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THE BEHAVIOR OF ß-LACTOGLOBULIN PROTEIN IN PLATE HEAT EXCHANGER'S CHANNEL DURING MILK HEAT TREATMENT

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A major problem related to heat treatment of milk is formation of deposits. This is due to the chemical alteration of β-lactoglobulin protein that leads to formation of coatings on the walls of the equipment. These deposits induce losses in the thermal performance of the heat exchanger. In order to provide an adequate prediction of these deposits, we present a two-dimensional (2D) modelling study of the channel of a plate heat exchanger. Compared with the former studies, where the domain is related to a single plate, our work is related to the area between the two plates. This approach will allow a better understanding and description of the evolution and behaviour of β-lactoglobulin protein. Equations for fluid flow, energy, and fouling were resolved. Special boundary conditions had been implemented to link the amount of deposits with thermal transfer. Predicted results for the amount of fouling deposit on the wall were validated by comparisons with experimental data available in literature. The behaviour of β-lactoglobulin protein was studied by analyzing its distribution in the channel.

Keywords: milk fouling, plate heat exchanger, modelling, β-lactoglobulin

The major problem encountered during milk heat treatment in plate heat exchangers is the deposit of unwanted materials on heat treatment surfaces, which induce performance loss of the equipment (Belmarbeiny et al., 1993). Consequently, regular cleaning steps of equipment must be scheduled generating losses in capital, energy, and production time (Visser & Jeurnink, 1997). Many authors studied the main parameters affecting milk fouling in plate heat exchangers (Bansal & Chen, 2006). These parameters involve hydrodynamics of fluid flow and thermal processes (Belmarbeiny et al., 1993), plate heat exchanger configuration and heat surface properties (Huang & Goddard, 2015). Proteins of milk, especially β-lactoglobulin, represent a key component in deposit formation.

For several years, studying milk fouling mechanisms has been a well investigated field. Lalande and co-workers (1985) reported that the formation of deposit on the heat exchange surface is attributed to the chemical alteration of β-lactoglobulin protein. Toyoda and Fryer (1997) proposed a fouling model that takes into account the mass transfer between the fluid bulk and boundary layer. Georgiadis and Macchietto (2000) established a two-dimensional mathematical model using fluid flow to predict, with more accuracy, the temperature distribution. Jun and Puri (2006) developed the previous work into a dynamic 2D model to predict milk deposit patterns on the plate surface more precisely. De Bonis and Ruocco (2009) used commercial finite element solver in order to determine velocity profile, temperature distribution, and protein distribution of the deposit in corrugated channel of plate heat exchanger. Mahdi and co-workers (2009) proposed a two-dimensional dynamic fouling model for milk fouling in a plate heat exchanger, which takes into account fouling caused by both β-lactoglobulin protein and calcium phosphate.

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Few studies have attempted to determine the kinetics of β-lactoglobulin denaturation. The effect of calcium on β-lactoglobulin denaturation have been discussed by Petit and co-workers (2011) and Erabit and co-workers (2013), they concluded that high calcium concentrations catalyze unfolding and aggregation reactions. Khaldi and co-workers (2015) carried out experiments in order to determine the relationship between the deposit mass distribution generated in plate heat exchanger and the ratio between the unfolding and aggregation rate constants. Recently, Bouvier and co-workers (2014) used CFD (Computational Fluid Dynamics) as a tool to simulate β-lactoglobulin heat induced denaturation and aggregation in plate heat exchanger. They demonstrated that the unfolded β-lactoglobulin is the main precursor generating a dry deposit on the heat transfer wall.

All these works concentrate on the amount of deposits on the surface of plate heat exchangers and none has described the behaviour of β-lactoglobulin protein during milk flow in the channels of plate heat exchangers. Therefore, the present work adapts the milk fouling model of Jun and Puri (2006) to a two-dimensional channel situated between two plates of the plate heat exchanger in order to study the transformations that β-lactoglobulin protein has undergone along the channel. In order to obtain accurate results, we present a two-dimensional (2D) modelling study. Governing equations of momentum, heat, and mass transfer in the channel were resolved using the finite volume method for the discretization of the partial differential equations. In order to link heat and mass transfer, Biot number was used as a boundary condition at the walls. First, the simulation results were validated with experimental data of Georgiadis and co-workers (1998). Then, β-lactoglobulin protein distribution in its three forms, namely native, unfolded, and aggregated, was discussed in the first channel of plate heat exchanger during milk pasteurization.

