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A comprehensive review of milk fouling on heated surfaces

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Abstract

Heat exchanger performance degrades rapidly during operation due to formation of deposits on heat transfer surfaces which ultimately reduces service life of the equipment. Due to scaling product deteriorates which causes lack of proper heating. Chemistry of milk scaling is qualitatively understood and the mathematical models for fouling at low temperatures have been produced but the behavior of systems at ultra high temperature processing has to be studied further to understand in depth. In diversified field the effect of whey protein fouling along with pressure drop in heat exchangers were conducted by many researchers. Adding additives, treatment of heat exchanger surfaces and changing of heat exchanger configurations are notable areas of investigation in milk fouling. The present review highlighted information about previous work on fouling, influencing parameters of fouling and its mitigation approach and ends up with recommendations for retardation of milk fouling and necessary measures to perform the task.

Key words:

Heat exchanger, crystallization, milk fouling, dissolved salts, mitigation.

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NOMENCLATURE

AB	Acid/Base Interaction Forces
Br	Brownian motion
CIP	CLEANING IN PLACE
EL	ELECTROSTATIC DOUBLE LAYER INTERACTION FORCES
HIWPG	HEAT INDUCED WHEY PROTEIN GELS
LW	Lifshitz -Van der Waals interaction forces
NTU	NUMBER OF TRANSFER UNITS METHOD
R_f	FOULING RESISTANCE
SMUF	SIMULATED MILK ULTRA FILTRATE
UHT	ULTRA HIGH-TEMPERATURE PROCESSING
WPC	WHEY PROTEIN CONCENTRATE
α -La	α -LACTALBUMIN
β -Lg	β -LACTOGLOBULIN
γ^-	ELECTRON DONOR

1. