EXPERIMENTAL AND COMPUTATIONAL ANALYSIS OF REDUCTION IN FOULING BY LOW ENERGY SURFACES

A Dissertation in
Agricultural and Biological Engineering
by
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ABSTRACT

Pasteurization is essential to ensure the microbial safety and to increase the shelf life of food products such as dairy products and fruit juices. Thermal instability of fluid milk, focus of this research, causes formation of solid fouling deposits, as a result of specific reactions of milk components, on the contact surfaces of the heat transfer equipment resulting in: (1) reduction of heat transfer coefficient, (2) increase in pressure drop, (3) higher risk for microbial growth, and (4) adverse effect on product quality. Due to economic, microbial safety and quality concerns, and environmental problems caused by fouling of heat exchangers, mitigating fouling during the operation is desirable. Fouling is also a widely pervasive issue in several other industries such as automotive, aerospace, chemical, petroleum, and waste water treatment. Therefore, the breadth of industries affected by fouling, served as the motivation of this research.

Accordingly, the goal of this work was to develop a computational model which estimates the amount and rate of fouling on the contact surfaces of heat transfer equipment under various operating conditions. Towards this purpose, first a bench-scale test set-up that sufficiently represented the thermo-hydraulic behavior in industrial plate heat exchangers was designed and fabricated. Next, stainless steel SS316 (control) and four coated surfaces Microlube/PTFE, TM117P, AMC148, and CNT were tested to evaluate their fouling behavior. In the computational model, the same test set-up was modeled with FLUENT to obtain the temperature profile in the flow channel of the bench scale tester. The calculated temperature profiles formed the basis for determining the change in thermal performance of the system with time due to fouling. Then with this data, the deposit amount on the test surface was calculated based on the effect of additional conductive heat transfer resistance caused by foulants deposits on the surface. Finally, the effect of surface characteristic, particularly the contact angle, of the test surfaces, on the rate and amount of foulants deposits were determined by introducing a semi-empirical correlation.

Fouling behavior on the test surfaces was analyzed experimentally and computationally for four different test conditions; two different milk flow rates, 3 g/s and 10 g/s, and two different inlet milk temperatures, 40°C and 60°C. Experimental results showed that there was up to 50% less deposit formation when the inlet milk temperature was 60°C compared to when it was 40°C. Results did not provide any definitive trend of deposit amount as a function of flow rate.

A relationship between the contact angle of a liquid on a solid surface that is a major indicator of fouling tendency of the surface, and fouling amount for the test surfaces was found based on experimental results. When the contact angle of the liquid on the solid surface is large, the wettability of the surface is less and thus the surface is less prone to fouling. In the case of AMC148 and CNT coated surfaces, which have relatively higher contact angles (145° and 180°, respectively) compared to the contact
angle of the SS316 control surface (72°), the fouling amount was reduced approximately by 90%. Whereas, Teflon® based coated surfaces, Microlube/PTFE and TM117P (contact angles are 107° and 112°, respectively) did not reduce the fouling amount as much as AMC148 and CNT coated surfaces; that is only 20 to 70% depending on the operating conditions. The subpar performance of Teflon® based surfaces was attributed to insufficient improvement of the wettability of these surfaces.

The results of the computational model for each test case indicate that there is a good agreement with the measurements for control and Teflon® based coated surfaces; the average percent difference between the measured and calculated deposit amount for four test conditions was from 11.1% to 24.5%. In contrast, the average percent differences are the largest for the surfaces that are least prone to fouling (AMC148 and CNT, -57.1% and -68.2%, respectively). In other words, for the surfaces that have relatively higher contact angles the calculated and experimental fouling amount differ more. This is most likely due to the insufficiency of the semi-empirical correlation between the contact angle and deposit amount that was obtained by using limited number of test surfaces.

In this study, a computational model was proposed and validated to estimate the amount of foulants deposits that can be used to improve the thermal performance of the heat exchangers. The results of this study for bench scale tester show that the proposed computational model can be used to predict the fouling amount on any surface with known contact angle at different operating conditions with less experimental work. Furthermore, for the milk industry AMC148 coated surface is recommended for further investigation since it gave the most promising results in fouling mitigation. The same methodology can be extended to other industries where fouling is a major problem.

Keywords: Fouling, milk pasteurization, protein denaturation, beta lactoglobulin, plate heat exchanger, FLUENT, contact angle, wettability
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Dedicated to my mother…
Chapter 1

INTRODUCTION

Heat treatment is a very common and one of the most important operations in the dairy industry. It is done to ensure the microbial safety and to increase the shelf life of the end product.

Milk and milk-based products are subjected to various heat treatment operations. Heat treatment is mainly of two types; direct and indirect. In direct heating, the milk is brought into contact with the heating medium, steam. In indirect heating, milk and the heating medium are separated by a conducting material in the form of heat exchangers, and heat transfer is by conduction and convection.

Heat exchangers are devices where two moving fluid streams exchange heat without mixing. Plate heat exchanger (PHE), a very common type of heat exchanger in diverse industries, consists of a pack of gasketed and corrugated metal plates, pressed together in a frame. Through a series of interconnected parallel-to-each-other and narrow flow channels, fluid flows alternately and heat exchange takes place through highly conductive very thin metal plates. PHEs are widely used in the food industry involving liquid-liquid heat transfer because of their compactness, easy maintenance, high thermal performance, and low heat loss. However, PHEs are particularly prone to fouling because of their small hydraulic diameter.

The fluid milk is thermally unstable. Consequently, during heat treatment, solid fouling deposits form, as a result of specific reactions of milk components, on the contact surfaces of the heat transfer equipment. These typical undesired reactions: (1) reduce the heat transfer coefficient, (2) increase the pressure drop, (3) promote the microbial growth, (4) compromise safety and quality of the product, and (5) increase product losses that result in higher operating cost. Operating cost of fouling in early 1990s pasteurized milk production in US was estimated to be approximately $140 millions per year (Sandu and
Singh, 1991). In addition to the economic and microbiological impacts, cleaning of fouled equipment with aggressive cleaning agents and excessive use of water is of environmental concern. For all these reasons, the problem of fouling in a dairy plant needs to be understood and minimized for more efficient and economic processing.

Deposit formation in milk involves at least two processes: chemical reaction fouling and crystallization fouling (Prakash et al., 2005). Chemical reaction fouling is due to denaturation of proteins, particularly β-lactoglobulin, major constituent of whey protein in fluid milk. Adsorption of whey proteins even occurs at low temperatures but increases and becomes significant at higher temperatures. In pasteurization of milk, protein aggregates are formed at temperature of around 80°C and higher. Fouling as a result of denaturation of whey proteins is dominant but there are other components of fouling deposits such as salts, mainly calcium phosphate, and fats. Crystallization fouling of calcium phosphate is due to its solubility being exceeded at high temperature. Although the deposits are mainly composed of protein and salts, it has been established that first layer is always proteinaceous and presence of this protein layer on the surface accelerates the fouling (Visser and Jeurnink, 1997). For this reason, most of the studies in fouling of heat exchangers in milk pasteurization have been focused on factors affecting the denaturation of β-lactoglobulin and adhesion to surface.

A number of authors have modeled milk fouling in PHEs based on a simple representation of the process hydrodynamics. The thermal simulation of plate heat exchangers have been performed based on plug flow assumption inside channels and an equal distribution of fluid into the channels from inlet.

Flow rate, surface and inlet stream temperatures are the operational factors affecting fouling behavior. Surface temperature becomes important in fouling due to dependence of the denaturation of proteins on temperature. Hiddink et al. (1986) stated that surface becomes prone to fouling when the difference between surface temperature and fluid temperature exceeds 10-15°C. Inlet stream velocity affects fouling in two ways: (1) when velocity of the stream is high, rate of fouling decreases due to increase in shear forces, (2) as fluid flows faster between the plates of heat exchanger length of the plate
prone to fouling increases due to undeveloped thermal behavior. Therefore, hydrodynamic and thermodynamic modeling in heat exchangers has been studied extensively. There have been 1D, 2D, and 3D models describing the dynamic behavior in heat exchangers (Jun and Puri, 2005\textsuperscript{a}). Accurate prediction and analysis of fouling dynamics based on understanding of the chemistry and fluid dynamics are useful in predicting how real process equipment is likely to respond (Jun and Puri, 2005\textsuperscript{b}).

Investigation of anti-fouling methods has received considerable attention. A variety of chemical and mechanical methods have been suggested to reduce the deposit formation on the equipment surfaces. For PHEs, chemical mitigation is one of the possible ways. Unfortunately, using chemical additives, especially in food industry, may cause undesired product contamination. The use of magnetic, electric, radiation or catalytic treatments, as Donaldson and Grimes (1988) stated, have not yet given consistent and reliable results. Modifying the surface properties of the heating surfaces has been suggested recently for finding a solution. Several novel techniques such as direct ion implantation, magnetron sputtering, dynamic mixing, plasma enhanced vapor deposition, and surface coating have been considered for this purpose.

Experimental results have shown that the free surface energy plays a predominant role and the roughness a minor role in the level of fouling and cleaning efficiency. In addition, damaged surfaces and the conditioning film also influenced the surface properties.

Many factors play a critical role in fouling process which eventually causes inefficient processing, high cost, and aggressive cleaning requirements. For all these reasons, the problem of fouling in a dairy plant needs to be understood and minimized for more efficient and economic processing. To be able to achieve this goal, the mechanism of fouling and the factors affecting the formation of fouling deposits should be understood well. Consequently, modeling of thermal behavior in a plate heat exchanger supported with experimental data and coupled with surface study forms the scope of this study.
Chapter 2

OBJECTIVES

The cost of fouling in heat exchangers and the problems caused by fouling were discussed in the previous chapter. There have been studies to model fouling, however, not sufficient research has been conducted for mitigating fouling. This study aims to minimize fouling in milk pasteurization unit, plate heat exchanger. The goal of the proposed research was to understand and mitigate fouling in plate heat exchangers. Towards this end, a detailed study of bench scale, plate type heat exchanger was performed. Rationale for using bench scale tester includes: (1) rapid evaluation of surfaces compared to pilot scale plate heat exchanger, (2) cost and effectiveness, (3) quick assembly and disassembly, and (4) portability.

Specific objectives leading to this goal were to:

Objective 1: analyze the dynamics of bench scale plate heat exchanger experimentally and numerically,

Objective 2: characterize differently treated surfaces,

Objective 3: examine the effect of surface properties on milk fouling,

Objective 4: correlate surface properties with adhesion of deposits, and

Objective 5: develop a computational model including surface properties, fouling kinetics, and PHE dynamics that describes milk fouling in bench scale plate heat exchanger.
Chapter 3

LITERATURE REVIEW

This chapter includes information about milk and currently available literature on fouling dynamics. In addition, hydrodynamic and thermal models that describe the fluid flow and heat transfer in plate heat exchangers, effect of surface properties and other factors affecting fouling are summarized.

Fouling can be defined as the accumulation of undesirable substances on a surface. In general, fouling is classified according to the principal process as particulate, crystallization, corrosion, bio-fouling, and chemical reaction. In this context, the term refers to the undesirable deposits formed because of thermal instability of food products on the heat exchanger surface resulting in both technical and economical problems; higher heat transfer resistance, increase in pressure drop, and possible areas for microbial growth. Fouling is very severe in the food industry and needs cleaning more frequently compared to other industries (Fryer and Belmar-Beiny, 1991). Therefore, cleaning cost, production losses and environmental issues resulting from excessive use of water and aggressive chemicals are of concern too. Consequently, due to aforementioned reasons, it is beneficial to minimize this very serious problem.

As mentioned in Chapter 1, fouling needs to be understood together with the hydrodynamic and thermodynamic patterns of flow stream in the heat exchangers. Synergistic interactions between the physicochemical, hydro- and thermodynamic fundamentals need to be included to have an accurate prediction and analysis of fouling dynamics. The purpose of fouling models is to enable an assessment of the impact of fouling on heat exchanger performance.

3.1 Milk

Milk is secreted as a complex mixture of water, carbohydrate, lipid, protein, minerals, and vitamins by all female mammals to supply nutrition and immunological protection to the young. The interspecies variation in the quantitative composition is due
to differences in the metabolic process and nutritional and physiological requirements among species.

In the food industry, the term milk is used for cow’s milk. If used otherwise it is specified as goat milk, sheep milk, or buffalo milk (Spreer, 1998; Wong, 1988; Harper and Hall, 1976). The composition of milk is given in Table 3.1 (Wong, 1988).

Table 3.1  Composition of milk

<table>
<thead>
<tr>
<th>Species</th>
<th>% Total solids</th>
<th>% Fat</th>
<th>% Protein</th>
<th>% Lactose</th>
<th>% Ash</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cow</td>
<td>12.6</td>
<td>3.7</td>
<td>3.4</td>
<td>4.8</td>
<td>0.7</td>
</tr>
</tbody>
</table>

3.1.1 Milk Proteins

The proteins of milk can be divided into two groups: the caseins and the whey proteins. The approximate milk protein concentrations are given in Table 3.2. The difference between these protein groups is that caseins are heat sensitive and precipitate upon acidification; whereas, the whey proteins in their native form are heat sensitive, more soluble in water and do not precipitate at their isoelectric pH (Hui, 1993; Wong, 1988).

There is a link between fouling and heat denaturation of whey proteins when the processing temperature is higher than 70°C, the temperature at which whey proteins undergo irreversible changes (Visser and Jeurnink, 1997). The role of caseins in fouling process is less clear.

3.1.1.1 Casein

The main constituent of milk proteins is casein with approximately 80% of total milk protein (Table 3.2). Casein is present in the form of micelles or particles of macromolecular sizes. Casein proteins, made up of number of components, α_{S1}-casein, α_{S2}-casein, β-casein, κ-casein, and γ-casein, together forming complex particles, have very little secondary structure (Wong, 1988; Hui, 1993).
3.1.1.2 Whey Proteins

The whey protein fraction, approximately 17% of milk protein (Table 3.2), comprises noncasein proteins that remain in the milk after precipitation of casein at pH 4.6. Whey proteins are more stable to acid than casein but very sensitive to heat. In addition to solubility dissimilarity between casein and whey proteins, unlike casein, whey proteins have more organized secondary and chemical structure (Wong, 1988).

The main fractions of whey proteins are β-lactoglobulins, α-lactalbumins, bovine serum albumin (BSA), and immunoglobulins (Ig). β-lactoglobulin is quantitatively the most dominant protein. For this reason and also due to its more heat sensitivity compared to α-lactalbumin, fouling studies are focused mainly on the behavior of this protein (Visser and Jeurnink, 1997).

Table 3.2 Distribution of milk proteins

<table>
<thead>
<tr>
<th>Component</th>
<th>% Total protein</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Caseins</strong></td>
<td>≈ 83</td>
</tr>
<tr>
<td>αS1-casein</td>
<td>36</td>
</tr>
<tr>
<td>αS2-casein</td>
<td>9</td>
</tr>
<tr>
<td>β-casein</td>
<td>21</td>
</tr>
<tr>
<td>κ-casein</td>
<td>12</td>
</tr>
<tr>
<td>γ-casein</td>
<td>4</td>
</tr>
<tr>
<td><strong>Whey</strong></td>
<td>≈ 17</td>
</tr>
<tr>
<td>β-lactoglobulin</td>
<td>10</td>
</tr>
<tr>
<td>α-lactalbumin</td>
<td>2</td>
</tr>
<tr>
<td>Immunoglobulin</td>
<td>2</td>
</tr>
<tr>
<td>Serum Albumin</td>
<td>1</td>
</tr>
<tr>
<td>Minor proteins</td>
<td>2</td>
</tr>
</tbody>
</table>
β-lactoglobulin: A major component of whey protein, β-lactoglobulin, shown in Figure 3.1 (Brownlow et al., 1997), exists in colloidal form in milk. It is composed of 162 amino acids. Though there are eight genetic variants, A, B, C, D, Dr, E, F, and G; variants A and B prevail in most breeds (Hui, 1993). Two monomers, each approximately 18,000 molecular weight, are linked to form a dimer by forming intramolecular disulfide bridges. Free thiol group is unreactive in the native protein. When the protein is partially unfolded, for instance by heat treatment at near-neutral pH, one thiol group per β-lactoglobulin molecule becomes available for reaction (Visser and Jeurnink, 1997; Hui, 1993).

β-lactoglobulin is composed of 162 amino acid residues including two disulfide bonds (Cys66–Cys160 and Cys109–Cys119) and one free cysteine (Cys121). It is documented that β-lactoglobulin irreversibly denaturates as a result of mainly the formation of intermolecular and nonnative intramolecular disulfide bonds initiated by a single sulfhydryl group during unfolding (Burova et al., 1998).

![Figure 3.1 Schematic representation of the native structure of bovine β-lactoglobulin](http://www.ifr.ac.uk/Science/programme/F1/projects.html) (with permission)

Under physiological conditions, β-lactoglobulin exists as a dimer consisting of anti-parallel β-sheets formed by nine strands labeled A to I and one α-helix. Each monomer has two disulfide bonds and one free sulfhydryl group buried within the protein structure on the β-strand H. At neutral pH, upon heat treatment, the native dimers dissociate into native monomers. Then, reactive monomer with a partially disrupted helix
is formed leading to the exposure of free sulfhydryl group and hydrophobic amino acids. These reactive species undergo intermolecular sulfhydryl/disulfide interchange to give non-native dimers and larger species by thiol catalyzed disulfide bond interchange and the involvement of non-covalent interactions depending on the salt concentration of the environment (Croguennec et al. 2003).

The protein fouling is dependent on the reactions involving –SH and S-S groups. When temperature increases, first, the three-dimensional structure held together by intramolecular disulphide bridges, ionic interactions, and van der Waals forces of the protein is altered. This stage, reversible upon cooling, is followed by unfolding of the protein. As the secondary and tertiary structure of the protein unfolds, free thiol groups are exposed and thus more of the inner structure can react with other molecules. Consequently, insoluble mass of protein is formed irreversibly. As a result of accumulation of these aggregates, deposits are formed on the heating surface.

3.1.2 Properties of Milk

Knowing the chemical and physical properties of milk is critical to understanding the dairy processing and associated problems. Milk is a complex system; a dilute emulsion, colloidal dispersion, and solution. For this reason, the physical properties of milk, though very similar to its major constituent, water, depend on compositional and structural factors.

3.1.2.1 Density

The density of liquid mixtures is the average of the densities of each component. Thus, the density of milk depends on concentrations of components. Since degree of hydration of proteins and specific volume of milk fat are functions of temperature, density of milk changes with temperature.

3.1.2.2 Viscosity

Viscosity can be simply defined as the resistance of a liquid to flow. Rheological behavior of dairy products depends on state of dispersion of the solid components and
thus can be Newtonian or non-Newtonian. Milk does not differ much from Newtonian behavior. Temperature and composition of milk affects its viscosity.

### 3.1.2.3 Heat Capacity

Heat capacity of milk is inversely related with fat content at a given temperature. From 15 to 40°C, heat capacity decreases and above 40°C it gradually increases. The specific heat variation is not very significant with temperature variation.

### 3.1.2.4 Thermal Conductivity

Thermal conductivity of milk, at temperatures above the freezing point, is inversely related with fat content, and increases with temperature. The average physical properties of milk at room temperature are given in Table 3.3. For comparison, the corresponding properties of water are given also in this table.

<table>
<thead>
<tr>
<th>Table 3.3 Physical properties of milk and water</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal conductivity (W/m.K)</td>
</tr>
<tr>
<td>Milk</td>
</tr>
<tr>
<td>Water</td>
</tr>
</tbody>
</table>

Nema and Datta (2006) gave the properties of milk as a function of temperature. These are presented as in Table 3.4.

<table>
<thead>
<tr>
<th>Table 3.4 Physical properties of milk as a function of temperature (in K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal conductivity (W/m.K)</td>
</tr>
<tr>
<td>0.00133T+0.539911</td>
</tr>
</tbody>
</table>

### 3.1.3 Thermal Processing of Milk

In dairy plants, pasteurization of milk is an important process. Different types of equipment are used for this purpose. However, the most common one is the plate heat exchangers.
To ensure destruction of pathogenic microorganisms in the milk, time and temperature combinations of the pasteurization process are regulated. To ensure safe product, thermal lethality of the microorganisms that are likely to be present in milk are determined. Tolerance of each organism to heat is different. During thermal treatment, rate of destruction of microorganisms changes logarithmically and depends both on the process temperatures and time. The extent of the pasteurization treatment required is determined by the heat resistance of the most heat-resistant enzyme or microorganism in the specified food. In the case of milk pasteurization, *Mycobacterium tuberculosis* and *Coxiella burnetti* are the key bacteria that the required time temperature relationship is based on. Accordingly, at 63°C process shall not be less than 30 minutes, or milk should be kept at 72°C for not less than 15 seconds (FDA, 2004).

### 3.2 Plate Heat Exchanger

Compact heat exchangers are found in almost every field of engineering increasingly. As mentioned previously, two types of heat exchangers exist based on their heating approach; direct and indirect. In direct heating, the fluid is heated in a heated tank; whereas, in the case of indirect heating, two moving fluid streams exchange heat without mixing. Plate heat exchanger (PHE), very common type of heat exchangers used in various industries, consists of a pack of gasketed and corrugated metal plates, pressed together in a frame. Narrow flow channels are interconnected, and the fluid flows through these parallel channels alternately. Heat exchange takes place between the hot fluid on one side and the cold fluid on the other side of the highly conductive very thin metal plates. An example of a gasketed plate-and-frame heat exchanger for industrial applications is shown in Figure 3.2.

The number of plates, their corrugation, the type and position of the gaskets and the location of inlet and outlet ports characterize the equipment configuration. This is important as it affects the flow profile within the channels. In plate and frame heat exchangers, the corrugated plates are the elementary components. The metal plates, usually of stainless steel, are closely packed in a frame so that the orientation of the corrugation between adjacent plates is non-aligned. This configuration improves heat
transfer by providing a larger effective surface area and by generating a complex flow structure in the patterns between the plates. Corrugations also strengthen the plates and give a high structural stability due to the multitude of contact points. The corrugation determines the thermal-hydraulic performance of the heat exchanger.