1. Mathematical formulation

In order to predict the evolution of β-lactoglobulin protein denaturation during milk flow in a channel of plate heat exchanger, we solve numerically the governing equations of fluid flow, heat and mass transfer, which represent the model of milk fouling. Figure 1 describes the channel geometry used in this study. Milk, at uniform inlet temperature T_{in} was rising between two smooth and parallel plates of 75 cm length (L) and 20 cm width (W) making a channel of 4 mm thickness (e). The plates are held at constant temperature of 90 °C.

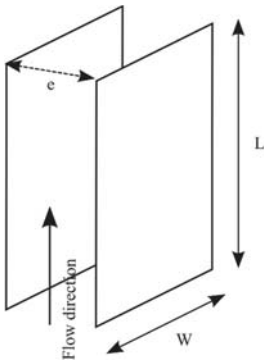

Fig. 1. Descriptive scheme of channel geometry

1.1. Fluid flow and energy equations

The equations of continuity (1) and momentum (2–3) according to axial and vertical directions for the two-dimensional laminar flow in the channel are given as:

$$\nabla \cdot \left(\rho \vec{V}\right) = 0 \tag{1}$$

$$\rho \frac{\partial u}{\partial t} + \nabla \cdot \left(\rho \vec{V} u\right) = -\frac{\partial p}{\partial x} + \mu\left(\nabla^2 \cdot u\right) \tag{2}$$

$$\rho \frac{\partial v}{\partial t} + \nabla \cdot \left(\rho \vec{V} v\right) = -\frac{\partial p}{\partial y} + \mu\left(\nabla^2 \cdot v\right) \tag{3}$$

where u and v denote, respectively, axial and vertical component of velocity, ρ is the density of milk, μ its dynamic viscosity, p represents the pressure and t time. Heat transfer in the channel is governed by:

$$\frac{\partial T}{\partial t} + \nabla \cdot \left(\vec{V} T\right) = \alpha\left(\nabla^2 \cdot T\right) \tag{4}$$

where T is the temperature and α the thermal diffusivity of milk.

1.2. Fouling model

Fouling model used in this work is based on the model presented by Jun and Puri (2006), which is mainly adapted from the work of Georgiadis and Macchietto (2000). This model is based on the chemical reactions of derivates of β-lactoglobulin protein at 65 °C and mass transfer between the thermal boundary layer and the bulk of fluid. Indeed, when milk temperature reaches 65 °C, β-lactoglobulin protein becomes thermally unstable, it is prone to chemical alteration producing molecules containing sulfhydryl groups (-SH), which polymerize irreversibly giving an insoluble aggregate. Aggregated protein adheres to the wall of the equipment. Figure 2 represents the reaction scheme of this fouling model (Georgiadis & Macchietto, 2000).

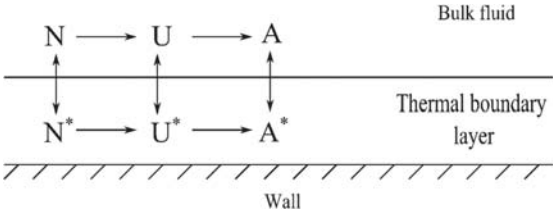

Fig. 2. Reaction scheme of β-lactoglobulin

This model is represented mathematically by mass transport of native, unfolded, and aggregated forms of β-lactoglobulin protein as mentioned below by equations (5–7), where C_i denotes the concentrations of different forms of the protein and D_i is their molecular diffusivity. $\Re_U$ and $\Re_A$ represent, respectively, the unfolding and aggregation reactions.

