Bernardo, eds., Kluwer Academic, Dordrecht, The Netherlands, 557 (1988).

Lalande, M., J. P. Tissier, and G. Corrieu, "Fouling of Heat Transfer Surfaces Related to β-Lactoglobulin Denaturation during Heat Processing of Milk," *Biotechnol. Proc.*, **1**(2), 131 (1985).

Murray, J. D., *Mathematical Biology I: Introduction*, 3rd Edition, Springer, New York (2002).

Nagata, A., T. Sakiyama, H. Itoh, T. Toyomasu, E. Enomoto, T. Nagai, T. Saeki, and K. Nakanishi, "Comparative Study on Caustic and Enzymatic Cleanings of Stainless Steel Surface Fouled with β-Lactoglobulin," *Biosci. Biotechnol. Biochem.*, **59**(12), 2277 (1995).

Narasimhan, B., "Mathematical Models Describing Polymer Dissolution:

Consequences for Drug Delivery," *Adv. Drug Deliv. Rev.*, **48**, 195 (2001).

Papanu, J. S., and D. S. Soane, "Transport Models for Swelling and Dissolution of Thin Polymer Films," *J. Appl. Polym. Sci.*, **38**, 859 (1989).

Parker, A., F. Vigouroux, and W. F. Reed, "Dissolution Kinetics of Polymer Powders," *AIChE J.*, **46**(7), 1290 (2000).

Peppas, N. A., and J. C. Wu, "Mathematical Modeling and Experimental Characterization of Polymer Dissolution," *Macromolecules*, **27**, 5626 (1994).

Plett, E. A., "Cleaning of Fouled Surfaces," *Fouling and Cleaning in Food Processing*, Lund University, Sweden, 286 (1985).

Ranade, V. V., and R. A. Mashelkar, "Convective Diffusion from a Dissolving Polymeric Particle," *AIChE J.*, **41**(3), 666 (1995).

Schlussler, H. J., "Cleaning of Hard Surface in the Food Industry," *Milchwissenschaft*, **25**(3), 133 (1970).

Tu, Y., and A. C. Ouano, "Model for the Kinematics of Polymer Dissolution," *IBM J. Res. Dev.*, **Mar.**, 131 (1977).

Tuladhar, T. R., "Development of a Novel Sensor for Cleaning Studies," PhD Dissertation, University of Cambridge, UK (2001).

Xin, H., X. D. Chen, and N. Özkan, "Whey Protein Based Gel as a Model Material for Studying the Initial Cleaning Mechanisms of Milk Fouling," *J. Food Sci.*, **67**(7), 2702 (2002a).

Xin, H., X. D. Chen, and N. Özkan, "Cleaning Rate in the Uniform Cleaning Stage for Whey Protein Gel Deposits," *Trans. IChemE*, **80**(Part C), 240 (2002b).

Manuscript received Sep. 10, 2002, and revision received Oct. 30, 2003.