![Plate heat exchanger diagram](www.genemco.com) (with permission)

### 3.3 Fouling Dynamics and Models

Milk processing is an important example of fouling in food industry. Whey proteins, mainly β-lactoglobulin, and calcium phosphate are involved in the fouling process through interaction mechanism. β-lactoglobulin fouls after denaturation and aggregation while calcium phosphate fouls as a result of supersaturation (Changani et al., 1997). On cold surfaces, fat and protein dominate milk deposits, while amount of minerals is minimal. As temperature increases, mineral content in the deposits increases and fat content decreases. Deposits formed at bulk temperatures at around 70-110°C contain 50-60% protein and 30-35% minerals (Karlsson et al., 1996). Fouling starts as soon as milk is brought into contact with a surface at a molecular level and invisible to
the naked eye with whey protein adhesion. Upon heating, the whey proteins start to unfold and expose a free S-OH group, changing into an activated state that allows attachment to the protein layer initially formed (Visser and Jeurnink, 1997). For this reason, properties of the surfaces are very important. If the formation of first layer is delayed, fouling can be reduced.

3.3.1 Deposit Composition

Burton (1968) described two types of fouling in terms of composition and structure. Type A has white, soft like spongy and voluminous structure. It starts to form at temperatures above 75°C and is highest between 95 and 110°C. The composition is protein rich with 50-70%, and concentration of minerals in the deposit is around 30-40%. The deposit that forms in pasteurization is of this type. Type B deposits are formed at temperatures above 120°C, due to high mineral content, as high as 70-80%. Generally, type B deposit is hard, granular, and brittle.

3.3.2 Denaturation Kinetics of β-lactoglobulin

Mathematical models for prediction of product properties are very helpful in process optimization. Generally, the models that are used in food processing are process models, kinetic models, and predictive models for cost estimation.

Of concern, herein are kinetic models that describe, for instance, the denaturation and aggregation of proteins, the inactivation of enzymes, contamination and formation of reaction products. de Wit and Klarenbeek (1989) suggested the denaturation of β-lactoglobulin as a consecutive reaction of unfolding and aggregation.

\[ N \rightarrow U \rightarrow A \]

(3.1)

where N, U, and A are the native, the unfolded, and the aggregated β-lactoglobulin, respectively. The equations that describe kinetics of the reaction, i.e., the rates of disappearance and formation are given by:

\[ -\frac{dC_N}{dt} = k_U C_N^U \]

(3.2)
\[
\frac{dC_U}{dt} = k_U C_N^p - k_A C_U^q
\]

(3.3)

\[
\frac{dC_A}{dt} = k_A C_U^q
\]

(3.4)

where \(C_N\), \(C_U\), and \(C_A\) are the concentrations of native, unfolded, and aggregated \(\beta\)-lactoglobulin, respectively. The reaction orders of unfolding native protein and aggregated unfolded protein are \(p\) and \(q\). The reaction rate constants \(k_U\) and \(k_A\) are related to the absolute temperature according to the Arrhenius relation:

\[
k = k_0 \exp\left(-\frac{E_a}{RT}\right)
\]

(3.5)

where \(k_0\) is the pre-exponential factor and \(E_a\) is the activation energy of the reaction.

Based on experimental data, the kinetic data for unfolding and aggregation of \(\beta\)-lactoglobulin is given in Table 3.5.

Table 3.5 Kinetic data of \(\beta\)-lactoglobulin

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Temperature (°C)</th>
<th>Reaction Order</th>
<th>(E_a) (kJ/mol)</th>
<th>(\ln k_0)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unfolding</td>
<td>70-90</td>
<td>(p=1)</td>
<td>261</td>
<td>86.41</td>
<td>de Wit and Klarenbeek (1989)</td>
</tr>
<tr>
<td>Aggregation</td>
<td>70-90</td>
<td>(q=2)</td>
<td>312</td>
<td>99.32</td>
<td>de Wit and Klarenbeek (1989)</td>
</tr>
<tr>
<td>Aggregation</td>
<td>90-150</td>
<td>(q=2)</td>
<td>56</td>
<td>14.42</td>
<td>Dannenberg and Kessler (1988)</td>
</tr>
</tbody>
</table>

### 3.3.3 Fouling Kinetics

Local fouling is heterogeneous adsorption of milk constituents at the surface governed by mass transfer and reaction in series. The aggregated protein diffuses through boundary layer and adheres to surface. Concentration profile describing the fouling process is shown as in Figure 3.3 (de Jong et al., 1992).

Mass transfer through boundary layer:

\[
J_F = \frac{D_F}{x_{blg} \delta} \left(\bar{C}_U - C_{Uli}\right)
\]

(3.6)

where \(\delta\) is the boundary layer thickness, \(J_F\) is diffusion rate, \(D_F\) is diffusivity of foulant, \(x_{blg}\) is the fraction \(\beta\)-lactoglobulin in the deposit. \(C_{Uli}\) is the local concentration of
unfolded β-lactoglobulin near the surface in equilibrium with the concentration $\overline{C}_U$ in the deposit layer at the interface.

Reaction rate equation is:

$$R_F^* = -k'' C_{Ui}^n$$  \hspace{1cm} (3.7)

where $n$ is the reaction rate, and $k''$ is the rate constant.

Reaction occurring at the surface is transported through, therefore the mass balance equation is:

$$J_F = -R_F^*$$  \hspace{1cm} (3.8)

$$C_{Ui} = \left( \frac{J_F}{k''} \right)^{1/n}$$  \hspace{1cm} (3.9)

$$J_F = \frac{D_F}{x_{bg}\delta} \left( \overline{C}_U - \left( \frac{J_F}{k''} \right)^{1/n} \right)$$  \hspace{1cm} (3.10)

In order to control fouling problem, straightforward approach is first to develop a model based on physics and chemistry of fouling mechanism. However, since milk is not a well defined system, exact mathematical description of the problem is not straightforward. Thus, models based on kinetics of fouling are useful (de Jong, 1997).

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{concentration_profile.png}
\caption{Concentration profile for deposit formation (de Jong et al., 1992)}
\end{figure}
There have been a number of studies on the mechanics and kinetics of fouling (Grijspeerdt et al., 2003). There seems to be an agreement that the key role in fouling of dairy products is played by proteins and especially β-lactoglobulin, at least when the temperature is below 90°C. The heat denaturation of this whey protein governs the milk deposit formation on the heat transfer area. As temperature increases, depending on the pH, mineral precipitation, specifically calcium phosphate, gradually becomes more dominant.

3.3.4 Fouling Dynamics

The exact mechanism of fouling is not known due to complexity of reactions and dependence on so many variables, such as equipment design, operating condition affecting the transfer in the boundary layer and adhesion of deposits (Changani et al., 1997). In addition to external physical factors playing crucial role in deposit formation, the chemistry, physicochemical properties of the milk and its interaction with the equipment surface must be considered.

3.3.5 Fouling Models

Epstein (1981) stated the fouling steps are divided into initiation, transport, attachment, removal, and aging. Overall, result of these processes is the net deposition of material on the heat transfer surface. Thus, fouling is a transient process. In other words, it is time dependent; the heat transfer equipment is clean at the beginning and becomes fouled in time. After the induction period, depending upon the deposition and removal rates fouling curve has its shape when the difference between deposition and removal rate is constant, the fouling resistance curve will be a straight line. In this case, unless the cleaning is employed fouling will continue to increase. Asymptotic curve is obtained if the attachment rate is constant and the removal rate is proportional to the fouling thickness. Falling rate, when the deposition rate is inversely proportional to the deposit thickness, is between linear and asymptotic curves (Figure 3.4). In food processing, usually linear or falling rate fouling takes place (Changani et al., 1997).
In the induction period, the conditions do not change significantly. Thus, it is usually ignored in the modeling of fouling and assumed that fouling period begins as soon as fluid flows through the heat exchanger.

Mass transfer between the bulk and thermal boundary layers takes place for each protein. Only the aggregated protein is deposited on the wall. The deposition rate is proportional to the concentration of aggregated protein in the thermal boundary layer. The fouling resistance to heat transfer is proportional to the thickness of the deposit.

The slowest step is the controlling step of the overall rate of fouling. If fouling is mass transfer controlled, then deposition would not be expected to be a strong function of temperature. However, if fouling is reaction controlled, deposit formation will be a function of wall or bulk temperature, depending on the position of the controlling reaction. A more complete picture of milk fouling might thus be given by considering separately the contribution of both surface and bulk reactions to solids deposition.

According to Bott (1995), not much research has been done about the problem of surface fouling in heat exchangers. However, currently mitigating fouling in heat transfer equipment is very important in many industries since energy is often a factor in the economics of a particular process. Understanding the mechanism of fouling that causes accumulation of deposits on the surfaces of the equipment resulting in heat transfer efficiency is the key step to suggest ways for reducing or eliminating this problem. Three basic stages in relation to deposition on surfaces from a moving fluid are:

1. The diffusional transport of the foulant across the boundary layers adjacent to the solid surface within the flowing fluid.
2. The adhesion of the deposit to the surface and to itself.
3. The transport of material away from the surface.

Sum of these stages determines the growth of the deposit on the surface and may be called deposit growth. Figure 3.4 shows idealized asymptotic graph for the rate of deposit growth on a surface.
In region A in Figure 3.4, induction or conditioning period, adhesion starts. In region B, the process where there is competition between deposition and removal, deposit on the surface grows steadily. When the rate of removal and the rate of deposition become equal as in region C, the deposit thickness remains almost constant.

The ability to transfer heat efficiently is the central feature of heat transfer processes. For that reason, there have been numerous research publications focused on understanding the heat transfer mechanisms better and development of suitable correlations and techniques that may describe the system better and be applied to the design of heat exchangers. The accumulation of deposits on the surfaces of heat exchangers increases the resistance to heat transfer.

**3.3.6 General Models of Fouling**

As previously stated, rate of build-up of deposit on a surface can be defined by the difference between deposition rate and removal rate. The mass balance across fluid/solid interface is:
\[ \text{Accumulation} = \text{Input} - \text{Output} \] (3.11)

\[ \frac{dm}{dt} = \phi_D - \phi_R \] (3.12)

where \( m \) is mass of the deposit, \( \phi_D \) and \( \phi_R \) are the deposition and removal mass rates, respectively.

As mentioned earlier, the deposit layer increases the resistance to heat transfer. Besides this undesired effect of fouling, there is a condition when the deposit is appreciably thick; the area for fluid flow is reduced hence for the same volumetric flow, the fluid velocity increases. In addition, the foulant layer on the surface changes the surface roughness. Roughness of the deposit surface will be greater than that of heat exchanger surface. This will produce higher turbulence particularly near the surface. Consequently, change in heat transfer coefficient due to fouling can be expressed as the sum of these effects; change due to thermal resistance of deposit layer, change due to higher roughness of foulant, and lastly change due to change in Reynolds number.

Idealized picture of the development of a deposit on the surface with time is shown in Figure 3.5. During fouling, two distinct time periods can be identified; induction and fouling periods.

Fouling is a transient process. There may be an induction period during which conditions do not change significantly, followed by a fouling period during which the heat transfer coefficient decreases and the pressure drop increases. Most mathematical models that have been developed ignore the induction period. In other words, they assume fouling begins as soon as fluid flows through the heat exchanger. Therefore, most attempts have been made to develop the generalized models for the fouling period (Bott 1995). The fouling period consists of a deposition and a removal process. The rate of buildup of deposit on a surface could be defined by the simple concept of the difference between the rates of deposition and removal. This is discussed in the following paragraphs.
3.3.6.1 Fouling Period

During the fouling period, both deposition and removal takes place. Denatured protein deposits on the wall and swept away simultaneously.

3.3.6.1.1 A General Model

Simplest model for fouling is shown as linear fouling in Figure 3.5. If the induction period is neglected, the deposit thickness $x_f$ at time $t$ can be estimated using:

$$x_f = \frac{dx_f}{dt} t$$

(3.13)

where $\frac{dx_f}{dt}$ is the rate of increase of deposit thickness

When induction period ($t_i$) is included in the deposit thickness equation

$$x_f = \frac{dx_f}{dt} (t - t_i)$$

(3.14)

Fouling thermal resistance at time $t$, $R_{ft}$, can be calculated to be:
\[
R_f = \frac{x_f}{\lambda_f} = \frac{dR_f}{dt} (t - t_i)
\] (3.15)

where \(x_f\) is the fouling layer thickness at time \(t\), \(\lambda_f\) is the thermal conductivity of the deposit, and \(R_f\) is extra resistance to heat transfer due to deposit formation.

### 3.3.6.1.2 Asymptotic Fouling

The model developed by Kern and Seaton (1959) is essentially a mathematical interpretation of the asymptotic fouling curve neglecting the induction period in Figure 3.4, fouling thermal resistance at time \(t\) is;

\[
R_f = R_{\infty} \left(1 - e^{\beta t}\right)
\] (3.16)

where \(R_{\infty}\) is the asymptotic value of fouling thermal resistance at infinite time, and \(\beta\) is a constant that depends on system properties.

### 3.3.6.1.3 Falling Rate Fouling

The behavior of \(\beta\)-lactoglobulin during heat treatment has been the basis for a number of fouling models. The amount of deposits at any point in the equipment has been correlated to the local \(\beta\)-lactoglobulin denaturation determined by local temperature and fouling kinetics of \(\beta\)-lactoglobulin (Changani et al., 1997).

de Jong et al. (1992) studied fouling in relation to \(\beta\)-lactoglobulin by performing experiments in plate heat exchanger during milk pasteurization. Effect of time and temperature on \(\beta\)-lactoglobulin denaturation rate was examined by measuring temperature at different locations and aggregation was measured by chromatographic methods. They used experimental data to determine fouling kinetics. de Jong et al. (1992) concluded that the main mechanism in fouling process was a reaction controlled adsorption of unfolded \(\beta\)-lactoglobulin on heat exchanger surface.

Initial stages of fouling, in other words during induction period, need to be understood for better modeling of the process since the sequence and rate of events taking place in this critical, but often ignored, stage are crucially important. Delplace et al. (1994) demonstrated that fouling starts at contact points between heat exchanger plates.
Before aggregation starts conformational changes of the protein takes some time called lag phase.

### 3.3.7 Forces Involved in Fouling

There are two key issues that play key role in the behavior of a particular particle in contact with a flat surface. The first one is the physicochemical interactions that keeps a particle attached on the surface and determines the nature and strength of the adhesive bonds. Second group of forces is the hydrodynamic interactions because of fluid flow. Flowing fluid tends to break the adhesive bonds between the surfaces and hence dislodge the particle from the surface.

Fouling period consists of a deposition and removal processes. The relationship between these two is given as:

\[
\frac{dR_f}{dt} = \frac{1}{\lambda_f \rho_f} (\dot{m}_d - \dot{m}_r) \tag{3.17}
\]

where \(\lambda_f\) is the thermal conductivity and \(\rho_f\) is the density of the fouling layer. Deposition and removal mass rates are shown as \(\dot{m}_d\) and \(\dot{m}_r\), respectively.

Förster and Bohnet (1999) and Förster et al. (1999) investigated the surface fouling of plate heat exchangers by crystalline deposits. They stated that both the deposition and removal processes are influenced by the surface free energy of the heat transfer surface. Since induction period is shorter for high energy surfaces, the main interest is towards producing low energy surfaces to prevent or at least mitigate fouling. Low energy surfaces would help to reduce both the nucleation rate and adhesive strength between crystals and the surface as Förster and Bohnet (1999) mentioned. However, the assumption is also applicable for proteinaceous systems. They recommended developing model describing the fouling process with induction period. They suggested that the imperfection of the surface need to be considered.
3.3.8 Protein Thermal Stability and Fouling

The three phases of fouling; induction, fouling, post-fouling, obtained by Fryer and Slater (1985) in heating of reconstituted milk in tubular heat exchanger was validated by Delplace et al. (1994) in plate heat exchanger with whey protein solutions. Delplace et al. (1994) determined the theoretical values of native β-lactoglobulin concentration after an average residence time using the denaturation kinetics proposed by Lyster (1970) and numerically found bulk temperature profile. They also carried out short fouling experiments with whey protein solutions to observe the initiation of fouling. They detected the beginning of deposit layer formation around a contact point between two plates of a channel. The deposit around the contact points might lead to increase in turbulence as explained by Fryer and Slater (1985) and hence resulted in a slightly higher heat transfer coefficient than it was during induction period before it started to fall. It was observed that fouling occurred where the velocity was low and eddies were significant. Experiments in a PHE with complex flow arrangement consisting of 13 plates at steady-state were performed. The numerical determination of temperature profile for each channel was used to simulate the quantity of denatured β-lactoglobulin, and was compared with the experimental measurement using an immunodiffusion method. Their fouling model, summarized below, permitted the prediction of native β-lactoglobulin at the outlet of the PHE with an error of less than 10%.

\[
C(t) = \frac{C_0}{1 + kC_0 t}
\]

\[
T \leq 363.15K \quad \log k = 37.95 - \frac{(14.51)10^3}{T}
\]

\[
T \geq 363.15K \quad \log k = 5.98 - \frac{(2.86)10^3}{T}
\]

where \( C \) is the β-lactoglobulin concentration, \( C_0 \) is the initial concentration, \( k \) is the second order rate constant, \( t \) is the time, and \( T \) is the temperature.

Delplace and Leuliet (1995) examined the milk fouling in different PHE flow arrangements by measuring the heat transfer coefficient and amount of deposit and developed an empirical model to predict the dry mass of deposit in each channel by
calculating the heat denaturation of \( \beta \)-lactoglobulin. The proposed empirical model relies on steady-state numerical simulation for the prediction of temperature profiles as denaturation and adhesion are temperature dependent.

In contrast to one and two protein phases are bulk-controlled homogeneous reaction processes, the four-phase model is closely related to a mass transfer or surface reaction process (Visser and Jeurnink, 1997). Four phases described stand for protein characteristics varying during denaturation process, such as native, unfolded, aggregated and deposited. de Jong et al. (1992) and de Jong (1996) have developed a mathematical fouling model wherein both surface and bulk reactions are considered. The denaturation of \( \beta \)-lactoglobulin and the fouling process were described as reactions in parallel. It was assumed that the participation of casein micelles in the fouling process plays a minor role. The denaturation of \( \beta \)-lactoglobulin was described as a consecutive reaction kinetics of unfolding and aggregation (de Wit and Klarenbeek, 1989). The reaction scheme is:

\[
N \rightarrow U \rightarrow A
\]

(3.19)

where N, U, and A are the native, the unfolded, and the aggregated \( \beta \)-lactoglobulin, respectively. The rate of disappearance and formation are given by:

\[
-\frac{dC_N}{dt} = k_U C_N
\]

(3.20)

\[
\frac{dC_U}{dt} = k_U C_N - k_A C_U^2
\]

(3.21)

\[
\frac{dC_A}{dt} = k_A C_U^2
\]

(3.22)

where \( t \) is the time, \( C_N, C_U, C_A \) are the concentrations of native, unfolded and aggregated \( \beta \)-lactoglobulin, respectively, and \( k_U \) and \( k_A \) are the reaction rate constants.

The local fouling process can be considered as a heterogeneous adsorption reaction of milk constituents at the surface with mass transfer and reaction series: transport in the boundary layer and the reaction process at the surface.

In the study of Grijspeerdt et al. (2004), the chemical reactions leading to fouling were assumed to follow a two-stage pathway. Native \( \beta \)-lactoglobulin (N) becomes
unfolded (U). Then it is converted to aggregate β-lactoglobulin (A). It was considered that aggregated β-lactoglobulin had no significant role in the fouling process. de Jong (1996) determined this aggregation step to be a second order reaction. Unfolded protein can also react with milk constituents (M) to form aggregated milk constituents (F) which then can be adsorbed to the heat exchanger wall causing fouling (F*). The fouling reaction was experimentally verified to be a reaction rate controlled process, not limited by mass transfer (de Jong, 1996). The reaction scheme is as follows:

\[
N \rightarrow U \quad (3.23)
\]
\[
2U \rightarrow A \quad (3.24)
\]
\[
U + M \rightarrow F \rightarrow F* \quad (3.25)
\]

The model of de Jong focuses on fouling caused by β-lactoglobulin and does not include fouling caused by precipitation of minerals.

Toyoda and Fryer (1997) assumed that denaturation and aggregation reaction are first and second order, respectively, following the approach of de Jong et al. (1992). The assumptions for the reaction/mass transfer schemes given in Figure 3.6 are as follows:

1. Proteins react in both the bulk and the thermal boundary layer in the fluid milk. Native protein N is first transformed to denatured protein D, in a first order reaction, and denatured protein then reacts to give aggregated protein A in a second order reaction.

2. Mass transfer between the bulk and the thermal boundary layer takes place for each protein.

3. Only the aggregated protein is deposited on the wall. The deposition rate is proportional to the concentration of aggregated protein in the thermal boundary layer.

4. The fouling resistance to heat transfer is proportional to the deposit amount.
The fouling scheme in Figure 3.6 was proposed by Georgiadis and Macchietto (2000) that was adopted from Toyoda and Fryer (1997). Proteins react in both the bulk and the thermal boundary layer in the milk. Native protein $N$ is transformed to denatured protein $D$, in a first order reaction. The denatured protein then reacts to give aggregated protein $A$ in a second order reaction. $N^*$, $D^*$, and $A^*$ are the native, denatured, and aggregated protein in thermal boundary layer, respectively. Aggregated protein in the thermal boundary layer, $A^*$, adheres to surfaces causing fouling deposits (Jun and Puri, 2004).