For modelling purposes, the variation of milk physical properties with temperature was neglected and the properties were taken as skimmed milk ones (Georgiadis & Macchietto, 2000).

$$\frac{\partial C_N}{\partial t} + \nabla \cdot \left(\vec{V} C_N\right) = D_N \left(\nabla^2 \cdot C_N\right) - \Re_U \tag{5}$$

$$\frac{\partial C_U}{\partial t} + \nabla \cdot \left(\vec{V} C_U\right) = D_U \left(\nabla^2 \cdot C_U\right) + \Re_U - \Re_A \tag{6}$$

$$\frac{\partial C_A}{\partial t} + \nabla \cdot \left(\vec{V} C_A\right) = D_A \left(\nabla^2 \cdot C_A\right) + \Re_A \tag{7}$$

Kinetics of $\Re_U$ and $\Re_A$ depend on temperature by the Arrhenius equation as described below.

$$\Re_U = k_{U_0}\, exp\left(\frac{E_U}{RT}\right) C_N^{n_U} \tag{8}$$

$$\Re_A = k_{A_0}\, exp\left(\frac{E_A}{RT}\right) C_U^{n_A} \tag{9}$$

The values of reaction order, $n_{i,}$ reaction rates constants, $k_{i_0,}$ and activation energies, $E_{i,}$ are summarized in Table 1. Mass transfer takes place in the interface between the thermal boundary layer and the bulk of fluid. Its expression, according to x position inside or outside the thermal boundary layer, is as follows:

Inside boundary layer: $$\left[-\frac{k_{m_i}}{\delta_T}\left(C_i^* - C_i\right)\right] \tag{10}$$

Outside boundary layer: $$\left[-\frac{k_{m_i}}{\delta_T}\left(C_i - C_i^*\right)\right] \tag{11}$$

where k_{m_i} represents mass transfer coefficient for each protein, δ_T the thermal boundary layer thickness and C_i^* the concentration of the protein in the first x position inside the thermal boundary layer. Calculation details of mass transfer coefficients and thermal boundary layer are presented in the work of Jun and Puri (2006).

Table 1. Kinetics of β-lactoglobulin denaturation reactions*

Reaction	Unfolding	Aggregation
k_{i_0} ($kg^{1-n}\ m^{3(n-1)}\ s^{-1}$)	3.42×10^{40}	3.25×10^{9}
E_i (kJ mol^{-1})	276.3	79.7
n_i	1.5	2

*: Bouvier and co-workers, 2014

Deposition rate of fouling is represented by the dimensionless Biot number. It is calculated by equation (12), where β=129 m² kg^{-1} (Georgiadis & Macchietto, 2000) represents an experimental coefficient, which depends on the flow configuration of the plate heat exchanger, and k_w=10^{-7} m s^{-1} is the mass transfer coefficient of the deposit on the wall.

$$\frac{\partial Bi(y)}{\partial t} = \beta k_w C_A(0,y) \tag{12}$$

Then, equation (13) gives the expression, in kg m^{-2}, of the mass deposits along the plates.

$$m(0,y) = \frac{\rho_d \, Bi(y) \lambda_d}{U_0} \tag{13}$$

where ρ_d = 1030 kg m^{-3} is the density of deposit, λ_d=0.5 W m^{-1} K^{-1} its thermal conductivity, and U_0 the overall heat transfer coefficient at clean conditions. The average mass of deposit on the wall is defined by equation (14), where W is the plate width.

$$\overline{M} = W \int_0^L M(y) dy \tag{14}$$

The mass of deposit on the right wall is the same as that deposited on the left one due to the symmetry prevailing in the channel.

1.3. Initial and boundary conditions

Milk arrives into the bottom of channel with constant temperature T_{in}=333 ºK and constant flow rate corresponding to Reynolds number of 2600. Concentration of native β-lactoglobulin protein is 5 kg m^{-3}. The diffusion flux in the direction normal to the outlet of channel is assumed to be zero for all variables.

The initial temperature of the walls is T_0=363 ºK. In order to take into account the effect of deposit on the thermal performance of plates, conditions imposed for energy and mass transfer are expressed by equations (15–17), e represents the channel thickness.

$$T(x=0,y) = T(x=e,y) = \frac{T_0}{1+Bi(y)} \tag{15}$$

$$C_N = C_U = 0 \tag{16}$$

$$\left.\frac{\partial C_A}{\partial x}\right|_{x=0} = \left.\frac{\partial C_A}{\partial x}\right|_{x=e} = \frac{k_w}{\delta_T} C_A \tag{17}$$

1.4. Numerical procedure

The equations presented in this model are a set of partial differential equations. In order to solve it, a computer program was written. It uses the finite volume method for spatial discretization with power law scheme. Establishment of pressure / velocity linkage is done with SIMPLE algorithm proposed by Patankar (1980). The implicit scheme was used for temporal discretization. The plate heat exchanger channel is assumed as a rectangular domain that consists of 80 x 160 nodes in staggered grid. The run of simulation has taken about 2 days of computing time on a Xeon server (2.53 GHz CPU, 6 Go RAM). Detailed procedure is described in Appendix A.