3.3.9 Calcium Phosphate Fouling

Calcium phosphate deposition and adhesion on a solid surface is the result of a competition between different types of interactions involving calcium phosphate particles, the deposition surface, the solvent and any other solute present in the system. The process starts with calcium phosphate particle formation in the bulk upon heating. These particles can then adhere to the surface following a process that is dependent on the forces established between the foulants and the surface. After the first layer is formed, other particles coming from the bulk liquid adhere on top of this layer and develop a more or less structured and compacted deposit. The structure of this deposit depends both on the structure of the first layer, which depends mainly on the surface properties, and on the particles and ions present in solution. These factors and their interactions also determine resistance to removal (Rosmaninho et al., 2005).
3.4 Hydrodynamic and Thermal Modeling of PHE

Fouling in food industry is more severe than in other industries resulting in increased pressure drop and resistance to heat transfer from heat transfer standpoint. Due to severity of deposit formation, in order to prevent these unwanted results affect the process, the plants need to be cleaned often. As this causes increased cleaning cost and time loss in addition to environmental concerns, fouling needs to be minimized by accurate design of the equipment and the process. For this reason understanding the hydro and thermal behavior in the heat exchanger is very crucial and the first step in order to be able to propose better design and operating conditions.

Understanding the need for reducing the cost of heat transfer processes in heat exchangers during fouling is crucial from industry profitability. Sandu and Lund (1982) pointed the potential for optimal design and operation of heat exchangers to reduce fouling. They stated that the dynamics of fouling and design and operation factors of heat transfer equipment are strongly related.

Fouling has been widely studied in many industries, especially chemical industry where salt and organic materials, precipitation and adhesion are dominant similar to prevalence of fouling in food industry. However, in most cases fouling in heat transfer units has been modeled with a simple representation of the heat exchanger hydrodynamics. Nevertheless, it has been long known that there are strong interactions between the physicochemical, hydrodynamics and thermal fundamentals involved in fouling process. For this reason, it is important to consider aforementioned phenomena together with fouling kinetics. Therefore, fouling of PHEs is a process that can be analyzed through momentum, heat, and mass transfer phenomena coupled with protein chemistry, denaturation kinetics, and adhesion to surface which is related with surface properties.

Although milk fouling has been extensively studied, design and operation of the heat exchangers have not been explored much. Complexity of the dynamics of fouling process makes the process analysis and optimization difficult.
Suggested experimental methods include the comparison of effect of operating methods on temperature profile by increasing the flow rate and inlet temperature of heating medium (Yoon and Lund, 1994). Flow maldistribution within the plate and from channel-to-channel plays a highly significant role in performance of plate heat exchangers. The mathematical model developed by Georgiadis et al. (1998a) for optimization describes the fouling process based on a complex reaction scheme, the hydro and thermal behavior in the heat exchanger, modeling of the transport phenomena and monitoring of fouling dynamics. The mathematical model that is used for describing the process was developed, which explains; (1) fouling process based on a complex reaction scheme, (2) the hydrodynamics and thermal behavior, through modeling of transport phenomena, (3) heat transfer critical in pasteurization and protein denaturation, and (4) mass transfer that takes place in the form of diffusion of aggregated protein through boundary layer. For optimization of the process, the optimal heating time, the optimal control profiles and optimal design must all come together. In a later study, Georgiadis et al. (1998b) simulated the model for different heat exchanger configurations.

As mentioned earlier, in engineering practice, fouling is accounted for by including a fouling resistance, \( R_F \), in the overall heat transfer coefficient equation \( U_c \). Such an approach is justified if deposition occurs uniformly throughout the exchanger.

\[
\frac{1}{U_f} = \frac{1}{U_c} + R_F 
\]  

(3.26)

where \( R_f \) is the resistance to heat transfer due to foling, \( U_f \), and \( U_c \) are the overall heat transfer coefficients for fouled and clean heat exchanger, respectively.

Assuming that the deposit thickness is uniform, \( \lambda \), and the thermal conductivity of the deposit is \( k \), then the fouling resistance is:

\[
R_F = \frac{\lambda}{k} 
\]  

(3.27)
However, fouling which occurs due to chemical reactions at liquid-solid interfaces or due to solidification of a fluid component at a wall often depends on local temperature conditions and may occur unevenly.

As the fouling rate is temperature dependent, then deposition on a clean wall occurs most rapidly in those parts of the heat exchanger where the wall is warmest. Therefore, due to non-uniform temperature profile within the heat exchanger, uneven deposition occurs with the result that the local overall heat transfer coefficient varies with position in the equipment.

### 3.4.1 Local Fouling Rate Model

In order to accurately model the performance of heat exchanger during fouling process, realistic models for local rates of deposition need to take account of (i) the rates of chemical and physical processes that leads to deposition, (ii) local temperature distribution along with the affect of local deposit thickness on heat transfer, and (iii) the influence of hydrodynamics of the equipment on the solid deposition and re-entrainment. The net rate of solid accumulation is the difference between the rate of deposition and rate of re-entrainment due to shear forces because of fluid flow (Fryer and Slater, 1985).

The complex fouling process involves mass transfer in addition to fluid flow and thermal behavior in the equipment. The transport of materials towards the solid surface being fouled can be explained by the mass transfer principles. Whenever there is a concentration gradient of a particular component, to reduce the concentration gradient, the component of concern will move from higher to lower concentration. When there is no flow or flow is laminar, mass transfer occurs as a result of the random motion of the molecules that is called Brownian motion (Bott, 1995).

When a component A in a mixture is diffusing through a fluid B towards a surface, the Fick’s diffusion equation in x direction has been used to describe the process;

\[ N_A = -D_{AB} \frac{dc_A}{dx} \]  

(3.28)
where $N_A$ is the rate of diffusion of molecules A, $c_A$ is the concentration of A, and $D_{AB}$ is the diffusivity of A through B. The diffusion is a function of physical properties of the individual components and the fluid and flow conditions.

In the case of turbulent flow, eddy diffusion is superimposed on the Brownian motion. Eddy diffusion is the result of the random physical movement of particles of fluid brought about by the turbulent conditions of the flow. Under these conditions, diffusion equation becomes:

$$
N_A = -\left(D_{AB} + E_D\right)\frac{dc_A}{dx}
$$

(3.29)

where $E_D$ is eddy diffusion. Usually, when the flow is turbulent eddy diffusion is very large and mass transfer is governed by eddy diffusion.

During accumulation of deposits on the surface of the plate heat exchanger there are considerable changes taking place in the character of fouling precursors that reach the surface. Such changes occur because of system parameters such as velocity of the flow, temperature, and coverage of surface with deposits in time (Bott, 1995). Effect of velocity on the accumulation and removal can be explained as follows. As the bulk velocity increases the velocity gradient increases which in turn results in increased shear force near deposit. Therefore, when flow velocity increases deposit thickness decreases as the increased shear force will remove more solid from the surface.

3.4.2 Dynamic Behavior in PHE

Das et al. (1995) studied experimentally the affect of parameters which demonstrate the whole spectrum of dynamic behavior of plate heat exchangers, number of transfer units (NTU), heat transfer rate ratio, and number of plates. It was stated that the experimental results conformed closely to the mathematical model obtained by simplifying the dispersion model suggested by Das and Roetzel (1995). It was found that the dispersion model gives better simulation than the heat transfer coefficient correction approach for plug flow model. They stated that the dynamic response to change in parameter magnitudes could be characterized by either a first order or a second order
system with a delay period. They concluded that the number of plates and capacity rate ratio affected initial delay period of the response.

Mehrabian and Poulter (2000) showed the variation in local hydrodynamic and thermal characteristics of the flow between two plates, and the effect of corrugation angle on the performance with fixed plate spacing. They used Computational Fluid Dynamics (CFD) modeling to investigate the microscopic flow and thermal characteristics within the channels so that complex flow patterns could be understood. Understanding the flow within the channels of the plate heat exchanger enables one to grasp the idea how further enhancements in heat transfer could be achieved. Factors such as corrugation, pitch to height ratio, corrugation angle, and flow direction are the important that affect flow and hence the heat transfer. Determining the general trends of how these important factors play critical roles is a step in the way towards designing thermal hydraulically optimized plates. The details of the model developed by Mehrabian and Poulter (2000) are summarized in the following paragraph.

Three-dimensional numerical simulation of fluid flow and heat transfer based on finite difference/finite volume technique was performed. Geometry of the system was identical with typical APV or Alpha-Laval plate heat exchanger units. Equation of continuity, momentum, and energy were discretized on the mesh and solved numerically. It was assumed that the flow was fully developed. Flow, pressure, and temperature distributions were demonstrated, and affect of corrugation angle on them was examined. Their model was capable of showing the momentum and heat transfer phenomena in the plate heat exchanger.

The study of Delplace et al. (1997) focused on verification whether or not the numerical approach proposed by René et al. (1991) was valid for heat denaturation of β-lactoglobulin in PHE channels to predict, to determine bulk temperature profiles and flow distributions in a complex flow arrangement PHE. A fouling experiment with low Reynolds number values was performed to test the hypothesis that only proteins denaturated in the thermal boundary layer were able to stick to heat transfer surfaces. The link between heat denaturation of β-lactoglobulin and the fouling of the PHE was studied.
through the overall heat transfer coefficient and dry masses of deposit measurements. The assumption that only the proteins denaturated in the thermal boundary layer were involved in deposit formation was shown not to be valid.

With the aim to initiate the use of advanced hydrodynamic calculations for the simulation of flow of milk through plate heat exchangers, flow pattern of milk between two corrugated plates was carried out using 2D and 3D computational fluid dynamics was conducted by (Grijspeerdt et al., 2003). They claimed that suitable hydrodynamic model for the equipment was basis for the simulation of the other processes occurring during thermal treatment. Hence, when an adequate model is available the process can be optimized to obtain the best operating conditions that would help to minimize undesired side effects of heating, such as fouling. The simulations were done using FINE-turbo software employing CFD code. The simulations showed that 2D ones were not good enough to explain the system response.

**3.4.3 Hydrodynamic and Thermodynamic Model**

Fouling process was simulated using the CFD code FLUENT by Brahim et al. (2003). Based on previous experimental results of Hirsch (Bohnet et al., 1999) they developed models for density of the fouling layer as a function of position and time hence the deposit thickness, and for describing heat flux distribution along the heat transfer surface. These models provided fairly good description of the fouling process.

As the denaturation reaction is temperature dependent, the most likely place for aggregation to occur is the hot surface of the heat exchanger with strong probability that surface will be fouled. Temperature distribution will affect the uniformity of deposit accumulation, where the surface is hottest it is expected that there will be more deposits accumulated. Moreover, hydrodynamics of the flow between the channels is of importance, too. For this reason, the distribution of fouling within a heat exchanger is non-uniform.

A suitable hydrodynamic model for the heat exchanger is also the basis for the simulation of the other processes occurring during the thermal treatment. When an adequate model is available, the process can be optimized to obtain the best operating
conditions that minimize undesirable side effects of the heating, while protecting the microbiological quality of the milk. According to de Jong (1996), such an optimization could potentially reduce the operating cost of a typical system by more than 50%.

The goal of modeling fouling is to develop a design tool that heat exchanger designers and plant operators can benefit from for assuring minimum fouling. It is very crucial to combine the reaction kinetics and thermo-hydraulics of the fluid flowing between the plates. A mathematical model which combined a simple model for a fluid flow with the reaction kinetics of β-lactoglobulin was proposed by Toyoda and Fryer (1997). The model incorporated i) temperature differences between the bulk and boundary layer of the fluid, ii) protein denaturation and aggregation kinetics, iii) protein mass transfer, iv) wall reaction terms. A good fit of the model to the experimental data for fouling as a function of concentration and flow rate demonstrated the validity of the model.

Grijspeerdt et al. (2003) performed advanced hydrodynamic calculations for the simulation of flow of milk through plate heat exchangers. Flow pattern of milk between two corrugated plates was carried out using 2D and 3D computational fluid dynamics. Authors claimed that suitable hydrodynamic model for the equipment was basis for the simulation of the other processes occurring during thermal treatment. Hence, when an adequate model is available the process can be optimized to obtain the best operating conditions that would help to minimize undesired side effects of heating, such as fouling.

In 2D calculation, the effect of the corrugation shape was shown. However, the 3D calculations are necessary to investigate the influence of corrugation orientation. In a later study conducted by Grijspeerrdt et al. (2004), a model for indirect thermal treatment was implemented to commercial heat exchanger systems. The model produced good results for time-temperature profile in these systems, tubular and plate heat exchangers.

Fouling studies done in laboratory are not easy to scale up to predict the behavior of realistic scale. Scherier and Fryer (1995) conducted a theoretical study that examines the scale-up of fouling from bulk and surface processes. The study included developing techniques for the measurement and modeling of food fouling, and local fouling models.
Jang et al. (1999) solved the heat transfer and fluid flow equations in 3D numerically using control volume based finite difference formulation. Algorithm proposed by Patankar (1981) was used to solve the system of finite-difference equations.

Most of the modeling studies have dealt with flow between plates or tubes. However, the plates of plate heat exchanger are not smooth but they are corrugated, and the pattern of corrugation is usually defined mathematically. Manglik and Ding (1997) analyzed the fluid flow and heat transfer of both shear-thinning and shear-thickening flows in sinusoidally corrugated channels. The analytical solutions were based on the Galerkin integral method.

Fabbri (2000) studied the effect of corrugation on the flow and PHE performance. Experimental and theoretical studies are available on the fluid dynamics and thermal behavior in corrugated channels. Characteristics of the flow depend on the corrugation properties. Corrugation increases the heat transfer surface and, more importantly, generates turbulence even at low Reynolds numbers. In terms of pressure drop, it is higher in corrugated wall channels than flat channels. Comparing the overall effectiveness, as the local convection coefficient is reduced due to reasons aforementioned, although the heat transfer surface is extended, the heat transfer effectiveness for both cases are not significantly different.

Gut and Pinto (2003) presented a PHE modeling framework that can be used for any configuration to examine the affects of configuration on the performance of heat exchanger. Total number of channels, number of passed at each side, hot fluid location, feed connection relative location, type of flow in the channels were the parameters used to define the configuration in this study. Other than the experimental work, mathematical model was conducted too. As it had been the case for most of the modeling studies, the assumptions were steady state operation with plug flow and uniform distribution of flow through the channels. Thermal effectiveness and pressure drop were calculated. The authors stated that it was not possible to derive a model that was explicitly a function of configuration parameters. For this reason, they suggested use of an algorithm for generalized model applicable for different configurations. They outlined the steps for the
algorithm and explained it with a given case study. It was concluded that assembling algorithm made the simulation more flexible. It could be used to develop optimization methods for selecting the plate configuration.

Strelow (2000) proposed a methodology for exact determination of temperature profiles in the channels. It was suggested that this method could be used for the simulation of heat flux along the walls of the plates of heat exchangers. The method was intended to determine the stationary outlet and intermediate temperatures as a function of inlet temperatures, mass streams, specific heat capacities, heat transfer coefficients, and heat transfer areas. Strelow (2000) represented the heat exchanger system using systems of linear equations, and differential equations. The developed vector and matrix calculations without iterations were recommended as a quick and effective heat transfer analysis for practical use. The numerical method used for defining the behavior of heat exchangers guaranteed exactness as no iteration was included.

Ribeiro and Andrade (2002) developed an algorithm for steady-state simulation of plate heat exchangers. The usability of the algorithm included for co-current or countercurrent flow, and series or parallel flow pattern, with either single or multi pass arrangements. The temperature profile was calculated by approximating the solution for each channel within the plate heat exchanger by a linear combination of exponential functions. They compared the simulation results with both the analytical solution and the experimental data. The algorithm described was based on the assumptions that the heat transfer between the streams can be expressed by a system of linear, first order, ordinary differential equations with constant coefficients referring to the work of Wolf (1964).

\[
\frac{d\theta_i}{dz} = \frac{UA}{\dot{m}_iC_p}[\theta_{i-1} + \theta_{i+1} - 2\theta_i] \\
1 \leq i \leq n
\]  
(3.30)

where \(n\) is the number of the channels, \(A\) is the heat transfer area of a plate, \(\dot{m}\) is the mass flow rate of the liquid in channel \(i\), \(z\) is the dimensionless position in the channel given as \(z = x/L\), and \(\theta\) is the dimensionless temperature defined as:

\[
\theta_i = \frac{T_i - T_c}{T_b - T_c} \\
0 \leq \theta_i \leq 1
\]  
(3.31)
in which, subscripts i, c and h denote for channel i, cold and hot temperatures, respectively.

This system of equations could be solved analytically. However, calculations are cumbersome, for this reason numerical methods were employed. Ribeiro and Andrade (2002) used the exponential method proposed by Zaleski and Klepacka (1992). The method was based on the approximation of the temperature profiles by a linear combination of exponential functions.

\[ \theta_i(z) = c_{i0} + c_{i1} \exp(\gamma_1 z) + c_{i2} \exp(-\gamma_1 z) + c_{i3} \exp(\gamma_2 z) + c_{i4} \exp(-\gamma_2 z) \]  

where \( c_{ij} \) are unknown constants, and \( \gamma_1 \) and \( \gamma_2 \) are the eigenvalues of the system of PHE in two channels. They are computed using:

\[ \gamma_1 = UA \left( \frac{1}{m_h C_p_h} + \frac{1}{m_c C_p_c} \right) \]  
\[ \gamma_2 = UA \left( \frac{1}{m_h C_p_h} - \frac{1}{m_c C_p_c} \right) \]

where \( m_h \) and \( m_c \) are flow rates, \( C_p_h \) and \( C_p_c \) are specific heats of hot and cold fluids, respectively. \( U \) is overall heat transfer coefficient and \( A \) is the heat transfer area.

Stream temperatures were calculated once the constants \( c_{ij} \) values were known. The validity of the developed algorithm was tested by comparing it with (1) exact solution for cases including both series and parallel, and cocurrent and counter current flow pattern, (2) experimental data obtained by different flow rates, inlet temperatures, number of plates, (3) multipass orange juice pasteurization unit data. The algorithm was validated and used for simulation in industrial PHEs.

Lakshmanan and Potter (1990) developed a “cinematic model” to simulate the dynamic performance of PHEs where time and space were considered to be independent. The cinematic approach requires multiple discrete batch “plugs” instead of just one as in the steady-state case. Each of the snapshots of time can be considered as a sequence of instantaneous moves in space followed by a holding time. The “cinematic model” requires a minimal amount of computations to accurately simulate the dynamics of PHEs.
It is advantageous as it computes the dynamic and steady-state profiles in one sweep. However, the drawback is that this approach cannot be used to predict the temperature profiles over the whole heat exchanger areas but is limited to inlet and outlet stream temperatures.

Das and Murugesan (2000) predicted the transient response of multi-pass PHE based on an axial heat dispersion model in the fluid taking the deviation from ideal plug flow. The PHEs are different from shell-and-tube heat exchangers because of the phase lag at the entry and the successive channels. These phase lags including mixing of the fluid between passes was first taken into consideration in the solution algorithm.

Luo et al. (2003) modeled a dynamic behavior of one-dimensional flow multi-stream heat exchangers. Most of the current available literature involves numerical or analytical analysis of the steady state behavior of multi-stream or multi-channel heat exchangers. However, in industry, heat exchangers frequently undergo transients.

There have been studies in different engineering fields about the flow profile between the corrugated plates. Ding and Manglik (1996) and Manglik and Ding (1997) obtained analytical solutions by the Galerkin function-based integral method for fully developed 2D turbulent flow in double-sine ducts. The most widely used plate surface pattern consists of chevron-type corrugations with a sinusoidal profile. Fischer and Martin (1997) analyzed the fully 2D turbulent flow in straight ducts of constant cross section with sinusoidally curved wall by the finite element method. The Navier-Stokes equation and energy balance equation described for the cross-sectional plane (x, y) of wall corrugation were employed. The models cannot describe the hydrodynamic and thermodynamic performances of PHEs over the entire plate surface. A steady, 2D, laminar fluid flow between two horizontal parallel plates was investigated numerically by Ingham et al. (1995). The governing equations of mass, momentum and energy retain their respective coupling and nonlinearity. Hence, changing the duct temperature affects both the fluid and thermal fields, i.e., stratification or recirculation patterns of flow streams.
Jun et al. (2003) compared the 2D model that took into account the hydrodynamics of fluid flow with the 1D model by predicting the temperature distribution of flow in PHEs. 2D model followed the magnitudes and trends of the experimental data more accurately and could also identify zones most prone to milk deposits, where the temperature differentials between the cold and hot streams, more precisely.

Rao et al. (2002) analyzed the PHE performance considering the heat transfer coefficient inside the channels as a function of flow rate in that particular channel different from the previous studies which assumed equal flow in all channels. The assumption of plug flow inside channels and equal distribution within the plate is not realistic. The flow maldistribution within the plate and from channel-to-channel affects the actual performance of the equipment.

### 3.5 Surface Chemistry and Thermodynamics

The initial interaction between the fluid and hot solid surface involves protein adsorption. The nature and the properties of the solid thus influence the extent and rate of protein adsorption. All surfaces are energetically unfavorable in that they have a positive free energy of formation. A simple rationalization for this comes from considering the formation of new surfaces by cleavage of a solid and recognizing that bonds have to be broken between atoms on either side of the cleavage plane in order to split the solid and create the surfaces. Breaking bonds requires work to be done on the system, so the surface free energy (surface tension) contribution to the total free energy of a system must therefore be positive. Surface chemistry is concerned with the formation, the chemical structure and changes as well as the physical behavior of surfaces and interfaces. Surface tension is defined as equivalent force to do the work necessary to create a new piece or surface (per unit area) and therefore a quantity for the energy content of a liquid surface (SI-units: mN/m and mJ/m²)
3.5.1 Surface Energy, Wettability, and Contact Angle

When a drop of water is put on a solid surface it attaches to the surface to some extent determined by surface and water interaction, and thus forms a sessile drop. It is also observed that many liquids can climb up to a certain height in a capillary tube and form a meniscus at the top. The angle formed between the liquid-vapor interface and the liquid-solid interface at the solid-liquid-vapor three phase contact line is defined as the contact angle. In other words, contact angle is the angle which encloses the tangent line on the drop shape from the three-phase point to the base line on the solid surface.

Wettability denotes the ability of liquids to spread on a surface; the better the wettability the lower is the contact angle. Surfaces are called incompletely wettable in case of an occurring contact angle less than 90°. Spreading is associated with wetting of a surface by a liquid with a contact angle of about 0°.

Wetting may be considered as a process of achieving molecular contact. The extent of wetting can be defined as the number of molecular contacts between two phases. Contact angle is a measure of the wettability of a surface or interface with a different liquid phase; the smaller the contact angle, the better the wettability. Presently, interpretation of contact angles is one of the ways for determining surface tensions of solids. However, unfortunately, a contact angle phenomenon is very complex.