2. Results and discussion

2.1. Validation

The computational code was validated by comparing the amount of deposit on the wall obtained by simulation with the experimental data available in literature (Georgiadis & Macchietto, 2000). The data correspond to the first channel of plate heat exchanger, as milk enters in the channel at 333 ºK with a flow rate corresponding to Re = 2600 and walls maintained at constant temperature of 363 ºK. Figure 3 illustrates the evolution of average mass of deposit with time for both experimental data and simulation results. We have found that the simulation results are in good agreement with experimental values, where the maximal relative error does not exceed 0.66%. It can be seen that the amount of fouling increases linearly with time.

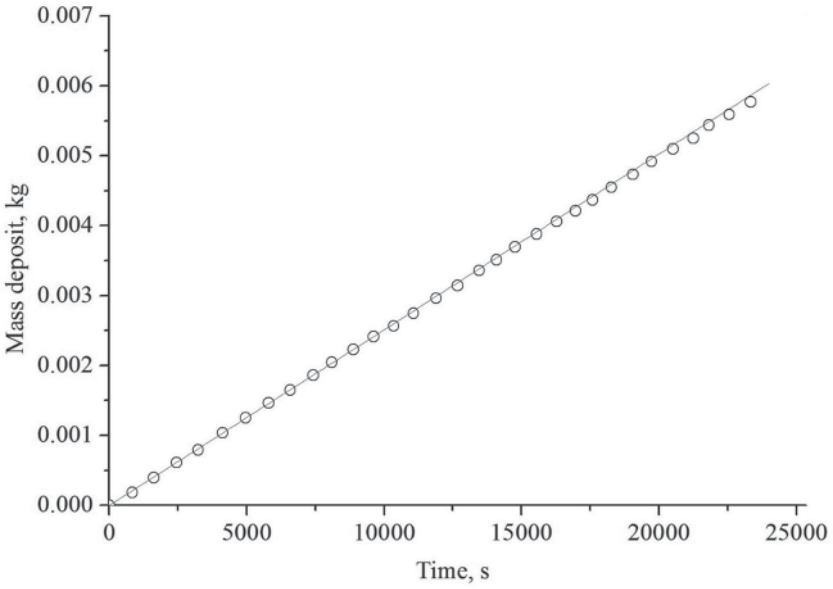

Fig. 3. Deposit profiles of simulation results (–) comparison with experimental data (o)

2.2. β-lactoglobulin behaviour

Figure 4 shows the distribution of β-lactoglobulin concentration in its three forms, native (A), unfolded (B), and aggregated (C), in half of the channel after 24 000 sec of milk treatment. The native protein concentrates in the bulk of fluid. The value of the concentration decreases towards the wall. As for aggregated protein, its concentration is higher near the wall. This is due to the fact that it adheres to the hot wall. Mass transfer between the thermal boundary layer and the bulk explains the concentration gradient of the native and aggregated forms of the protein in this zone of the channel. We have found that the aggregated protein concentration is higher at the top than at the bottom of the channel. This was also the case in the work of De Bonis & Ruocco (2009), where the deposit of fouling was mainly created in the outlet section of the channel. The increase in the concentration of aggregated protein is predictable,

since milk becomes hotter approaching the outlet of the channel. This is in good accord with previous works of Simmons and co-workers (2007) and Petit and co-workers (2013), saying that β-lactoglobulin denaturation increases at high temperature.