Equilibrium contact angles are functions of the surface free energies of the solid surface and the liquid in contact with it and of the free energy of the interface in between. Minimizing the overall free energy of a system consisting of a liquid in contact with a solid yields the Laplace equation of capillarity (Lamb, 1928)

\[
\gamma_{lv} \left( \frac{1}{R_1} + \frac{1}{R_2} \right) = \Delta \rho g z + c = \Delta P
\]

and Young’s equation

\[
\gamma_{lv} \cos \theta_c = \gamma_{sv} - \gamma_{sl}
\]
where $R_1$ and $R_2$ are the principal radii of curvature at a point of the liquid surface, $\theta_e$ is the equilibrium contact angle that the drop makes with the surface, $\gamma_{lv}$, $\gamma_{sv}$, and $\gamma_{sl}$ are the interfacial tensions between the liquid and the vapor, the solid and the liquid, and the solid and the vapor, respectively.

Surface free energy or surface energy denotes the total energy content of a solid surface in equivalence to the surface tension of a liquid; the surface free energy can be decomposed for most practical considerations into polar and dispersive contributions. In SI-units, the unit of surface energy is as N/m and mJ/m².

Surface free energy of a solid is approximated by a critical surface tension. Critical surface tension divides the liquids forming zero contact angle on the solid those forming contact angles greater than zero.

$$\gamma_{so} = \frac{\left[\gamma_1 (1 + \cos \theta) + \pi \Phi \right]^2}{4 \Phi^2 \gamma_1}$$

(3.37)

where $\Phi$ is a property of the particular liquid-solid system and can be calculated by using the molecular properties of the two substances; dipole moment, polarizability, ionization energy, and molecular radius (Good, 1960).

### 3.5.2 Thin Liquid Film/Contact Angle Phenomena

Young’s equation is not a universal formulation, i.e., is not applicable in all situations. The adsorption of the liquid component on the solid surface may affect the magnitude of contact angle. One of the ways to consider adsorption on a solid surface is thermodynamic approach established by Gibbs. This approach takes adsorption into account by considering a solid-vapor (sv) interface with a surface tension $\gamma_{sv}$ rather than taking only pure solid-vacuum interface surface tension $\gamma_s$. the difference between these surface tension, $\gamma_{sv} - \gamma_s$, is defined as the equilibrium spreading pressure. Generally, adsorption onto surface decreases the solid surface tension, so the equilibrium spreading pressure is positive.
The determination of $\gamma_{sv}$ and $\gamma_{sl}$ interfacial tensions is important. Due to complexities in measuring directly surface tensions, indirect approaches can be used. Measuring contact angle is one of the indirect ways for determining interfacial tensions (Li and Neumann, 1996).

Attractiveness of using contact angle measurement for estimation of interfacial tensions is because of ease of the measurement. In order to estimate interfacial tensions an equation involving three interfacial tensions is required in addition to Young’s equation (3.36). These equations mainly fall into two groups; (1) based on the surface tension components approach, and (2) based on the equation of state approach.

Although there are many methods developed for the measurement of contact angles, for now, measurements are performed mainly by three groups of methods (Moy and Neumann, 1996);

1. The methods based on the Axisymmetric Drop Shape Analysis (ADSA) of sessile drops.
2. The method of capillary rise on a vertical plate.
3. The use of a goniometer telescope for the direct, approximate measurement of contact angles.

Surface energy denotes the wetting capability of the surface. Surface energy is quantified in terms of the forces acting on a unit length at the solid-air or the solid-liquid interface. At the liquid-solid surface interface, if the molecules of the liquid have a stronger attraction to the molecules of the solid surface than to each other (the adhesive forces are stronger than the cohesive forces), then wetting of the surface occurs. If the liquid molecules are more strongly attracted to each other and not the molecules of the solid surface (the cohesive forces are stronger than the adhesive forces), then the liquid beads-up and does not wet the surface of the part. One way to quantify a liquid's surface wetting characteristics is to measure the contact angle of a drop of liquid placed on the surface of the subject object.
By viewing small droplets of liquid on a surface in profile, the effects of interfacial tension can be readily observed. In order to define these droplet profiles it is common to draw a line tangent to the curve of the droplet at the point when the droplet intersects the solid surface. The angle formed by its tangent line and the solid surface is called the contact angle. Liquids wet surfaces when the contact angle is less than 90 degrees.

3.5.3 Adsorption

The adsorption of molecules onto a surface is a necessary prerequisite to any surface mediated chemical process. Adsorption is the enrichment or agglomeration of particles on a surface or interface.

Protein adsorption onto solid surfaces results from a competition of various interactions involving the protein, the surface, the solvent and any other solute, which might be fat globules, minerals in protein adsorption, present in the system (Haynes and Nordes, 1994). These interactions are significantly affected by the surface properties. Surface free energy of the solid–liquid interfaces is one factor that can give some indication of the degree of foulant adsorption. Although it is known that adsorption is influenced by surface energy, there is no quantitative prediction relating surface energy and adsorbed amount of foulant.

3.5.4 Adhesion

Adhesion is simply defined as the chemical or mechanical bonding (strength) of one material to another material. Work of adhesion is the term used for the intermolecular (attraction) energy between to phases per unit area being in contact to each other; amount of energy necessary to perform adhesion processes.

The physical phenomenon adhesion can be defined simply as sticking of two materials together is very important in food industry due to increased cleaning costs and sanitary concerns as well as fouling problems. Mechanisms involved in the adhesion are mechanics, thermodynamics, and chemistry. Mechanical interlocking is the oldest adhesion theory that explains adhesion process due to locking through materials pores.
and asperities. Rugosity increases the actual available surface and hooking sites. Moreover, good wettability of a fluid adhesive on the solid, rugosity would improve adhesion. However, if the wettability is low, rugosity would decrease adhesion. Surface defects, finish and porosity on ability to retain adhered microbial contamination. Surface rugosity may also influence food flow and adhesion on the equipment (Michalski et al., 1999).

Surface tension, surface energy and adhesion are all interrelated. The numerical difference between the surface tension of a coating and the surface energy of a substrate has a profound effect on the way in which the liquid coating flows over the substrate and on the strength of the adhesive bond between the substrate and the dry film.

If the surface tension of the coating is greater than the surface energy of the substrate then the coating will not spread out and form a film. As the surface energy increases, a stage can be reached where the coating will spread out and form a film but, when dry, has poor adhesion. Further increases in the surface energy will result in easier wet-film formation and better dry-film adhesion.

Adhesive testing involves the application of force to remove the coating from the substrate. The intention is to measure the force needed to overcome the forces of adhesion between coating and substrate. In practice however, the cohesive strength of the coating and of the substrate both have an effect on how easy it is to remove the coating.

Fouling occurs when gravitational forces are negligible, i.e., only particulate materials of colloidal size, with dimensions less than 1 μm, adsorbs onto the solid surfaces immersed in fluids. Larger particles are not able to adhere to the surface since gravitational and hydrodynamic forces are strong enough to remove them. The theories that explain adhesion are the adsorption theory, the diffusion theory and the electrostatic theory.

**3.5.5 Kinetics and Thermodynamics of Protein Adsorption**

Protein adsorption on surfaces is a very complex process. When the adsorbing particles are large, as it is the case usually, then the surface protein interactions are
usually long range and the strength of these interactions is very high. In addition, particles may undergo conformational changes upon adsorption. Explanation of kinetics and thermodynamics of these changes is nontrivial.

In a closed system of a liquid and a solid in equilibrium, the liquid is in contact with a plane surface of an isotropic solid that is not soluble in that particular liquid, and saturated vapor of the liquid. The Young-Dupré equation (Chow, 1998) is:

$$\gamma_{s,v_2} - \gamma_{s,L_2} = \gamma_{L,v_2} \cos \theta$$  \hspace{1cm} (3.38)

When the surface of the solid is an equilibrium surface, the surface tension and the surface free energy of the solid are the same, therefore;

$$F_{s,v_2} - F_{s,L_2} = F_{L,v_2} \cos \theta$$  \hspace{1cm} (3.39)

Maximum reversible work of adhesion of liquid $L_2$ to $S_1$ is

$$W_{adh} = F_{s,v_2} + F_{L,v_2} - F_{s,L_2}$$  \hspace{1cm} (3.40)

where $F_{s_1}^o$ is the surface free energy of the solid $S_1$ in vacuum.

$$W_{adh} = F_{s,v_2} + F_{L,v_2} - \left( F_{s,v_2} - F_{L,v_2} \cos \theta \right)$$

$$= F_{L,v_2} \left( 1 + \cos \theta \right) + F_{s,v_2} - F_{s,v_2}$$  \hspace{1cm} (3.41)

### 3.5.5.1 Wetting and Thermodynamical Adsorption

A liquid drop put on a solid surface either spreads and forms a film or forms a bead on the surface. The extent of drop spreading is called wetting. A good wettability means that liquid and solid have strong affinity and are likely to adhere well. Wettability between the materials should be sufficient for the adhesion to occur. This is satisfied if materials are close enough because of the short action distance of intermolecular forces that govern wetting phenomena to thermodynamical adsorption.
Young’s forces equation relates solid and liquid surface tensions, solid-liquid interfacial tension and the liquid contact angle at the solid/liquid/air triple line.

\[ \gamma_s = \gamma_{sl} + \gamma_L \cos \theta \]  \hspace{1cm} (3.42)

According to thermodynamical adsorption theory, adhesion is due to electodynamical intermolecular forces; van der Waals forces and hydrogen bonds, acting at the interfaces. Interfacial attraction between adhering materials is expressed in terms of work of adhesion. The work of adhesion of \( W_a \) refers to the free energy difference between two defined states, the first of two phases in contact in equilibrium and the second comprising the two phases separate in equilibrium with their own vapor.

\[ W_a = \gamma_s + \gamma_L - \gamma_{sl} \]  \hspace{1cm} (3.43)

Instead of measuring adhesive forces directly, they can be deduced from wettability using the adsorption theory. Contact angle can be used for the calculation of work of adhesion by combining Young and Dupré equations.

\[ W_a = \gamma_L (1 + \cos \theta) \]  \hspace{1cm} (3.44)

In the equation for calculating work of adhesion, parameter other than contact angle that needs to be known in addition to contact angle is the liquid surface tension. The most useful method given in literature is by using tensiometer. It involves measurement of force. In the Wilhelmy method with tensiometer, a vertically suspended platinium plate is made to contact the liquid sample. This special plate allows for perfect wetting of the solid by the liquid. The sample pulls the plate into the liquid and the force required to pull the plate back to the sample surface level is measured. Therefore, surface tension expressed in mJ/m\(^2\) can be calculated by dividing the measured force with the wetting length.

The surface energies \( \gamma \) are defined using Gibbs G or Helmholtz F free energies. Gibbs free energy is used for constant pressure and Helmholtz free energy is for constant temperature situation.
The liquids that have lower surface tension than critical surface tension, liquid-vapor surface tension that is found by correlating with contact angle and finding the corresponding value when cosine function of the measured contact angles is equal to 1, spread on and completely wet the solid surface. On the contrary, the liquids those have higher surface tension than the critical value of surface tension can only partially wet the surface and hence resulting in a three-phase contact point with a noticeable contact angle. Contact angle measurements have been very useful to develop models for estimation of solid surface energies (Packham, 1996).

The main conclusion of the researchers that investigated the wetting behavior of liquids on low-energy surfaces is that contact angle, measured through the liquid, increases with increasing liquid-vapor surface tension. In addition to this crucial statement, with some surfaces, it was found that the contact angle of a drop appears to depend almost entirely on the liquid-vapor surface tension and much less on the other properties of the liquid (van Giessen et al., 1997). Zismann et al. (1964) suggested that the contact angle of a liquid drop on a particular solid surface could be determined only by the liquid-vapor surface tension. In a more recent study conducted by van Giessen et al. (1997), it was found theoretically that the same trend of \( \cos(\theta) \) versus liquid-vapor surface tension by using a generalized van der Waals theory. They concluded that, both the contact angle and the liquid-vapor surface tension depend on fluid properties rather than being functions of each other (Bahramian and Danesh, 2004).

Various topographies and physicochemical properties of stainless steel, due to production in various grades and surface finishes, affect adhesion. In order to define surface topography that plays vital role in adhesion process, surface roughness has been investigated. For more accurate description, use of parameters other than the average surface roughness such as reduced valley depth was proposed by Mettler and Carpentier (1999). Since stainless steel is amongst the materials that display non homogenous surface free energies especially in the case of complex composition of fluid, for instance presence of suspending media, it is difficult to recommend a relation between adhesion and surface free energy (Jullien et al., 2002).
Protein adsorption is associated with the initiation of adhesion to surface, which then becomes difficult to remove because of high surface temperature. The adsorption behavior of globular proteins, such as β-lactoglobulin, at interfaces has been studied by many techniques; reflectometry, cyclic voltammetry, adsorption spectroscopy and ellipsometry (Murray and Cros, 1998).

3.5.5.2 Thermodynamics of Wetting

The wetting state of a fluid/solid system depends on the relative value of the surface tensions acting at different interfaces. Unfortunately, free energy of a surface is not directly measurable. For a pure liquid and perfectly smooth surface it is easy to understand the concept and measure the contact angle, used to define the degree of wettability of the surface by the stated fluid. However, in reality surfaces are not ideally smooth, i.e., roughness and chemical heterogeneity of the surface brings up some challenges in the measurement. In the case of pure liquid, three states of wetting can be distinguished; complete, partial, or nonwetting. For partial wetting or nonwetting systems, contact angle that liquid forms on the surface can be defined for a drop of liquid in contact with a solid. Contact angle of 90° is considered as the limit between partial and nonwetting cases. When the equilibrium conditions are satisfied, the contact angle is related to the interfacial tension by Young-Dupré equation (Denoyel et al., 2004).

Contact angle measurement: Contact angle gives information about surface energetics, hydrophobicity, roughness, and chemical heterogeneity. There is no direct method for surface tensions. Indirect methods include contact angle measurement for different liquids, direct force measurements, solidification front techniques, film floatations, sedimentation techniques, gradient theory, and the Lifshitz theory of van der Waals forces. Among them, contact angle is the most appropriate one for determining surface tension due to its simplicity.

Contact angle of a liquid drop on a solid surface is governed by mechanical equilibrium under the effect of interfacial tensions, liquid-vapor, solid-vapor, and solid-liquid surface tensions, represented as $\gamma_{lv}$, $\gamma_{sv}$, and $\gamma_{sl}$, respectively. This equilibrium relation is known as Young’s equation:
\[ \gamma_n \cos \theta_Y = \gamma_{sv} - \gamma_{sl} \]  

(3.45)

where \( \theta_Y \) is the Young contact angle. Only \( \gamma_n \) and \( \theta_Y \) are measurable (Tavana et al., 2004).

The thermodynamical adsorption theory is based on Young force equation and Dupré energy equation. Young’s equation relates solid and liquid surface tensions \( \gamma_s \) and \( \gamma_L \), solid-liquid interfacial tension \( \gamma_{sl} \), and the liquid contact angle \( \theta \) at the solid/liquid drop/air line shown in Figure 3.7.

![Figure 3.7 Contact angle](image)

Adesso and Lund (1997) studied the influence of solid surface energy on protein adsorption. They examined Teflon and different types of stainless steel for \( \alpha \)-lactoalbumin and \( \beta \)-lactoglobulin adsorption. The results of the experiments showed that the critical surface tensions determined by contact angle measurements were significantly higher for stainless steel. They stated that Wilhelmy Plate Technique for contact angle measurement did not give initial unchanged adsorbed protein because of conformational changes that was likely to occur at protein/air interface. It was found that adsorption of \( \alpha \)-lactoalbumin resulted in decrease in critical surface tension, whereas \( \beta \)-lactoglobulin did not had significant effect. They stated that although the measured contact angles were different for different surfaces, the protein adsorption onto surfaces was not less for low energy surfaces. Therefore, they concluded that the critical surface tension determined by contact angle measurement cannot be used to predict the rate and extent of milk whey protein fouling.

The concept of measuring strength of adhesion measurement in terms of work of adhesion was introduced by Harkins. In principal, the work of adhesion is a useful
measure of the strength of adhesion in a particular system. Clint and Wicks (2001) calculated the work of adhesion of liquid to solid surfaces under water, which was new compared to other studies conducted in air. They used purified water in the system. Contact angle measurements were performed for the calculation. They concluded that contact angles for the liquids examined, allow components of solid surface energies. For the calculation in addition to surface energy terms, interfacial tension was used too. The important result of this study was that the adhesion of oily material to solid surfaces in air or under water was entirely different. It can be concluded that since adhesion in the system is different and changes with the fluid it is present in, then by suitable surface modification adhesion can be inhibited or promoted as desired.

It has been long known that protein deposition is one of the key factors in fouling in dairy processing. Adsorption of denatured protein onto solid surfaces results from competition between interactions involving the protein, the surface, the fluid and the solutes present in the system. The important point is that these interactions are affected by the surface properties.

Surface free energy of the solid-liquid interfaces is defined by the Young’s equation (3.36). It gives indication of the degree of adsorption. Janocha et al. (2001) observed that the adsorbed amount of albumin decreased with increasing surface energy.

Fouling starts by the adsorption of proteins on the surface, followed by deposition of protein and other substances on top of the initially adsorbed layer. For this reason, the effect of surface material on fouling is very strong especially in the initiation stage. Once the plate is fully covered with deposits, the original material will be less important (Visser and Jeurnink, 1997). In the initiation stage, the interaction is between the heat exchanger surface and milk protein β-lactoglobulin. These interactions are significantly influenced by the surface properties (Santos et al., 2004). Surfaces with high surface energy, for instance stainless steel, are more easily wetted than those with low surface energy and thus more prone to fouling. Investigation of use of low surface energy materials in at least certain parts of heat exchanger, where possibility of fouling is high, is suggested as a way for minimization of fouling.
3.6 Recent Studies about Surface Coating and Fouling

It has been long known that protein deposition is one of the key factors in fouling in dairy processing. Adsorption of denatured protein onto solid surfaces results from competition between interactions involving the protein, the surface, the fluid and the solutes present in the system. The important point is that these interactions are affected by the surface properties. Fouling starts by the adsorption of proteins on the surface, followed by deposition of protein and other substances on top of the initially adsorbed layer. For this reason, the effect of surface material on fouling is very strong especially in the initiation stage. Once the plate is fully covered with deposits, the original material will be less important (Visser and Jeurnink, 1997). In the initiation stage, the interaction is between the heat exchanger surface and milk protein β-lactoglobulin. These interactions are significantly influenced by the surface properties (Santos et al., 2004). Surfaces with high surface energy, for instance stainless steel, are more easily wetted than those with low surface energy and thus more prone to fouling. Investigation of use of low surface energy materials in at least certain parts of heat exchanger, where possibility of fouling is high, is suggested as a way for minimization of fouling.

Adesso and Lund (1997) studied the influence of solid surface energy on protein adsorption. They examined Teflon and different types of stainless steel for α-lactoalbumin and β-lactoglobulin adsorption. The results of the experiments showed that the critical surface tensions determined by contact angle measurements were significantly higher for stainless steel. They stated that Wilhelmy Plate Technique for contact angle measurement did not give initial unchanged adsorbed protein because of conformational changes that was likely to occur at protein/air interface. It was found that adsorption of α-lactoalbumin resulted in decrease in critical surface tension, whereas β-lactoglobulin did not had significant effect. They stated that although the measured contact angles were different for different surfaces, the protein adsorption onto surfaces was not less for low energy surfaces. Therefore, they concluded that the critical surface tension determined by contact angle measurement cannot be used to predict the rate and extent of milk whey protein fouling.
Müller-Steinhagen and Zhao (1997) developed stainless steel surfaces with low surface energy by ion implantation. They found that CaSO$_4$ fouling was considerably reduced.

In previous studies on the adsorption of protein on surfaces with different surface energies conducted by Yoon and Lund (1994), Adesso and Lund (1997), and Janocha et al. (2001) no clear quantitative relationship was found between the amount of deposit and the surface energy.

Polymer coatings have been studied due to their low surface energy (Adesso and Lund, 1997, Britten et al., 1988, Santos et al., 2001), however, the main drawback is reduced abrasion characteristics, and heat transfer properties. Other coatings such as DLC, SiOx, Ni-P-PTFE (Rosmaninho et al., 2003; Santos et al., 2001), and Ti (Addesso and Lund, 1997) have also been studied.

Santos et al. (2001) found that modification of the stainless steel surface influenced the adsorption of protein. They found that both Ni-P-PTFE and SiOx coatings reduced the amount by which the heat transfer coefficient decreased. However, the compact deposit formed on the Ni-P-PTFE surface was harder to remove during cleaning. The SiF$_3^+$ ion implanted surface showed slow adsorption kinetics but rinsing did not remove as much protein as for stainless steel surfaces. MoS$_2$ ion implanted surface did not show a significant improvement from stainless steel fouling behavior.

Beuf et al. (2003) investigated the effect of different surface treatments, including Diamond Like Carbon (DLC), Silica, SiOx, Ni-P-PTFE, Excalibur®, Xylan® and ion implantation (SiF$_3^+$, MoS$_2$), on fouling and cleaning of plate heat exchangers during milk processing. The decrease in the overall heat transfer coefficient, the increase in the pressure drop and the quantity of denatured β-lactoglobulin after the heat treatment were used for comparison of surfaces. After fouling, they did not observe any significant difference between all the modified steels and the reference unmodified stainless steel. They suggested that the free surface energy plays a predominant role and the roughness a minor role in the level of fouling and cleaning efficiency. In addition, damaged surfaces and the conditioning film also influenced the surface properties.
Zhao et al. (2002) examined the effect of cationic surfactant and PTFE emulsion addition on Ni-P-PTFE coating rate and PTFE content in the coatings, and described a method of improving the adhesion of the Ni-P-PTFE coating. The adhesion of the Ni–P–PTFE layer was significantly improved by gradually increasing the PTFE content from the substrate to the top surface. It has been demonstrated that these graded electroless Ni–P–PTFE coatings reduced the formation of deposits on heat exchanger surfaces significantly.

Modified surfaces, Diamondlike Carbon (DLC) and Titanium Nitride (TiN), were tested by Ramachandra et al. (2005). Some differences in deposit appearance were found between whey protein deposits on stainless steel and titanium nitride surfaces. It is possible that the deposit on stainless steel forms more uniformly than for titanium nitride, where a more patchy deposit was found.

Rosmaninho et al. (2005) studied the fouling caused by calcium phosphate. In this work, reactive sputtering was used as an anti-fouling technique. They obtained a number of stainless steel materials with similar surface composition and morphology but variable surface energy values. By this way, they tested the effect of surface energy on deposit formation under constant conditions of temperature, pH, calcium concentration and hydrodynamics. All surfaces were 316 2R (bright annealed) based and were prepared by a surface modification technique called reactive sputtering coating (with Ti and N). They concluded that surfaces with surface energies had different deposition curves. Under similar conditions, more deposit was formed at 70°C than at 44°C for all the surfaces at initial stages of deposition; however, for longer deposition times, this behavior was only found on surfaces with higher surface energy. After cleaning less deposit remained on the surfaces with the highest and the lowest surface energy.

Rosmaninho et al. (2007) investigated the fouling behavior of stainless steel surfaces with different properties for different dairy products under different condition. Modification techniques used were \( \text{SiF}_3^+ \), \( \text{MoS}_2^{2+} \), and TiC ion implantation; diamond-like carbon (DLC) sputtering; DLC, DLC-Si-O and SiO\(_x\), plasma enhanced chemical vapor deposition (PECVD); autocatalytic Ni-P-PTFE and silica coating. Aqueous
solutions that simulate milk were used. In all cases, the fouling behavior was affected by the surface material either in deposition or cleaning phases. Among all the surfaces, they found that Ni-P-PTFE surface promoted less deposit build up and was easiest to clean.

The goal of modeling fouling is to develop a design tool that heat exchanger designers and plant operators can benefit from for assuring minimum fouling. It is very crucial to combine the reaction kinetics, thermohydraulics of the fluid flowing between the plates, and surface properties of the equipment material. By the developed model, approximate reduction in deposit formation for a given surface compared to common surface in industrial PHEs, stainless steel, at any operating conditions can be predicted.
Chapter 4

METHODOLOGY

Experimental work followed four main steps. First, the bench scale plate heat exchanger was designed and fabricated. Second, test surfaces were prepared and characterized. Third, coated surfaces were tested for fouling. Finally, the milk foulants deposit was quantified. The experimental design for this study is summarized in Table 4.1. Vitamin D enriched whole milk purchased from a local grocery store was used in the tests.

4.1 Bench Scale Tester

While designing the set up, a system that would sufficiently represent the thermal-hydraulic behavior of parallel plate heat exchanger was the top priority. By this way, the cost of investigating the effect of surface energy on fouling in plate heat exchangers was reduced by scaling down to bench scale tester. Other advantages of using the bench scale parallel heat exchanger are summarized in terms of experimental and computational benefits.

- Measurement: If real plate heat exchanger is used for the test, performing surface characterization measurements will be difficult. Since goniometer size is not suitable to fit the large size plates of the heat exchanger, small test surfaces were prepared for the bench scale tester. After the experiments were performed, amount of deposit on the test surfaces were measured. For measuring the deposit on the plates of full size plate heat exchanger, this measurement can be done by scraping the fouling from the surface. This might lead to increase in error of measurement. Another error will be in temperature measurement. There will be a need for more thermocouples on the larger plates. Thus, the error of measurement will be higher.
• Model simplification: On the plates of heat exchanger, the corrugation of the surface might effect the distribution and amount of fouling; however, in this study, flat smooth surface was used

4.1.1 Test Set-up

After characterization of the surfaces, they were tested under different conditions on a small scale set-up manufactured by Omega Piezo Technologies, Inc. (Figure 4.1). Milk was flowing between two parallel stainless steel, electrically heated plates (Figure 4.2 (a)). The coated test samples were placed on one of these stainless steel surfaces.

![Figure 4.1 Bench scale milk fouling test set-up](image)

4.1.1.1 Pump

A centrifugal pump was used with variable speed pump drive. The pump drive contains the pump drive motor and the pump which is magnetically coupled to the motor. The pump can deliver up to 26.5 L/min (7 gal/min) of liquid and has speed range of 90 to 9,000 rpm. The maximum fluid temperature it can handle is 121°C. It has line speed regulation of ±1% and load speed regulation of ±2% of maximum speed.

4.1.1.2 Coiled Tube Heat Exchanger

Heat exchanger was used to cool the outlet milk to the desired temperature before returning to the holding tank to keep the inlet milk temperature constant. Hot milk exiting
from the parallel plate channel passes through a coil made of stainless steel 316 tubing enclosed in a PVC casing. Water enters the PVC casing from the bottom and leaves from the top. Water flow rate can be adjusted to control the amount of heat removed from the milk.

4.1.1.3 Milk Tank

Milk tank made of high density plastic was used as a reservoir and which can hold more than 37.9 L (10 gallons) of milk.

4.1.1.4 Flow Meter

A variable area rotameter was used to control and monitor milk flow rate. The rotameter’s accuracy was ±5%. Besides, mass of the milk collected in 60 seconds was measured every 15 minutes using a beaker and stop watch.

4.1.1.5 Line Heater

A rope-type heater was wrapped around the tube to heat the milk to a desired temperature before it entered the test channel.

4.1.1.6 Parallel Plate Test Channel

Figure 4.1 and Figure 4.2 show the different components of the test section and how they were assembled. The main body, plenum and flanges were made of clear acrylic so that test plates and side walls could be observed from outside. The flanges and the plenums were clamped to the main body by the four threaded rods and butterfly nuts. The side wall with test plates, the side wall without test plates, test plate holders as well as the test plates were made of stainless steel 316. The main body had gaskets on the top and bottom to prevent leakage of milk. Both the side walls were screwed down to the main body. The main body and side walls formed a rectangular passage of 5 mm x 60 mm with 184 mm long (Figures 4.3 (a) and 4.4 (a)).

Milk entered the plenum, following the filling of plenum, it flows through the rectangular passage. Thermocouples were used to measure milk temperatures inside the inlet and exit plenums (Figure 4.2 (a)).
The side wall with test specimens, each were 25 mm x 25 mm in size, contained the test plate holder and the three test plates. Thermocouples were inserted into the test plate from the back of the side walls (Figure 4.4 (b)). The test plate holder had o-ring grooves (Figure 4.3 (d)) so that milk did not leak through the holes for thermocouples and was bolted to the side wall from the back. Thermocouples were also inserted into opposite side wall, which does not contain any test specimens, at exactly the same locations on opposite wall.

Figure 4.2 Components of bench scale parallel plate exchanger unit

Flexible wall heaters, two 80 W heaters on each side, i.e., 160 W each side, were attached to the side walls from the outside to heat the stainless steel walls to the desired temperature. Potentiometers were used to control the amount of heating. Insulation was used to prevent heat loss from the heater.
Figure 4.3 Components of test plate

(a) Side wall with test plates
(b) Test plates and test plate holder

Figure 4.4 (a) Flow direction of the parallel plate channel and (b) Position of thermocouple locations on the test surface
4.2 Materials

Vitamin D enriched whole milk purchased from a local grocery store was used in the tests. The milk was preheated in a pot on hot plate to the test temperature to simulate the thermal behavior of milk flow in the heating section of the PHE system.

4.3 Surface Preparation

4.3.1 Cleaning the Stainless Steel Surface

Stainless steel 316 surface was cleaned with 25 ml 6% sodium hypochlorite diluted with 5 L water at 40°C followed by water rinse. This treatment dissolves milk fats, proteins, and carbohydrates, and loosens and suspends other soil particles. Sodium hypochlorite is typically used at concentrations of 100 to 200 ppm active chlorine for applications to food contact surfaces. 25 ml phosphoric and sulfuric acid mixture was diluted in 5 L at 60°C. Acid cleaners remove or prevent accumulated mineral deposits or milkstone buildup. Coated surface did not require any cleaning agents as the foulants could easily be removed with warm water.

4.3.2 Surface Coating

Four different surfaces were used in this study in addition to the control surface, stainless steel. Procedure for preparation of each of them is given in the following subsections.

4.3.2.1 Microlube/PTFE

The microlube/PTFE (Poly Tetra Fluoro Ethylene) Nickel coating (Micro Plating Inc., Erie, PA) is an electroless nickel composite plating process that co-deposits PTFE at 20 to 25% by volume as occluded particles in the plated coating. The Teflon® based Ni-P-PTFE coating contains about 15-25% Teflon®, 60-65% nickel and about 10-12% phosphorus. The Teflon® present in the coating helps to reduce the friction by imparting lubricity to the surface. The corrosion resistance of the coating is controlled by the amount of phosphorus. As the amount of phosphorus in the mixture increases, the resistance of the coating to corrosion is more. These Teflon-based coatings have a...
thermal conductivity nearly equal to stainless steel owing to the high percentage of nickel present.

4.3.2.2 TM117P

TM117P deposit is a composite of 12-25% by volume of Teflon\textsuperscript{®} particles dispersed in high phosphorous electroless nickel matrix (Techmetals, Inc., Dayton, OH). It is hard, corrosion resistant and has very good friction characteristics. This coating is also named as graded Teflon\textsuperscript{®} and is very similar to uniformly dispersed PTFE except that the dispersion is graded with stronger nickel bonding to the stainless steel surface.

4.3.2.3 AMC148

AMC148 coating (AMCX, Lemont, PA) comprises of hard materials. In the process, the AMC148 surface is strongly diffusion bonded to the substrate. AMC148 is non-reactive and resistant to most chemicals, all known solvents, most bases, and with the exception of hot hydrofluoric, all acids as well. In fact, once applied, there is no practical method of removing the AMC148 surface from the substrate without the partial destruction of the substrate. AMC148-18 is essentially inert and non-reactive, and is suitable for use in acids, solvents and most liquids. It is hydrophobic, lipophobic, and has extremely low non-specific protein binding characteristics. Adhesion to stainless steel is better than Teflon. Its surface tension is small as Teflon. However, when there are scratches on Teflon surface, the surface tension goes up. That is advantage of AMC148, i.e., it is not easily removed from the surface.

4.3.2.4 Carbon Nanotube (CNT)

Carbon nanotubes are fullerene-related structures which consist of graphene cylinders closed at either end with caps containing pentagonal rings. These tubes were deposited onto the stainless steel 316 test specimen covering the central 10 mm x 10 mm portion due to cost constraints (GVD Corporation, Cambridge, MA).
4.4 Surface Characterization

Wetting phenomenon is important in understanding adhesion to surfaces. In cases such as adhesion, the behavior of drop on a solid surface is dependent on the three interfacial tensions between the solid, gas, and liquid phases as shown in the Figure 4.5.

![Force balance on three-phase interface](image)

Figure 4.5 Force balance on three-phase interface

In this study, goniometer (Figure 4.6 (a)) was used for contact angle (Figure 4.6 (b)) measurement. As mentioned in Chapter 3, goniometry involves the observation of a sessile drop of test liquid on a solid surface. Analysis of the shape of the drop that test liquid forms on the test solid is the basis for goniometry technique. The basic elements of a goniometer include a light source, sample stage, lens and image capture. Contact angle was found by measuring the angle formed between the solid and the tangent to the drop surface. (FTA1000-First Ten Angstroms, Portsmouth, VA)

![Schematic illustrating principle of goniometer and contact angle](image)

Figure 4.6 Schematic illustrating (a) principle of goniometer and (b) contact angle
4.5 Fouling Tests

4.5.1 Test Conditions

In this study, five surfaces were tested at two flow rates and two different inlet temperature conditions. Among these five surfaces, one of them was used for control purpose, stainless steel (SS-316), normally used in the industry. Other surfaces were selected considering their contact angle as it plays an important role in deposition on the surfaces. The typical flow rates for milk used in the dairy industry are about 4.66*10^{-5} m^3/s per channel. Taking the limitations of the test set up into account, worst case for fouling flow rates shown in Table 4.1 were chosen in this study. The inlet temperature for pasteurization is refrigeration temperature in the industry. However, in the experiments the inlet temperatures are much higher. The reason is that fouling occurs mostly after denaturation of the proteins. Test surfaces and conditions are summarized in Table 4.1.

Table 4.1. Experimental design for fouling test at different conditions.

<table>
<thead>
<tr>
<th>Surface type</th>
<th>Stainless steel 316 (control), Ni-P-PTFE (uniform), Ni-P-PTFE (TM117P), CNT, AMC148</th>
</tr>
</thead>
<tbody>
<tr>
<td>Milk flow rate in channel</td>
<td>3 and 10 g/s or 2.9 x 10^{-6} m^{3}/s and 9.8 x 10^{-6} m^{3}/s, respectively (corresponding to Re= 118 and Re=394)</td>
</tr>
<tr>
<td>Milk temperature at inlet of flow channel</td>
<td>40°C and 60°C</td>
</tr>
<tr>
<td>Top plate temperature</td>
<td>80°C</td>
</tr>
</tbody>
</table>

4.5.2 Test Procedure

1. The test plates were cleaned.

2. The thermocouples were inserted into the side wall holes as shown in Figure 4.4 (b), using thermal grease that can help for good contact with side walls, to measure the inside wall temperature. Thermocouple positions were secured using high temperature tape.
3. The flexible heaters, two on each side and 80 W each, were clamped down to the test section side walls and the test section was connected to the test set up.

4. The tank was filled with 7.57 L (2 gallons) of milk.

5. The pump was turned on and flow rate was fixed.

6. The heaters were turned on and the potentiometers were adjusted until the wall temperatures (both test plate and the other wall) reached 80°C. As constant heat source boundary condition was applied, the wall temperature varied from the inlet (bottom) to the outlet (top) of the test section. The test plate temperature of 80°C refers to the thermocouple value inserted in the top test specimen.

7. Water flow rate into the heat exchanger and the wall heat fluxes were adjusted manually until the desired test condition (inlet milk temperature and wall temperature) and steady state conditions were achieved.

8. Temperature at the inlet, outlet and on the test specimens were monitored through a midi logger (Graphtec, Santa Ana, CA) every 10 seconds until 6 hour of test period was completed.

9. After the steady state condition was achieved in about 10 minutes, the test was run for six hours continuously as cleaning is done every 6-8 hours for milk plants. On average 3.79 L (1 gallon) of milk in the tank was replaced with fresh milk every hour to prevent curdle formation due to change in acidity and microbial environment in the milk.

4.5.3 Measurements

After testing the surfaces for milk fouling for six hours, the surfaces were examined in terms of amount of deposit. The equipment was disassembled after operation to investigate the distribution and amount of the deposits. The amount of deposit was determined by weighing the plate before and after 6 hours following the test.
4.6 Numerical Modeling using Computer Software Package

Suitable hydrodynamic model for the equipment was basis for the simulation of the other processes occurring during thermal treatment. The hydro and thermal behavior in the heat exchanger and modeling of the transport phenomena is required for monitoring fouling dynamics.

4.6.1 Flow and Energy Model

Governing equations for 3-D unsteady flow are:

Continuity:
\[
\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x} (\rho u) + \frac{\partial}{\partial y} (\rho v) + \frac{\partial}{\partial z} (\rho w) = 0
\]  (4.1)

x-momentum:
\[
\frac{\partial}{\partial t} (\rho u) + u \frac{\partial}{\partial x} (\rho u) + v \frac{\partial}{\partial y} (\rho u) + w \frac{\partial}{\partial z} (\rho u) = -\frac{\partial P}{\partial x} + \frac{1}{\rho} \left( \frac{\partial^2}{\partial x^2} (\rho u) + \frac{\partial^2}{\partial y^2} (\rho u) + \frac{\partial^2}{\partial z^2} (\rho u) \right) + \rho g_x
\]  (4.2)

y-momentum:
\[
\frac{\partial}{\partial t} (\rho v) + u \frac{\partial}{\partial x} (\rho v) + v \frac{\partial}{\partial y} (\rho v) + w \frac{\partial}{\partial z} (\rho v) = -\frac{\partial P}{\partial y} + \frac{1}{\rho} \left( \frac{\partial^2}{\partial x^2} (\rho v) + \frac{\partial^2}{\partial y^2} (\rho v) + \frac{\partial^2}{\partial z^2} (\rho v) \right)
\]  (4.3)

z-momentum:
\[
\frac{\partial}{\partial t} (\rho w) + u \frac{\partial}{\partial x} (\rho w) + v \frac{\partial}{\partial y} (\rho w) + w \frac{\partial}{\partial z} (\rho w) = -\frac{\partial P}{\partial z} + \frac{1}{\rho} \left( \frac{\partial^2}{\partial x^2} (\rho w) + \frac{\partial^2}{\partial y^2} (\rho w) + \frac{\partial^2}{\partial z^2} (\rho w) \right)
\]  (4.4)

Energy:
\[
\rho C_p \left( \frac{\partial T}{\partial t} + u \frac{\partial T}{\partial x} + v \frac{\partial T}{\partial y} + w \frac{\partial T}{\partial z} \right) = k \left( \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} \right)
\]  (4.5)
where $\mu$ is the viscosity, $\rho$ is the density, $P$ is the pressure, $t$ is the time, $k$ is thermal conductivity, $C_p$ is heat capacity, and $u$, $v$, and $w$ are the velocity components in $x$, $y$, and $z$ directions, respectively. The coordinate system with respect to the flow channel is given in Figure 4.7. The 2-D form of the governing equations was solved in FLUENT. Initial and boundary conditions are;

**Initial condition**  \( @  \ t = 0 \quad T(x, y) = T_{in} \)

**Boundary conditions**  \( @  \ x = 0 \quad and \quad x = 5 \text{ mm} \quad T(t, y) = T_{wall} \)  
\( @  \ y = 0 \quad T = T_{in} \quad @  \ y = 184 \text{ mm} \quad T = T_{out} \)

Figure 4.7 Coordinate system of the test set up
4.6.2 Software

Heat transfer accompanies many fluid flow phenomena and FLUENT 6.0 (Fluent, 2003) offers a comprehensive suite of options for convection, conduction, and radiation. Other capabilities closely associated with heat transfer include models for cavitation, compressible liquids, heat exchangers, shell conduction, real gas, and wet steam. The straightforward addition of heat sources and a complete set of thermal boundary condition options round out the capabilities, making heat transfer modeling in FLUENT a reliable tool to meet the needs of this research. In this study, time-dependent two dimensional representations of equations (4.1)-(4.5) along with initial and boundary conditions were solved using FLUENT.

4.6.3 Model Parameters

Factors that play role in the performance of plate heat exchanger are included in the numerical formulation. The main independent parameters can be divided into design parameters and operational parameters. Surface properties, corrugation, plate height to length ratio, channel size are related to the design of the equipment. Whereas, input stream velocities, relative flow direction, number of transfer units employed in the process are operational factors.

4.6.4 Model Outputs

Velocity profile enables one to identify the eddy zones. These zones are also where the most fouling occurs. Temperature profile within the channels of PHE can be obtained by solving the energy equation numerically. As stated earlier, since fouling is temperature dependent, and is most intense when the temperature difference is more than approximately 10°C, temperature profile provides a good reference to compare with experimental data. Results of fouling tests with different surfaces enable to correlate surface properties with fouling. Test results, surface parameters, operational parameters, and fouling kinetics are included in the overall model.
4.7 Computational Methodology

Using the time dependent 2D formulation, the temperature variation in the channel, outlet temperature, and fouling amount were calculated for each test surface by using FLUENT. The mesh generator was GAMBIT. In addition to previous assumptions, the following assumptions were made for calculations:

- Wall temperature varies linearly. There were three thermocouples located on the wall surface (Figure 4.4(b)). The distance between each of them was known. In the FLUENT calculation linear variation of wall temperature was assumed.
- Milk properties do not change with time.
- It is assumed that there is symmetry with respect to center of the plate width (z direction) since there is no significant difference in fouling distribution.

4.7.1 Mesh Generation

Mesh generation was done using GAMBIT, the preprocessor of FLUENT. Dimensions of the test channel were given in the previous sections (Figure 4.4(b)). Plate length, 184 mm, and channel gap, 5 mm, were divided into 20 and 50 meshes, respectively. Since the variation of temperature in the direction of the flow is important only for the calculation of deposits on surfaces at different locations, 20 meshes were sufficient for calculating the deposits. Main variation in the temperature was found to be in the channel. Therefore, smaller mesh size was chosen for the gap. The mesh generation was repeated for different mesh sizes to get more accurate results with less computation time. In Chapter 5, sensitivity analysis for time and mesh size are presented and discussed.

4.7.2 Thermal Conductivity Calculations

Overall heat transfer coefficient was calculated using Eqn. 4.6 for each time interval in other words every 30 minutes.
\[ UA \left( T_{ave\ wall} - \frac{T_{in} + T_{out}}{2} \right) = \dot{m} C_p \left( T_{out} - T_{in} \right) \]  \hspace{1cm} (4.6)

Nusselt number, \( \text{Nu} \), was calculated using the wall temperature profile in FLUENT. Thermal conductivity, \( k = 0.62 \) W/mK, and channel gap, 5 mm, are known. Convective heat transfer coefficient, \( h \), was calculated using equation 4.7.

\[ h_{milk} = \frac{\text{Nu} \ k_{milk}}{\text{channel \ gap}} \]  \hspace{1cm} (4.7)

Thermal conductivity of foulant was calculated for each time interval along the plate using equation 4.8, which shows the resistance to heat transfer.

\[ \frac{1}{UA} = \frac{\lambda_{surface}}{k_{surface} A} + \frac{1}{h_{milk} A} + \frac{\lambda_{deposit}}{k_{deposit} A} \] \hspace{1cm} (4.8)

where \( U \) is overall heat transfer coefficient, \( h \) is convective heat transfer coefficient, \( k \) is conductive heat transfer coefficient, \( A \) is heat transfer area, \( m \) is milk flow rate, \( T \) is temperature, and \( \lambda \) is thickness. Subscripts in, out, surface, deposit denote for inlet, outlet, plate surface and deposit layer.