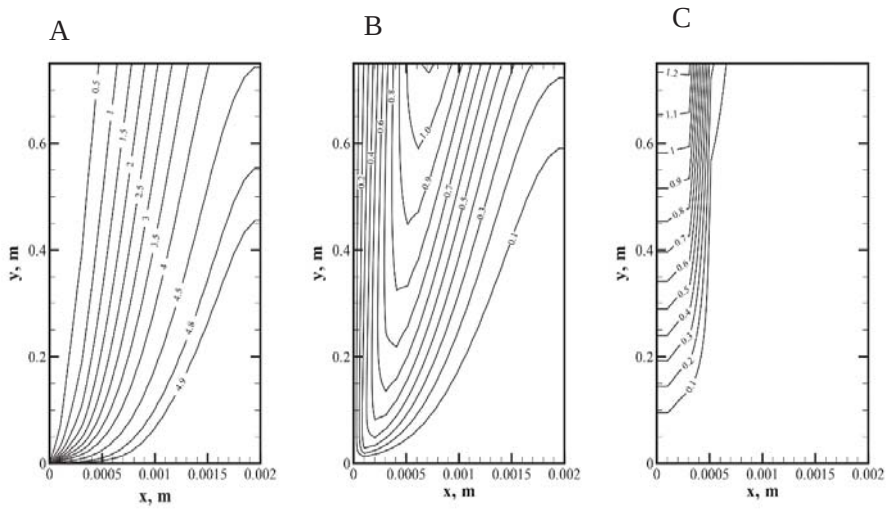

Fig. 4. Distribution of A: native; B: unfolded, and C: aggregated β-lactoglobulin protein concentration after 24 000 sec of operation for different values of milk inlet temperature

On the other hand, distribution of unfolded protein concentration is more complex, because this protein is a product of the unfolding reaction and target of the polymerization simultaneously. The concentration of this protein is negligible in the bulk of the fluid, where native protein is predominant, and near the wall, where the aggregate is deposited. Unfolded protein concentrates mainly around the interface between bulk of channel and thermal boundary layer. It is generated by mass transfer, which is dominant in this zone of the channel.

3. Conclusions

In this paper, we presented simulations of milk fouling in plate heat exchanger channel. Fluid flow, heat and mass transfer have been considered during milk heat treatment in order to study the concentration distribution of β-lactoglobulin over the channel.

The fouling model of Jun and Puri (2006) was used in this work and adapted to our calculation domain channel located between two plates of plate heat exchanger. The amount of deposit and heat transfer were coupled by using the Biot number in the expression of boundary condition. Numerical results of the amount of deposit were compared to experimental data in order to validate the model. Simulation results allowed validating the input data for the kinetic model of β-lactoglobulin denaturation. The native protein is concentrated in the bulk fluid unlike the aggregated form of the protein, which is present near hot walls and therefore adheres to it. However, unfolded form is focused around the interface between the thermal boundary layer and the bulk of fluid.

Finally, a three-dimensional study could allow a better understanding of the exact behaviour of fouling mechanisms, since it allows taking into account the corrugated form of the plates and also the plate ports for the milk inlet and outlet. The effects of turbulence neglected in this study so far will need to be considered in the further works.

Appendix A: Numerical procedure

The differential equations described in previous sections are resolved numerically by a calculation code developed in Fortran 90 language by our research team. This code uses finite volume method with a power law scheme for the transformation of differential equations to systems of algebraic equations. A staggered grid and the SIMPLE algorithm provide the pressure / velocity coupling. Algorithm steps are presented as follows:

1. Initialize pressure field;
2. Resolve the momentum equations to obtain U and V fields;
3. Resolve continuity equation to correct pressure;
4. Correct velocity field;
5. Return to step 1 until convergence.

Coupling between temperature distribution and fouling amount on walls is accomplished by the boundary condition imposed for temperature by equation (15).

The systems of equations obtained by discretization are solved via the line-by-line method, which is a semi-iterative method combining the direct method TDMA and the iterative one of Gauss Seidel.

Steps of the calculation procedure performed using the calculation code:

1. Initialize all variables and set physical properties values;
2. Introduce the time loop;
3. Resolve momentum equations;
4. Resolve continuity equations;
5. Correct velocity and pressure fields;
6. Resolve energy equation;
7. Resolve concentration equations of native, unfolded, and aggregated proteins;
8. Calculate fouling mass accumulated on walls;
9. Repeat steps (3) to (8) until convergence;
10. Use obtained results as initial variables and return to step (2);
11. The time loop is repeated until reaching the final time.

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