After calculating the thermal conductivity (\( k \)) of the deposit, a function describing the variation of \( k \) with time and distance along the plate was fit. A UDF (User Defined Function) for thermal conductivity and wall temperatures was developed in C and implemented to FLUENT.

4.7.7 FLUENT Calculations

4.7.7.1 Reading the Case File

The generated mesh was exported. A case file for the mesh was also copied. From the FLUENT menu the copied case file was read.

1. File \( \rightarrow \) Read case

After reading the file, grid check was done.
2. Grid → Check

Upto this point, this procedure is always same for any problem.

Next steps include defining the problem, operating and boundary conditions, exporting the UDF (User Defined Function) file for temperature profile at the wall and computing the surface Nusselt number that enables to find the convective heat transfer coefficient of milk in the channel with respect to channel length.

3. Define → Models → Solver. On the menu, 2D segregated implicit solution for unsteady state was marked.

4. Define → Models → Energy. Energy equation was selected.
5. Define ➔ Operating conditions. Ambient pressure was written as operating pressure. And the acceleration was in negative $y$ direction, $9.8 \text{ m/s}^2$.

6. Define ➔ User defined ➔ Functions ➔ Interpreted. UDF file was browsed and interpreted.

7. Define ➔ Materials. On the menu, material type was selected as fluid for milk and solid for test surface and deposit. The properties of milk were given in Chapter 3. The thermal properties of stainless steel in the FLUENT library were used. After calculation of Nusselt number, thermal conductivity ($k$) of deposit was calculated and a function was fit to $k$ as explained in the previous section.
8. Define Boundary conditions. Boundary conditions were set for interior as interior, for milk as fluid milk, for deposit walls as deposit, for interior and exterior walls of the channel as stainless steel. Inlet fluid velocity and temperature was set at this point. Outlet boundary condition is pressure outlet.
9. Solve → Initialize. Before proceeding to the computation, the solution was initialized.
10. Solve→Iterate. Time step size, number of time steps, and maximum iterations per time step was set.

11. File→Export. The temperature profile in the channel and outlet temperature was exported.

### 4.8. Calculation of Deposit Amount

Deposit amount calculation on each test surface with different contact angles was based on the change of thermal performance of the heat exchanger with time due to fouling. Overall heat transfer coefficient includes conductive and convective resistances.

\[
\frac{1}{U_{\text{clean}}} = \frac{1}{h} + \frac{\lambda_{\text{wall}}}{k_{\text{wall}}} \tag{4.9}
\]

When the surface is fouled, there is additional resistance \(\frac{\lambda_{\text{fouling}}}{k_{\text{fouling}}}\) due to this extra layer. Fouling resistance is often used as a parameter to monitor changes in performance. Therefore, overall heat transfer coefficient for fouled surface, \(U_f\), is expressed as;

\[
\frac{1}{U_f} = \frac{1}{h} + \frac{\lambda_{\text{wall}}}{k_{\text{wall}}} + \frac{\lambda_{\text{fouling}}}{k_{\text{fouling}}} \tag{4.10}
\]

\[
r_f = \frac{\lambda_{\text{fouling}}}{k_{\text{fouling}}} = \frac{1}{U_f} - \frac{1}{U_{\text{clean}}}
\]

where \(\lambda_{\text{wall}}\) and \(\lambda_{\text{deposit}}\) are thickness of the wall and the deposit, \(k_{\text{wall}}\) and \(k_{\text{deposit}}\) are the thermal conductivities of the wall and the deposit, \(U_{\text{clean}}\) and \(U_f\) are the overall heat transfer coefficient for clean and fouled surface, \(r_f\) is the total resistance due to fouling.

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Total heat transfer is;

\[ Q = U_{\text{clean}} A_{\text{clean}} \Delta T = U_f A_f \Delta T \]

(4.11)

\[ U_{\text{clean}} A_{\text{clean}} = U_f A_f \]

where \( A_f \) is the area required under fouled conditions and \( A_{\text{clean}} \) is the area required under clean conditions.

The fouling Biot number is a measure of the relative magnitude of fouling on a heat exchanger surface as described by Fryer and Slater (1985) and Somerscales et al. (1997).

\[ \frac{A_f}{A_{\text{clean}}} - 1 = \frac{U_{\text{clean}}}{U_f} - 1 = U_{\text{clean}} \left( \frac{1}{U_f} - \frac{1}{U_{\text{clean}}} \right) = U_{\text{clean}} (r_f)_\text{total} = B_i_f \]

(4.12)

where \( U_f \) and \( U_0 \) are heat transfer coefficients computed by FLUENT at time=360 min and at t=10 min, respectively.

When the fouling Biot number is large, the overall heat transfer resistance is dominated by fouling. On the other case, the overall heat transfer resistance is dominated by factors other than fouling.

The correction factor \( f(\theta) \) compares the fouling tendencies of the coated surfaces with stainless steel. It was found by using the experimental data. The experimentally found deposit mass was linearly correlated with contact angle. In this formulation, ratio of coated surface to the stainless steel was defined as \( f(\theta) \).

\[ \text{Fouling amount} = f(\theta) \left( \frac{\rho_{\text{deposit}} k_{\text{deposit}} B_i}{U_0} \right) \]

(4.13)

where \( f(\theta) \) is a correction factor function of contact angle \( \theta \), \( \rho_{\text{deposit}} \) and \( k_{\text{deposit}} \) are the density and thermal conductivity of the deposit, \( B_i \) is Biot number for fouling.
Chapter 5

RESULTS AND DISCUSSION

The goal of this work was to verify and propose an approach to mitigate fouling in dairy processing. As noted previously, fouling increases the cost, compromises the quality and safety, and leads to environmental issues (associated with CIP, Clean-In-Place) (Hauser, 1989). Therefore, four coatings with low surface energy were evaluated prior to their recommendation. In order to be able to test the hypothesis related to the effect of the surface properties on fouling, milk fouling was examined on differently treated surfaces in the bench scale test set-up. The surfaces tested and the test conditions are given in Chapter 4 (Table 4.1). Next step was computational modeling, which included: (1) calculation of the temperature profiles in the channel and the outlet temperatures for all surfaces and test conditions and (2) fouling amount on the surfaces by incorporating the fouling kinetics into the model. A relationship between the amount of deposits and contact angle was obtained by using the computational model with the physical properties of the test surfaces. In this way, amount of fouling for any test condition on any material with known contact angle could be estimated for bench scale tests.

In this chapter, effect of surface properties on fouling mitigation investigated in the bench scale parallel plate heat exchanger is presented and discussed. Under a variety of operating conditions, measured and computed temperature profiles for each test surface in the system, and foulants deposit amount on these surfaces are also given. Furthermore, the model developed including surface properties, fouling kinetics, and PHE dynamics that describes milk fouling in the bench scale plate heat exchanger is presented.
5.1 Experimental Results

The cost of investigating the effect of surface energy on fouling in plate heat exchangers was reduced by scaling-down to bench scale tester. There are benefits and drawbacks of the test set-up used for examining the fouling formation in plate heat exchanger. While designing the system, the simulation of thermal-hydraulic behavior of parallel plate heat exchanger was considered to be the first priority. However, certain assumptions, which have disadvantages as well as advantages, were made to simplify the system.

First, the need, significance, and advantages of the bench scale tester are summarized. Next, the drawbacks of the simplification are discussed.

Bench scale heat exchanger is advantageous in terms of the cost, acceptable accuracy, and ease of operation in performing experiments. Since available goniometer was not suitable to fit the full size plates of the heat exchanger, and the cost of coating whole plate is much higher than coating 2.5 cm x 2.5 cm test specimens, using small test surfaces that can be disassembled from the system was favored. Fouling was quantified by weighing the test specimens (2.5 cm x 2.5 cm) before and after the test (after 6 hours following the test). If the tests were performed in PHE, either there would be a need for a scale that could weigh full size plates with high sensitivity, to the extent of mg, or amount of deposit would have to be measured by scraping the foulants from the plates. In any case, the error would increase due to low sensitivity of the scale or loss of deposits during scraping. Temperature measurement is easier and more accurate for bench scale heat exchanger since the number of thermocouples needed is less. In other words, in plant-scale plate type heat exchangers, because of the need for more thermocouples due to their size and the practical difficulty of the thermocouples being inserted in the channels and mounted onto the plates, disturbance in the system is increased. Besides, measurement of plate temperature would be affected by the fluid (milk) flowing since thermocouples placed on the plates will be in contact with the stream. Moreover, the simplified geometry provided an opportunity for an accurate computational model including fundamental understanding of fouling kinetics.
In all cases, only the test specimen located at the top (Figure 4.4 (b)) was coated with low energy coating instead of whole plate for two reasons: to reduce the coating cost, and to examine the worst case scenario. Cost for coating the whole plate is almost twenty fold, \( \frac{184 \times 60 \, \text{mm}^2}{25.4 \times 25.4 \, \text{mm}^2} \approx 17 \). When entire plate is coated, deposits formed in the entrance (i.e., bottom) region will be less. As deposit amount decreases, resistance to heat transfer decreases. Thus, more heat is transferred to the fluid resulting in less temperature difference between the surface and the fluid with the same heat input. As stated in Chapter 3, fouling occurs most when the temperature difference between the wall (i.e., plate) and the milk is more, beyond denaturation temperature. Therefore, amount of deposits in actual practice is expected to be less than the measured values, i.e., the deposit amount over the effective area of the test specimens represented the worst case scenario.

In terms of the system drawbacks, milk was re-circulated in the test set-up in contrast to industrial PHE, in which it flows through once, i.e., is not recirculated. This both affects the amount of native \( \beta \)-lactoglobulin in the milk and the physical properties with time. Another reason for the variation of the milk properties is that the system is not totally a closed system. There was loss of water vapor from the milk tank resulting in higher density toward the end of operation vs. beginning of operation period. In addition to the change in density of the milk, viscosity increase might cause higher residence time of milk in the system. The level of the effect of physical property change was not examined in this study.

As mentioned in the Chapter 4, vitamin D enriched, pasteurized, whole milk purchased from a local grocery store was preheated in a pot on hot plate to the inlet temperature to reduce the time to reach steady state condition. That is to say, since the amount of the native protein in pasteurized milk is lower than unheated milk, due to the deposits during pasteurization, and there was fouling on the pot surface, the measured deposit amount would be more in pasteurizing raw milk in industrial scale plate heat exchanger. It is a known fact that vitamin D in milk is unaffected by pasteurization, boiling, or sterilization. Hence, fortification of milk with vitamin D was assumed to not
effect fouling during the experiments. Microbial safety and quality analyses of the milk
during or at the end of the test were not performed since the purpose of this study was
mainly to investigate fouling formation on the surfaces and mitigation of fouling. The
designed system was not suitable for these analyses as it did not provide sufficiently high
outlet milk temperature and residence time for pasteurization, was open to atmosphere,
i.e., prone to contamination, and pasteurized milk was re-circulated during testing.
Analysis to determine the contamination of milk with coating material could be suggested
after investigating the benefits of the coated surface in terms of fouling mitigation under
selected test conditions.

Smooth uncorrugated plane walls of bench scale tester were heated uniformly by
a constant heat source. On the other hand, to increase the heat transfer area, plates are
corrugated and for higher efficiency counter-flow is preferred in industrial PHEs. Due to
this discrepancy, temperature gradient between the hot fluid and milk in PHE, and wall
(i.e., plate) and milk in the bench scale heat exchanger would be different. For this
reason, fouling behavior is expected to have some similarities though not exactly the
same in these two cases.

In the following sections, surface characterization, and fouling on test surfaces,
stainless steel (SS316, control), Microlube/PTFE, AMC148, TM117P, and CNT are
presented. Surface characterization was done by measuring the contact angle between the
milk droplet and test surfaces. Fouling results are discussed in terms of change in heat
transfer resistance due to fouling and amount of deposits. Effect of flow rate and inlet
milk temperature on foulant formation is presented.

5.1.1 Contact Angle of Test Surfaces

The principle of the equipment used for contact angle measurement, goniometer,
is explained in Chapter 4. Table 5.1 shows the contact angle of test surfaces for fluid
whole milk. This data was used in the model that describes the amount of fouling on any
material with known surface properties. As expected, stainless steel (SS316) had the
smallest contact angle of 72° compared with coated surfaces. Of all the measured values,
AMC148 had the highest contact angle; therefore, is expected to have the smallest deposit amount.

Table 5.1 Contact angle of test surfaces with whole milk at room temperature

<table>
<thead>
<tr>
<th>Surface</th>
<th>Contact Angle</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stainless steel (SS316)*</td>
<td>72º</td>
</tr>
<tr>
<td>Microlube/PTFE</td>
<td>107º</td>
</tr>
<tr>
<td>TM117P</td>
<td>112º</td>
</tr>
<tr>
<td>AMC148</td>
<td>145º</td>
</tr>
<tr>
<td>CNT**</td>
<td>≈180º</td>
</tr>
</tbody>
</table>

*Control  
**Not measurable due to instability

Contact angle of CNT coated surface was not measurable by the goniometer due to the instability of the droplet on the surface. When the fluid droplet was placed on CNT, it was bouncing due to high contact angle between the liquid and the surface. Instead, a high speed camera was used to capture the falling and rolling of water droplet on the surface. Figure 5.1 shows the water droplet on the CNT coated surface. Water forms an almost spherical ball (static contact angle ≈ 180º) on the CNT coated surface (1cm x 1cm black area in the middle). The lower contact angles of the water droplets outside the CNT coated area are clearly noticeable.

Figure 5.1 Water droplet on Teflon® coated CNT surface

5.1.2 Temperature Profiles

The worst case for fouling formation was examined. For this purpose, the coated surfaces and the control surface were mounted at the top location of the test surface holder where fouling was expected to be most severe due to higher temperature (Figure 4.4b). In other words, as the comparison of deposit amount was done on the top surface, where the fouling occurred the most, the judgment would be more reliable vs. very little foulants near the bottom of the test plate. Time-temperature profiles are given in Figures 5.2 to 5.6 and discussed in ensuing paragraphs.
Figure 5.2 Temperature change with time for 40°C inlet milk temperature (a) 3 g/s, (b) 10 g/s for stainless steel, SS316 control surface
Figure 5.3 Temperature change with time for 40°C inlet milk temperature (a) 3 g/s, (b) 10 g/s for Microlube/PTFE coated surface
Figure 5.4 Temperature change with time for 40°C inlet milk temperature (a) 3 g/s, (b) 10 g/s for TM117P coated surface.
Figure 5.5 Temperature change with time for 40°C inlet milk temperature (a) 3 g/s, (b) 10 g/s for AMC148
Temperature profiles in the system and inlet and outlet milk temperatures when using control, Microlube/PTFE, TM117P, AMC 148, and CNT coated test surfaces are displayed in Figures 5.2 to 5.6, respectively. These figures include the inlet (T$_{in}$) and outlet (T$_{out}$) temperature variation with time of milk. In addition, temperature variation of walls at the three locations (T$_{top}$, T$_{middle}$, and T$_{bottom}$) are shown in these figures, which were measured at the mid position of test specimens (Figure 4.4 (b)). As noted previously, the CNT coated specimen could be used only once since it was not possible to clean the deposits without damaging and/or dislodging the CNT tubes. Therefore, it was decided to run the fouling trial at 40ºC and 3 g/s.

The variation in outlet milk temperature was because of flow rate fluctuation. Fluctuation at low flow rate was more since the rotameter control, while acceptable, was not nearly as good as it was at high flow rate. Additionally, the variation in the wall temperatures with time was attributed to changes in inlet milk temperature and manual control of heat input (due to cost considerations an automated system was not installed).
Temperature profiles show that there was first increase in the surface and outlet milk temperatures. In about 10-15 minutes, the system reached steady-state and surface temperatures did not vary much. However, outlet milk temperature decreased gradually with time. This was due to fouling formation on the surface. As the surface was fouled, the heat transfer resistance increased and the milk was not heated as much as it was at the beginning. Tables 5.2 to 5.5 summarize increase in temperature ΔT (outlet temperature-inlet temperature) for different conditions and surfaces. As shown in section 5.1.14 Fouling of Surfaces, when there is more fouling, it is expected that outlet milk temperature drop will be more. In Figures 5.2 to 5.6, the slopes of the curves of outlet milk temperature are different for this reason.

Table 5.2 Average temperature (±SD) and ΔT after the system reached steady state for flow rate of 3 g/s and inlet milk temperature 40°C

<table>
<thead>
<tr>
<th></th>
<th>Control</th>
<th>Microlube/PTFE</th>
<th>TM117P</th>
<th>AMC148</th>
<th>CNT</th>
</tr>
</thead>
<tbody>
<tr>
<td>T_{bottom} (ºC)</td>
<td>60.98±0.97</td>
<td>73.12±0.74</td>
<td>72.53±0.48</td>
<td>71.87±0.52</td>
<td>71.61</td>
</tr>
<tr>
<td>T_{middle}(ºC)</td>
<td>74.44±0.95</td>
<td>75.97±1.05</td>
<td>76.22±0.56</td>
<td>77.17±0.57</td>
<td>74.31</td>
</tr>
<tr>
<td>T_{top} (ºC)</td>
<td>80.31±0.18</td>
<td>80.58±1.13</td>
<td>80.27±0.42</td>
<td>80.27±0.41</td>
<td>80.21</td>
</tr>
<tr>
<td>T_{in} (ºC)</td>
<td>40.31±0.26</td>
<td>40.35±0.63</td>
<td>40.16±0.24</td>
<td>40.09±0.22</td>
<td>40.07</td>
</tr>
<tr>
<td>T_{out}(ºC)</td>
<td>65.24±0.64</td>
<td>66.37±0.37</td>
<td>65.30±0.53</td>
<td>65.47±0.21</td>
<td>63.50</td>
</tr>
<tr>
<td>ΔT, T_{out}-T_{in}</td>
<td>24.93±0.69</td>
<td>26.02±0.73</td>
<td>25.14±0.58</td>
<td>25.38±0.30</td>
<td>23.43</td>
</tr>
</tbody>
</table>

Table 5.3 Average temperature (±SD) and ΔT after the system reached steady state for flow rate of 10 g/s and inlet milk temperature 40°C

<table>
<thead>
<tr>
<th></th>
<th>Control</th>
<th>Microlube/PTFE</th>
<th>TM117P</th>
<th>AMC148</th>
</tr>
</thead>
<tbody>
<tr>
<td>T_{bottom} (ºC)</td>
<td>68.76±1.11</td>
<td>69.61±2.23</td>
<td>69.96±0.87</td>
<td>66.21±1.26</td>
</tr>
<tr>
<td>T_{middle}(ºC)</td>
<td>76.56±0.65</td>
<td>77.26±0.47</td>
<td>75.24±0.46</td>
<td>76.21±0.49</td>
</tr>
<tr>
<td>T_{top} (ºC)</td>
<td>80.26±0.22</td>
<td>80.41±0.38</td>
<td>80.32±0.29</td>
<td>80.21±0.46</td>
</tr>
<tr>
<td>T_{in}(ºC)</td>
<td>40.26±0.14</td>
<td>40.12±0.35</td>
<td>40.22±0.19</td>
<td>40.34±0.36</td>
</tr>
<tr>
<td>T_{out}(ºC)</td>
<td>48.11±0.30</td>
<td>48.43±0.21</td>
<td>49.23±0.31</td>
<td>48.17±0.22</td>
</tr>
<tr>
<td>ΔT, T_{out}-T_{in}</td>
<td>7.85±0.33</td>
<td>8.31±0.41</td>
<td>9.01±0.36</td>
<td>7.83±0.42</td>
</tr>
</tbody>
</table>
Table 5.4 Average temperature (±SD) and ΔT after the system reached steady state for flow rate of 3 g/s and inlet milk temperature 60°C

<table>
<thead>
<tr>
<th></th>
<th>Control</th>
<th>Microlube/PTFE</th>
<th>TM117P</th>
<th>AMC148</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_{\text{bottom}}$(ºC)</td>
<td>72.46±0.67</td>
<td>73.06±0.73</td>
<td>72.73±0.61</td>
<td>72.77±0.52</td>
</tr>
<tr>
<td>$T_{\text{middle}}$(ºC)</td>
<td>76.87±0.42</td>
<td>78.26±0.69</td>
<td>75.64±1.27</td>
<td>75.15±0.70</td>
</tr>
<tr>
<td>$T_{\text{top}}$(ºC)</td>
<td>80.52±0.30</td>
<td>80.43±0.26</td>
<td>80.18±0.60</td>
<td>80.19±0.48</td>
</tr>
<tr>
<td>$T_{\text{in}}$(ºC)</td>
<td>60.30±0.66</td>
<td>60.46±0.36</td>
<td>60.15±0.54</td>
<td>60.28±0.52</td>
</tr>
<tr>
<td>$T_{\text{out}}$(ºC)</td>
<td>71.81±0.16</td>
<td>72.82±0.16</td>
<td>71.65±0.21</td>
<td>72.27±0.26</td>
</tr>
<tr>
<td>ΔT, T_{\text{out}}-T_{\text{in}}</td>
<td>11.51±0.68</td>
<td>12.36±0.39</td>
<td>11.50±0.58</td>
<td>11.99±0.58</td>
</tr>
</tbody>
</table>

Table 5.5 Average temperature (±SD) and ΔT after the system reached steady state for flow rate of 10 g/s and inlet milk temperature 60°C

<table>
<thead>
<tr>
<th></th>
<th>Control</th>
<th>Microlube/PTFE</th>
<th>TM117P</th>
<th>AMC148</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_{\text{bottom}}$(ºC)</td>
<td>73.02±0.24</td>
<td>76.03±0.65</td>
<td>71.32±0.48</td>
<td>74.36±0.96</td>
</tr>
<tr>
<td>$T_{\text{middle}}$(ºC)</td>
<td>77.47±0.67</td>
<td>77.87±0.45</td>
<td>75.56±0.36</td>
<td>76.87±0.67</td>
</tr>
<tr>
<td>$T_{\text{top}}$(ºC)</td>
<td>80.34±0.29</td>
<td>80.41±0.22</td>
<td>80.21±0.18</td>
<td>80.22±0.34</td>
</tr>
<tr>
<td>$T_{\text{in}}$(ºC)</td>
<td>60.25±0.57</td>
<td>60.14±0.26</td>
<td>60.22±0.31</td>
<td>60.36±0.45</td>
</tr>
<tr>
<td>$T_{\text{out}}$(ºC)</td>
<td>67.19±0.26</td>
<td>66.09±0.21</td>
<td>66.93±0.35</td>
<td>66.83±0.15</td>
</tr>
<tr>
<td>ΔT, T_{\text{out}}-T_{\text{in}}</td>
<td>6.94±0.63</td>
<td>5.95±0.33</td>
<td>6.71±0.47</td>
<td>6.47±0.47</td>
</tr>
</tbody>
</table>

The surface based on which the comparison was made is stainless steel SS316 since it is the one currently used in the industry. In Tables 5.2 to 5.5, first three rows show the average temperature, at the center of the three test specimens inserted into the test plate, of three replications along with standard deviation for 3 g/s at 40°C, 10 g/s at 40°C, 3 g/s at 60°C, and 10 g/s at 60°C, respectively. In fourth and fifth rows, average inlet and outlet milk temperatures (of three replications) and standard deviation values are displayed. The values in Tables 5.2 to 5.5 are discussed further in the following subsections.
5.1.2.1 Effect of Mass Flow Rate

Figures 5.2(a) to 5.6(a) show typical temperature profiles for flow rate 3 g/s and inlet milk temperature 40°C when using control, Microlube/PTFE, TM117P, AMC 148, and CNT coated test surfaces, respectively. These figures include the inlet and outlet temperature change of milk and temperature change of wall at the three points, (T_top= Top plate, T_middle= Middle plate, and T_bottom= Bottom plate, Figure 4.4 (b)), each were measured at the mid position of test specimens. When the inlet milk temperature was 60°C at the same flow rate the behavior of the temperature profiles was similar. Typical temperature profiles during the test time (6 hours) for the condition 40°C inlet milk temperature and 10 g/s flow rate are shown in Figures 5.2 (b) to 5.6 (b).

For higher flow rate (10 g/s) change in milk temperature through the channel was 70% (Tables 5.2 and 5.3) and 55% (Tables 5.4 and 5.5) less for inlet milk temperature of 40°C and 60°C, respectively, compared to 3 g/s. As can be noted from equation 5.1, for a given input heat flux (as was for these tests) when the fluid flow rate is high, fluid temperature change is less.

\[ Q = mC_p \Delta T \]

where \( Q \) is the heat flux, \( m \) is the mass flow rate, \( C_p \) is the specific heat, and \( \Delta T \) is the temperature difference between outlet and inlet fluid.

Even though the heat input was controlled, the goal was to maintain the top wall temperature to be constant at 80°C (i.e., invariant with time) throughout the experimental runs. Therefore, based on the experimental data, the boundary condition in this study was approximately linear wall temperature; i.e., from inlet (bottom) to outlet (top) of bench scale tester. Equation 5.1 explains the change of milk temperature being less at higher flow rate. For this reason, the \( \Delta T \) change is not the same at different milk flow rates.

5.1.2.2 Effect of Inlet Milk Temperature

When the inlet milk temperature was 40°C, the heat transfer rate was higher compared to 60°C (Tables 5.2 vs. 5.4, and 5.3 vs. 5.5). This can be explained by the
difference between the surface temperature and the average milk temperature. The relationship between wall temperature and bulk temperature, i.e., average milk temperature, can be written as:

\[ Q = UA\Delta T \] (5.2)

where \( Q \) is the heat transfer rate, \( U \) is the overall heat transfer coefficient, \( \Delta T \) is the temperature gradient between wall and the bulk fluid. From equations 5.1 and 5.2:

\[ mCp(T_{out} - T_{in}) = UA\left[T_{wall} - \left(\frac{T_{out} + T_{in}}{2}\right)\right] \] (5.3)

In this study, the \( T_{wall} \) temperature at the top location was nearly constant throughout the experimental duration.

In Tables 5.2 to 5.5 the last row shows the increase in temperature of the milk as it flowed through the heating channel. As stated, at any flow rate the reason for milk to be less heated at higher inlet temperature is the decrease in the driving force for heat transfer. In other words, the temperature gradient between the wall and milk is less when the milk enters the channel at a higher temperature hence it is heated less. At flow rate of 3 g/s, comparison of \( \Delta T \) values for inlet milk temperature of 40°C (≈+25°C) and 60°C (≈+12°C) for each test surface indicates that there is \( \approx 50\% \) decrease in \( \Delta T \) for 60°C. At higher flow rate, 10 g/s, the same trend is observed; for the inlet milk temperature of 40°C, \( \Delta T \) is approximately +8.5°C; whereas, for 60°C \( \Delta T \) is approximately +6.5°C resulting in \( \approx 30\% \) decrease in \( \Delta T \) at higher inlet milk temperature (60°C). This shows that, effect of inlet milk temperature on the thermal performance changes with flow rate. At higher flow rate this effect is less, i.e., temperature difference between the outlet and inlet milk temperature is reduced by 30% when the inlet milk temperature is increased from 40°C to 60°C at 10 g/s flow rate compared (Table 5.3 vs. Table 5.5) to 50% decrease at 3 g/s flow rate (Table 5.2 vs. Table 5.4).
5.1.3 Outlet Milk Temperature

The outlet milk temperature is different after steady state was reached for different test conditions since milk is heated less at higher flow rate due to shorter residence time and at higher inlet milk temperature because of the smaller temperature difference between the surface and the milk. Final outlet temperature was not chosen for comparison purpose because the initial outlet temperature (i.e., following the steady state condition) was different for different surfaces and test conditions. For this reason, the comparison based on the difference between the outlet temperature at the end of six hours and at the start of the test is more meaningful.

In Table 5.6 decrease in outlet milk temperature after 6 hours of test time is shown. In all cases decrease in outlet milk temperature for stainless steel is the most and AMC148 and CNT are the least. This is related with the change in thermal performance of the heat exchanger during the test time. As denaturated proteins attach to the walls, they form a layer which in turn reduces the overall heat transfer coefficient resulting in reduced thermal performance. Change in outlet milk temperature decreases with increase in contact angle between the surface and milk (180°- Contact Angle of Test Surfaces, as shown in Table 5.1), i.e., for coated surfaces thermal performance of the system is not as much affected as stainless steel. This is further discussed in the following subsection.

Table 5.6 Decrease in outlet milk temperature in °C (experimental) after 6 hours of test time

<table>
<thead>
<tr>
<th></th>
<th>Control (108°)*</th>
<th>Microlube/PTFE (73°)*</th>
<th>TM117P (68°)*</th>
<th>AMC148 (35°)*</th>
<th>CNT (~0°)*</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 g/s, 40°C</td>
<td>10.3</td>
<td>7.2</td>
<td>6.9</td>
<td>6.7</td>
<td>4.8</td>
</tr>
<tr>
<td>3 g/s, 60°C</td>
<td>6.6</td>
<td>6.3</td>
<td>6.6</td>
<td>4.2</td>
<td>ND**</td>
</tr>
<tr>
<td>10 g/s, 40°C</td>
<td>5.6</td>
<td>4.9</td>
<td>5.2</td>
<td>2.8</td>
<td>ND**</td>
</tr>
<tr>
<td>10 g/s, 60°C</td>
<td>4.4</td>
<td>3.9</td>
<td>3.8</td>
<td>2.0</td>
<td>ND**</td>
</tr>
</tbody>
</table>

*180°-Contact Angle of Test Surfaces (Table 5.1)

**not determined
5.1.4 Fouling of Surfaces

In Table 5.7, average amount of deposits in grams on the control and coated surfaces are shown for different test conditions. The values for amount of fouling were average of three replicates.

As expected, fouling occurred the most on the stainless steel surface. It was lower for Microlube/PTFE and TM117P. CNT and AMC148 coated surfaces had the least amount of fouling. The differences in fouling amount were due to the contact angle of milk with the surface. In Table 5.1, contact angle of milk droplet on the test surfaces are given. High contact angle (i.e. low surface energy) between milk and these coated surfaces reduced the deposit amount as given in Table 5.7. Stainless steel has the lowest contact angle, 72°, which means it is more prone to fouling. As mentioned in Chapter 3, delaying the initial deposition of the denaturated protein onto the surface is crucial as once the surface is fouled it becomes more easy for other particles, in this case aggregated protein, to attach to the wall. When coated surfaces were used, the deposit amount decreased by large amounts; especially for surfaces having very high contact angle, i.e., AMC148 (145°) and CNT (≈180°) approximately 90% reduction in fouling amount.

Table 5.7 Amount of deposit in grams (±SD) on different surfaces for different conditions and flow rate variation (top surface temperature was 80°C)

<table>
<thead>
<tr>
<th></th>
<th>Control</th>
<th>Microlube/PTFE</th>
<th>TM117P</th>
<th>AMC148</th>
<th>CNT</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 g/s, 40°C</td>
<td>0.216±0.010</td>
<td>0.171±0.003</td>
<td>0.165±0.011</td>
<td>0.031±0.011</td>
<td>0.021</td>
</tr>
<tr>
<td>3 g/s, 60°C</td>
<td>0.205±0.057</td>
<td>0.065±0.008</td>
<td>0.056±0.005</td>
<td>0.021±0.007</td>
<td>ND**</td>
</tr>
<tr>
<td>10g/s, 40°C</td>
<td>0.220±0.007</td>
<td>0.162±0.003</td>
<td>0.144±0.014</td>
<td>0.054±0.006</td>
<td>ND**</td>
</tr>
<tr>
<td>10g/s, 60°C</td>
<td>0.111±0.020</td>
<td>0.089±0.011</td>
<td>0.097±0.011</td>
<td>0.009±0.003</td>
<td>ND**</td>
</tr>
</tbody>
</table>

*CNT coating was on 1 cm² area, and amount of fouling was multiplied with correction factor to be able compare with other surfaces; fouled area for other surfaces 2.54 cm x 2.54 cm = 6.45 cm²

Amount of foulants deposited was more (approximately two-fold) when inlet milk temperature was 40°C for all surfaces. Because the difference between the surface and the milk temperature was higher when the inlet temperature was 40°C compared to 60°C.
In other words, it was observed that the amount of deposit was more for larger temperature difference as stated in the literature that the larger the temperature difference between the surface and the fluid, the greater the fouling is. For the control surface, at a flow rate of 3 g/s and inlet temperature of 60°C deposit amount was very close to the condition at the same flow rate at 40°C inlet temperature (0.205 g vs. 0.216 g). It was because of variation of the deposit amount in three tests performed for 60°C. It is shown in Table 5.7 that standard deviation for this case, 0.057, was higher compared to the other cases. The deposit amount on AMC148 coated surface is more sensitive to temperature change at 10 g/s. The deposit amount at this flow rate for inlet milk temperature of 60°C is 85% less than for inlet milk temperature of 40°C, whereas for other surfaces it is approximately 50% less. This is mainly because of the deposit formation pattern on AMC148 coated surface, and sensitivity of the measurement scale. The proteins deposited mostly near the border of the AMC148 test specimen and stainless steel test plate (Figure 5.8 (c)), where the flow disturbance effect may take place. Sensitivity of the measurement scale, 1 mg, becomes important for this small deposit amount (9 mg).

There was not a significant effect of flow rate on fouling at these conditions. If flow rate was higher, effect of shear forces would have been observed particularly for surfaces with less deposits, i.e., coated surfaces. At higher flow rate, stronger shear forces would sweep away the foulants more from the surface with lower surface energy since deposits do not as strongly adhere on the coated surfaces, especially CNT and AMC148, when compared with the control surface.

Table 5.8 shows the percent decrease in deposit amount for the tested surfaces compared to the control surface. Microlube/PTFE, and TM117P have approximately same reduction in foulant amount for each test condition, contact angles of which are 107° and 112°, respectively. Except for the condition where the flow rate is 3 g/s and 60°C, percent deposit amount reductions for these surfaces are approximately 20%. AMC148 and CNT coated surfaces, with contact angles 145°and 180°, respectively, showed better potential for mitigating fouling, where the percent foulant reduction is about 90%. AMC148 has advantages over CNT. Most importantly, CNT is not suitable
for applications in the food industry. In addition, most of the fouling deposit on AMC148 was mainly at the boundary of the test specimen not on the surface itself (Figure 5.8 (c)). Coated surfaces were inserted into the stainless steel test specimen holder. Since contact angle of stainless steel is much lower than AMC148, when milk adheres onto the stainless steel side of SS316-holder interface, foulants accumulated on it would also attach to the neighboring coated surface. Therefore, when the entire wall is coated with AMC148, it is anticipated that percent decrease in fouling is expected to be higher than that noted in Table 5.8.

Table 5.8 Percent decrease in fouling compared to the control surface, stainless steel SS316

<table>
<thead>
<tr>
<th></th>
<th>Microlube/PTFE</th>
<th>TM117P</th>
<th>AMC148</th>
<th>CNT</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 g/s, 40°C</td>
<td>20.8</td>
<td>23.6</td>
<td>86.6</td>
<td>90.2</td>
</tr>
<tr>
<td>3 g/s, 60°C</td>
<td>68.9</td>
<td>72.6</td>
<td>87.8</td>
<td>ND**</td>
</tr>
<tr>
<td>10 g/s, 40°C</td>
<td>26.4</td>
<td>34.5</td>
<td>75.4</td>
<td>ND**</td>
</tr>
<tr>
<td>10 g/s, 60°C</td>
<td>19.8</td>
<td>12.6</td>
<td>92.7</td>
<td>ND**</td>
</tr>
</tbody>
</table>

There is a relation between deposit amount and change in outlet milk temperature (Table 5.6 and 5.8). For a given condition, change in outlet milk temperature during 6 hours is less for surfaces with less deposit on it. When the foulant amount is more, as on the control surface compared to the coated surfaces, the deposit thickness and hence the conductive resistance of foulant layer for heat transfer is increased. This results in less heat transfer when the amount of fouling is more. Consequently, milk was not heated as it was at the beginning, i.e., the thermal performance reduces the most when the deposit amount is higher. In section 5.2 Comparison of Computational and Experimental Results, how foulant amount can be predicted using the model based on this observation without performing experiments is discussed.

Cleaning of the fouled surfaces that have higher contact angle was much easier, i.e., there was no need for cleaning agents. This is very crucial in the industry as aggressive cleaning agents are used for cleaning of stainless steel plates of heat exchanger. These harsh chemical substances create environmental problems. In addition,
the cost of cleaning will be reduced as less chemicals will be needed, and the surfaces will not need to be cleaned as often. This also increases the overall capacity of the plant and prevents production losses.

5.1.5 Fouling Pictorial Observations

In Figures 5.7 and 5.8, fouling on the tested surfaces are shown. Fouling increases from bottom to top since top plate temperature was highest (Figure 5.7 (a)). For the surfaces with less fouling, the deposits show bubble like structure. When the contact with the surface is less (large contact angles) the fluid does not spread on the surface totally. Therefore, deposits would form like bubbles on the surface. For CNT coated surface, the surface is coated only at the center with CNT. Therefore, fouling at the center was considered for comparison.

Integrity of the coated surfaces except for CNT did not change during and after tests though some discoloration occurred on Teflon based coatings, i.e., Microlube/PTFE and TM117P, which is thought to be mainly caused by overheating and improper cleaning.

Figure 5.7 Fouling on the control surface (a) entire side wall of flow channel and all the test specimens (b) top specimen
5.2 Comparison of Computational and Experimental Results

This section covers the sensitivity analysis of the system to grid size and time step, comparison and discussion of computed temperature profiles in the channel of bench scale tester, the outlet temperatures of milk, and the foulants deposits on the test surfaces for different test cases studied. Thereafter, the reliability and applicability of the proposed fouling model is interpreted.

5.2.1 Sensitivity and Stability Analysis

In the computational studies, mesh size and time step are very important in terms of the accuracy and the reliability of the computed values. Having accurate results in a short computation time is important for the benefit of the user.

The dimensions of the bench scale tester (5 mm channel width x 184 mm in length), and the flow direction (from bottom to top) in the channel are displayed in Figure 4.4 (a). Grid number in the flow direction (y) was chosen as 20 after trials with 10, 20, 50, and 100. In the selection of the mesh size in y-direction, representing the wall temperature profile was the criteria with shorter computation time. In the selection of mesh size in the width of the channel (x-direction), affect of wall and boundary layer thicknesses was considered. After trials with 5, 10, 20, 50, and 100, the mesh number in x-direction was selected as 50; corresponding to mesh size of 0.1 mm. In Figure 5.9, generated mesh for the computation is given.
As an example of the sensitivity of the results to mesh size, the outlet milk temperatures at the end of 6 hours for the control surface at flow rate of 3 g/s and inlet milk temperature of 40°C were for minimum number of meshes (5x10), the selected case, (50x20), and maximum number of meshes (100x100) are 56.55°C (2.4% difference), 55.24°C (0.05%), and 55.21°C (0%, reference grid), respectively. There is not much difference in outlet milk temperature (~0.05%) as the grid points increased more than the selected case (Figure 5.10 and Table 5.9).

Table 5.9 Mesh size sensitivity of outlet milk temperature

<table>
<thead>
<tr>
<th>Mesh size</th>
<th>Outlet milk temperature, °C</th>
<th>% difference with respect to 100*100 mesh</th>
</tr>
</thead>
<tbody>
<tr>
<td>5*10</td>
<td>56.55</td>
<td>2.42</td>
</tr>
<tr>
<td>10*10</td>
<td>55.95</td>
<td>1.34</td>
</tr>
<tr>
<td>20*20</td>
<td>55.47</td>
<td>0.47</td>
</tr>
<tr>
<td>50*20</td>
<td>55.24</td>
<td>0.05</td>
</tr>
<tr>
<td>100*100</td>
<td>55.21</td>
<td>0.00</td>
</tr>
</tbody>
</table>
Since the FLUENT formulation is fully implicit, there was no stability criterion that needs to be met in determining the time step size. The choice of the time step was based on the number of iterations FLUENT needed to converge at each time step. As stated in the User Manual (Fluent, 2003) the ideal number of iterations per time step is between 10 and 20. When the number of iterations for the selected time step was few, the time step size was increased. In addition, initially there was a need for selecting the time step smaller to represent the transient stage. For this reason, for initial 30 minutes, the time step was selected as 5 seconds, and after 30 minutes, it was 60 seconds.

### 5.2.2 Outlet Milk Temperature

In Figure 5.11, computed versus measured outlet milk temperature is shown for a sample case, inlet milk temperature of 40°C and flow rate of 3 g/s. The slope of the line shows that there is good agreement between the measured and computed values. The ideal line should have slope of 1 with $R^2$ of 1; for this case the slope is 0.995 with $R^2$ of 0.96. For the other test cases and conditions, the slopes of the linear fit and the square of correlation coefficient ($R^2$) are summarized in the Table 5.10. Slopes for all the cases

![Figure 5.10 Outlet milk temperature for different mesh sizes for stainless steel (3 g/s flow rate, and 40°C inlet milk temperature)](image-url)
varied from 0.994 to 1.008 with $R^2$ from 0.87 to 0.99, showing very good to excellent prediction capability of the computational model.

![Graph showing linear relationship with $y = 0.995x$ and $R^2 = 0.9632$](image)

Figure 5.11 Comparison of computed and measured outlet milk temperature during the test for the control surface at 3 g/s and 40°C

Table 5.10 Slope and $R^2$ of the linear line fit for computed and measured outlet milk temperature

<table>
<thead>
<tr>
<th></th>
<th>Control</th>
<th>Microlube/PTFE</th>
<th>TM117P</th>
<th>AMC148</th>
<th>CNT</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 g/s, 40°C</td>
<td>0.995</td>
<td>1.008</td>
<td>0.996</td>
<td>1.002</td>
<td>1.005</td>
</tr>
<tr>
<td></td>
<td>0.96</td>
<td>0.99</td>
<td>0.87</td>
<td>0.98</td>
<td>0.94</td>
</tr>
<tr>
<td>3 g/s, 60°C</td>
<td>1.007</td>
<td>0.997</td>
<td>0.996</td>
<td>0.994</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>0.94</td>
<td>0.90</td>
<td>0.88</td>
<td>0.87</td>
<td></td>
</tr>
<tr>
<td>10 g/s, 40°C</td>
<td>0.997</td>
<td>1.020</td>
<td>1.010</td>
<td>1.003</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>0.97</td>
<td>0.99</td>
<td>0.99</td>
<td>0.987</td>
<td></td>
</tr>
<tr>
<td>10 g/s, 60°C</td>
<td>1.006</td>
<td>1.006</td>
<td>1.006</td>
<td>1.001</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>0.96</td>
<td>0.95</td>
<td>0.94</td>
<td>0.96</td>
<td></td>
</tr>
</tbody>
</table>

Outlet milk temperatures at the end of 6 hours are given for all the test conditions and test surfaces in Table 5.11. In all but five cases, the computed temperatures lie in the 95% Confidence Interval (+/- 2SD) of the experimental values. The outliers are italicized in Table 5.11. The maximum percent difference is 1.25% for TM117P coated surface at 3 g/s, 40°C. This shows again that a good representation of the system was achieved by the computational model.
Table 5.11 Outlet milk temperature (±SD) at the end of 6 hours for the test cases

<table>
<thead>
<tr>
<th>Test condition</th>
<th>Control</th>
<th>Microlube/PTFE</th>
<th>TM117P</th>
<th>AMC148</th>
<th>CNT</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 g/s, 40°C</td>
<td>54.93±0.06</td>
<td>57.27±0.32</td>
<td>58.37±0.35</td>
<td>57.73±0.25</td>
<td>58.93</td>
</tr>
<tr>
<td></td>
<td>55.24</td>
<td>57.38</td>
<td>59.10</td>
<td>57.94</td>
<td>58.40</td>
</tr>
<tr>
<td>10 g/s, 40°C</td>
<td>42.60±0.20</td>
<td>43.43±0.15</td>
<td>44.03±0.15</td>
<td>45.28±0.10</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>42.45</td>
<td>43.91</td>
<td>44.30</td>
<td>45.50</td>
<td>ND</td>
</tr>
<tr>
<td>3 g/s, 60°C</td>
<td>65.23±0.29</td>
<td>66.53±0.15</td>
<td>65.30±0.56</td>
<td>68.95±0.12</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>65.49</td>
<td>66.72</td>
<td>65.56</td>
<td>68.05</td>
<td>ND</td>
</tr>
<tr>
<td>10 g/s, 60°C</td>
<td>62.77±0.51</td>
<td>62.37±1.98</td>
<td>63.40±0.18</td>
<td>64.70±0.20</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>63.30</td>
<td>62.60</td>
<td>62.85</td>
<td>64.82</td>
<td>ND</td>
</tr>
</tbody>
</table>

5.2.3 Change in Outlet Milk Temperature

In Table 5.12 change (i.e., decrease) in outlet milk temperatures during the test time, i.e., the difference between the outlet temperature at the end of six hours and at the start of the test, are presented. Outlet milk temperature comparison based on this difference is more meaningful since it is different for each surface and test condition.

Table 5.12 Change (i.e., decrease) in outlet milk temperature for the test cases

<table>
<thead>
<tr>
<th>Test condition</th>
<th>Control</th>
<th>Microlube/PTFE</th>
<th>TM117P</th>
<th>AMC148</th>
<th>CNT</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 g/s, 40°C</td>
<td>10.3</td>
<td>7.2</td>
<td>6.9</td>
<td>6.7</td>
<td>4.8</td>
</tr>
<tr>
<td></td>
<td>10.5</td>
<td>7.9</td>
<td>7.2</td>
<td>6.8</td>
<td>6.2</td>
</tr>
<tr>
<td>% difference</td>
<td>+1.9</td>
<td>+9.7</td>
<td>+4.4</td>
<td>+1.49</td>
<td>+29.2</td>
</tr>
<tr>
<td>10 g/s, 40°C</td>
<td>6.6</td>
<td>6.3</td>
<td>6.6</td>
<td>4.2</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>6.9</td>
<td>6.2</td>
<td>6.8</td>
<td>4.4</td>
<td>ND</td>
</tr>
<tr>
<td>% difference</td>
<td>+4.5</td>
<td>-1.6</td>
<td>+3.0</td>
<td>+4.8</td>
<td>ND</td>
</tr>
<tr>
<td>3 g/s, 60°C</td>
<td>5.6</td>
<td>4.9</td>
<td>5.2</td>
<td>2.8</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>6.4</td>
<td>5.2</td>
<td>5.4</td>
<td>3.2</td>
<td>ND</td>
</tr>
<tr>
<td>% difference</td>
<td>+14.3</td>
<td>+6.1</td>
<td>+3.8</td>
<td>+14.3</td>
<td>ND</td>
</tr>
<tr>
<td>10 g/s, 60°C</td>
<td>4.4</td>
<td>3.9</td>
<td>5.2</td>
<td>2.0</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>4.6</td>
<td>4.6</td>
<td>4.4</td>
<td>2.2</td>
<td>ND</td>
</tr>
<tr>
<td>% difference</td>
<td>+4.5</td>
<td>+17.9</td>
<td>+15.8</td>
<td>+10.0</td>
<td>ND</td>
</tr>
</tbody>
</table>
For all cases except for CNT, the percent error between the experimental and calculated change in outlet milk temperatures was between 2% and 18%. The error for CNT coated surface was the highest, 29.2%. This can be because of lack of replication; only one test was performed for this surface due to the sensitivity of the surface itself. After cleaning the surface, carbon nanotubes’ layout structure changed, which did not allow more tests to be done.

There is no pattern among the test surfaces or test conditions for the error. The calculation of the conductive resistance due to fouling was based on the experimental data as mentioned in Chapter 4. Fluctuations of the inlet and outlet milk temperature during the test were unavoidable since the system is not auto-controlled. Hence, the error in the computations is mainly caused by the experimental data since the methodology for calculating the fouling resistance is semi-empirical.

For higher inlet temperature, 60°C, error was less for the control surface and higher for the coated surfaces. Since the thermal conductivity of the coatings are not known, that of stainless steel was used in the calculation. Even though a small portion of the channel walls was coated, only the top specimen, this might be one of the possible reasons for the error. Other than that, in the calculations physical properties of the milk were assumed to be unchanging with time. However, in reality, it was expected that thermal properties of milk would change due to recirculation of the milk. Moreover, since the milk tank was not totally a closed system, there was a loss of water vapor during the test which effects the change in density and viscosity. All of these factors contribute towards the difference between the computed and experimental values.

5.2.4 Temperature Profile in the Channel

Fouling formation on the contact surface reduces the thermal performance of the heat exchanger leading to more temperature gradient along the width of the channel, and less heat transfer along the flow direction resulting in reduced change in milk temperature through the channel. Temperature profiles in the flow channel at the end of 6 hours are shown for the test surfaces at 3 g/s milk flow rate and 40°C inlet milk temperature. For stainless steel test surface, on which the fouling was the most, temperature gradient along
the width of the channel, i.e., temperature difference at the outlet between the nodes adjacent to the wall and at the center is more, 340 K-320 K = 20 K (Figure 5.12 (a)). Temperature gradients for coated surfaces are, 18 K, 17 K, 14 K, and 13 K, for Microlube/PTFE, TM117P, AMC148, and CNT, respectively (Figure 5.12 (b), (c), (d), and (e)).

![Temperature contour using FLUENT](image)

Figure 5.12 Temperature contour using FLUENT (in K, K= °C+273.15) in the channel at the end of 6 hours for (a) Control, (b) Microlube/PTFE, (c) TM117P, (d) AMC148, and (e) CNT (at 3 g/s, 40°C)

### 5.2.5 Deposit Amount

Deposit amount was calculated as explained in Chapter 4. Biot number, shows the extra resistance due to fouling. It is formulated depending on the overall heat transfer coefficients on a clean and fouled surface, i.e. initial and after 6 hours of operation time. Overall heat transfer coefficient U is defined as the total resistance to heat transfer, which in turn is the total resistance caused by conductive and convective resistances, in the system. Conductive resistances are surface resistances and not dependent on the flow condition and temperature if the physical properties of the solid are not strongly temperature dependent. However, convective resistance, 1/hA
where \( h \) is convective heat transfer coefficient and \( A \) is the effective surface area, is temperature and flow condition dependent. Therefore, in the formulation of \( \text{Bi}_f \) effect of temperature and stream velocity is embedded. However, \( \text{Bi}_f \) by itself is not sufficient to calculate fouling amount for different surfaces having different surface energies. For this reason, a correction factor \( f(\theta) \) needs to be included, where \( \theta \) is the contact angle (Table 5.1).

The derivation and formulation for \( \text{Bi}_f \) and calculation of deposit amount are given Chapter 4. Since the interest is in the percent reduction of fouling when lower surface energy material is used, the value of the function \( f(\theta) \) is considered as 1 for the control surface. \( \text{Bi}_f \) (in the first row) and the correction factor (in the second row) for the test surfaces are given Table 5.13.

For milk flow rate of 10 g/s and inlet milk temperature of 40°C, the linear correlation based on measured deposits amounts for all surfaces vs. contact angle found is:

\[
m = -0.0023 \theta + 0.3928 \quad R^2=0.97
\]

where \( m \) is the deposit amount on the test specimen in grams and \( \theta \) is the contact angle.

As an example, for Microlube/PTFE \( f(\theta) \) becomes:

\[
f(\theta) = \frac{-0.0023 \times 107 + 0.3928}{-0.0023 \times 72 + 0.3928} = 0.65
\]

Table 5.13 Biot number for fouling and the surface dependent correction factor

<table>
<thead>
<tr>
<th></th>
<th>Control (72°)*</th>
<th>Microlube/PTFE (107°)*</th>
<th>TM117P (112°)*</th>
<th>AMC148 (145°)*</th>
<th>CNT (≈180°)*</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 g/s, 40°C</td>
<td>17.8</td>
<td>17.8</td>
<td>15.4</td>
<td>9.1</td>
<td>5.2</td>
</tr>
<tr>
<td></td>
<td>1.0</td>
<td>0.67</td>
<td>0.61</td>
<td>0.30</td>
<td>0.08</td>
</tr>
<tr>
<td>3 g/s, 60°C</td>
<td>11.6</td>
<td>10.8</td>
<td>7.2</td>
<td>6.3</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>1.0</td>
<td>0.53</td>
<td>0.46</td>
<td>0.01</td>
<td></td>
</tr>
<tr>
<td>10g/s, 40°C</td>
<td>18.4</td>
<td>18.2</td>
<td>15.9</td>
<td>8.9</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>1.0</td>
<td>0.65</td>
<td>0.60</td>
<td>0.26</td>
<td></td>
</tr>
<tr>
<td>10g/s, 60°C</td>
<td>8.9</td>
<td>10.2</td>
<td>8.9</td>
<td>2.8</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>1.0</td>
<td>0.62</td>
<td>0.56</td>
<td>0.19</td>
<td></td>
</tr>
</tbody>
</table>

*Contact angle
The correction factor, \( f(\theta) \), decreases with the increase in contact angle. It is consistent within test conditions except for the test condition where flow rate and inlet milk temperature were 3 g/s and 60°C, respectively. At this test condition, as discussed in section 5.1.4 and shown in Table 5.7, standard deviation in the deposit amount on stainless steel was very high compared to other surfaces (0.205 ± 0.057 g). Due to this variation in the deposit amount on the control surface, correction factor for this case was very small especially for AMC148 (0.01 g), which has the least amount of fouling (0.021 g). For this reason correction factor at the mentioned test condition for the coated surfaces is calculated based on the minimum value of deposit amount on the control surface and found as 0.57, 0.50, and 0.13 for Microlube/PTFE, TM117P, and AMC148, respectively. The correction factor is still small at this condition compared to other test conditions, and is not consistent among the test conditions. This is mainly due to the lack of sufficient reliability in the measurement of the deposit amounts. As mentioned in section 5.1.4, and shown in Figure 5.10(c), proteins mainly deposit on the stainless steel-AMC148 border. Considering the quantity of the deposits, flow disturbances around this border, and the sensitivity of the scale the deposit amount measurement for AMC148 cannot be expected to be accurate enough to make good judgement.

In Table 5.14, calculated deposit amounts, calculated by using equation 4.13, for the test surfaces and percent errors are given. The percent errors are higher for the coated surfaces, and the highest for the surface that is least prone to fouling (AMC148 and CNT, -57.1% and -68.2%, respectively) for all test conditions. That means for the surface with the highest contact angle, and thus very little foulant on, the computed and experimental fouling amount differ more.

The average percent difference values between measured and calculated deposit amounts were from 11.1% to 38.1% (except CNT) with overall average of 21.5%. Of the five surfaces tested, the average percent difference was small for Microlube/PTFE and stainless steel. The percent difference was large for TM117P and AMC148 primarily due to small amounts of deposits. This is discussed further in the following paragraphs.
Table 5.14 Calculated and measured (in parenthesis) amount of deposit on the test surfaces in unit area (g/m²) and percent difference

<table>
<thead>
<tr>
<th></th>
<th>Control</th>
<th>Microlube/PTFE</th>
<th>TM117P</th>
<th>AMC148</th>
<th>CNT</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 g/s, 40°C</td>
<td>377.6 (334.8)</td>
<td>235.2 (265.0)</td>
<td>208.0 (255.8)</td>
<td>60.8 (48.0)</td>
<td>11.2 (32.6)</td>
</tr>
<tr>
<td>% difference</td>
<td>+9.2</td>
<td>-14.0</td>
<td>-21.2</td>
<td>+22.6</td>
<td>-68.2</td>
</tr>
<tr>
<td>3 g/s, 60°C</td>
<td>310.4 (317.8)</td>
<td>128.0 (100.8)</td>
<td>147.2 (86.8)</td>
<td>14.4 (32.6)</td>
<td>ND</td>
</tr>
<tr>
<td>% difference</td>
<td>-5.4</td>
<td>+23.1</td>
<td>+46.4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>10 g/s, 40°C</td>
<td>348.8 (341.0)</td>
<td>249.6 (251.1)</td>
<td>203.2 (223.2)</td>
<td>56.0 (83.7)</td>
<td>ND</td>
</tr>
<tr>
<td>% difference</td>
<td>-0.9</td>
<td>-3.7</td>
<td>-11.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>10 g/s, 60°C</td>
<td>241.6 (172.1)</td>
<td>147.2 (137.9)</td>
<td>108.8 (150.4)</td>
<td>8.0 (13.4)</td>
<td>ND</td>
</tr>
<tr>
<td>% difference</td>
<td>+34.2</td>
<td>+3.4</td>
<td>-18.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Absolute average of % difference</td>
<td>12.4</td>
<td>11.1</td>
<td>24.5</td>
<td>38.1</td>
<td>ND</td>
</tr>
</tbody>
</table>

Highest error between the measured and computed deposit amount for coated surfaces is at the condition with a flow rate of 3 g/s and inlet milk temperature of 60°C (+23.1, +46.4, and -57.1%). As mentioned in the discussion of measured deposit amount in section 5.1.4 and aforementioned in this subsection in the discussion of the correction factor calculation, at 3 g/s and 40°C test condition the high standard deviation in the measured deposit amount on the control surface causes the correction factor, calculated based on the experimental data, to be considerably different especially for AMC148 coated surface. This leads to very high error between the computed and measured deposit amount (57.1%).

In addition, the error in the computed deposit amount is considerably high for AMC148 (-57.1% to +22.6), TM117P (-11.8% to +46.4%) and CNT (-68.2%) vs. Microlube/PTFE (-14.9% to +23.1%) and control (-0.9% to +34.2%). The effect of measurement sensitivity on the error was mentioned before. Another and main reason for the error in calculated values is the assumption in the computations. Amount of deposit was linearly correlated with the correction factor that is a function of contact angle. Although the surface wettability is related with contact angle, it may not be linearly
correlated with contact angle. For that reason, the linear fit obtained with limited number of surfaces, i.e., with limited data points, may not be enough to explain the fouling tendency of surfaces with high contact angle.

The percent difference alone is not an indicator for the error between the experimental data and computed results since it is calculated based on the average deposit amount. Figures 5.13 to 5.16 show the computed and measured (with standard deviation) deposit amount for control, Microlube/PTFE, TM117P, and AMC148 test surfaces, respectively.

Figure 5.13 Measured and calculated deposit amount on control surface, SS316

Figure 5.14 Measured and calculated deposit amount on Microlube/PTFE coated surface
Growth of foulants deposit on the test surfaces over time, for a given condition when flow rate is 3 g/s and inlet milk temperature is 40°C, is shown in Figure 5.17. Time for conditioning of the surface, i.e., induction period, is shorter for SS316 (<90 min). Right after the conditioning, foulants deposit on the surface with increase in rate of deposition. The rate of deposition, as can be observed from the slopes of the curves, is smaller for coated surfaces, and almost constant for AMC148 and CNT coated surfaces. This leads to conclusion that, in longer test time, the reduction in fouling amount when coated surfaces are used will be much more than that given in Table 5.8.
In this study, effect of surface coating on mitigation of fouling in the bench-scale heat exchanger was investigated. The coated surfaces were compared with stainless steel SS316 in terms of their tendency to fouling. Since this comparison was based on surface characteristics (contact angle with milk), the percent reduction of fouling amount will be approximately in the same order when the tests are performed in full-scale PHE. Effect of flow rate on foulant deposit amount will be more observable as the operation flow rate in full-scale PHE is higher than in the test conditions (≈3000 gpm versus maximum 600 gpm). At higher flow rate, both the residence time and the removal of foulants from the surface will be affected. At shorter residence times in PHEs, the deposit amount will be less. Deposits, especially when they are attached to the surface loosely as in the case of high contact angle surfaces, will be removed more at higher flow rate. Consequently, the deposit amount per unit area will be less in the PHE. However, as the purpose was to investigate the feasibility of using coated surfaces for mitigating fouling, the surfaces that
gave promising reduction in the deposit amount will still be of value to be further investigated.

The model for the bench-scale heat exchanger can be extended to full-scale PHEs by defining the heat exchanger wall as being corrugated and generating mesh accordingly. In addition, incorporating the boundary condition as heat flux from the heating medium (water) instead of wall temperature profile would be a more accurate next step.

5.3 Summary

The goal of this study was to investigate the appropriateness of proposal of low energy surfaces for mitigating fouling experimentally and computationally. With this motivation, four coated surfaces in addition to stainless steel, a commonly used material in heat exchanger designs, for control purpose were examined for their tendency to fouling. Experimental results showed that less wettable surfaces, which have high contact angle, are less prone to fouling; Microlube/PTFE and TM117P had 20%, and AMC148 and CNT surfaces had 90% less fouling. Computed outlet temperature profile and calculated deposit amount were in good agreement with the experiments. Percent difference between measured and calculated foulant amount was higher for AMC148 and CNT. Fouling of stainless steel surface was much faster than the coated surfaces, i.e., short induction period. Considering both rate and amount of deposits, coated surfaces, especially AMC148 and CNT, slowed down the deposition rate and reduced the fouling amount as shown by experiments and verified by the computational model.
Chapter 6

CONCLUSIONS AND RECOMMENDATIONS

6.1 Conclusions

The main purpose of this study was to recommend materials that can be used to mitigate fouling in milk pasteurization. Towards this end, surfaces having different surface characteristics were examined and a model that describes the fouling formation was proposed. Correlating the deposit formation with contact angle is a fundamental starting point to overcome fouling on the surfaces of heat transfer equipment, a severe problem in food industry and particularly in dairy industry. By this way, a material that is less prone to fouling can be recommended resulting in increase in quality of safer product with less operational cost and cleaning concerns. Towards this end, surfaces having different surface characteristics were examined and a model that describes the fouling formation was proposed. Using this model and surface properties of any material, fouling on the surfaces can be estimated with less experimental work.

For testing fouling formation on different surfaces, a bench scale tester was designed and fabricated. The assumptions in the design of the set-up are listed in Chapter 4. Although the simplifications have some disadvantages, there are benefits they have provided in terms of cost, ease, and reliability in performing the experiments, and in the computational part.

In this study, effects of inlet milk temperature, at two different conditions (40ºC and 60 ºC), and flow rate, 3 g/s and 10 g/s, were investigated. It was found that there was upto 50% less deposit formation when the inlet milk temperature was higher. Results did not provide a definitive trend about the effect of flow rate on deposit amount

When Teflon® based surfaces – Microlube/PTFE and TM117P – were used, fouling was reduced by 20%; whereas, CNT and AMC148 gave the most promising
results for mitigating fouling (90% reduction). However, considering CNT being not applicable in food industry, AMC 148 or any surface with similar properties is recommended for full scale testing.

Thermal performance of the bench scale heat exchanger was reduced the most on the stainless surface; 10°C decrease in outlet milk temperature. The reduction in thermal performance was 40 to 70% less for coated surfaces depending on the contact angle of the surfaces.

The calculations of deposit amount for each test case and the surfaces tested were in good agreement with the experiments (overall average of 21.5%). The average percent difference values between measured and calculated values were from 11.1% to 38.1% (except CNT). Of the five surfaces tested, the average percent difference was small for Microlube/PTFE (11.1%) and stainless steel (12.4%) and large for TM117P (24.5%) and AMC148 (38.1%).

6.2 Recommendations

In industrial plate heat exchangers, there are multiple channels, in each there is flow of milk or heating medium, water. In the experiments, single channel heat exchanger was used for both reducing the cost, and examining the fouling under the worst test conditions keeping in mind that the set up mimics the plate heat exchangers.

Bench scale plate heat exchanger tests have showed that surfaces with low surface energy, high contact angle, are less prone to fouling. For better mock-up of the plate heat exchanger, totally coated and corrugated surfaces can be tested in the future. The benefit that will offer is that the flow regime will not be disturbed as it was at the points test specimens inserted to the whole plate.

Performing the experiments with raw milk in continuous flow will enable better judgment and estimation of deposit amount on the test surfaces. Thereby, change in milk properties will be minimized, and fouling amount can be estimated more precisely.

The test conditions were little different, to examine the worst case, than they actually are in industrial applications. It is recommended to perform the fouling tests at
different and more test conditions, in terms of flow rate and inlet milk temperature. This will bring the advantage along in better correlation of deposit amount with test conditions in the overall model as well as more accurate assessment of fouling under different circumstances.

There were three thermocouples on the test surface. It is expected that if temperature measurement at the wall is done with more thermocouples, better wall temperature profile will be obtained which would result in more accurate computation of temperature profile, as well as outlet milk temperature, thus calculation of deposit amount can be done more precisely.

In order to improve overall model more surfaces having different contact angles, especially in the range 115°-145°, and various test conditions are suggested to be tested so that enables the model to incorporate more data yielding a model applicable to a range of surfaces.

In this study, change in thermal resistance to heat transfer was examined to be used in the computational model that describes fouling formation. The model can be improved by continuously adjusting the mesh as the deposits build up.

In order to be able to recommend the use of coated surfaces in the industry, safety and quality analysis of the product should be investigated with cost analysis. The economic losses due to fouling are mainly; higher operating and maintenance costs due to increased pressure drop, and impaired heat transfer, and hence unnecessary energy consumption. Considering the cleaning needs, cost of chemical and water use and its environmental impact is of concern. When coated surfaces are used due to less fouling on the surface, and ease of cleaning the cost for cleaning would be less without using excess amount of chemicals and water. Effect of chemicals used in CIP on the integrity of the coated surfaces needs to be investigated.

Microbial and chemical analyses of the product, when coated surfaces are used, need to be done to estimate quality of the product. The coating, although very thin, brings extra conduction resistance to the system. Effect of this extra resistance to heat transfer may become important in the food industry due to time-temperature combination in the
pasteurization systems. For this reason, in the pasteurization process, outlet temperature is very important in the thermal inactivation of pathogenic microorganisms and enzymes that may affect the quality. Integrity of the surface needs to be examined with time in addition to the concentration of surface coating material in the product.

Fouling problem is not limited to dairy or food industry. Mitigating fouling in industries such as petroleum, chemical, and waste water treatment by using coated surfaces can be further investigated.
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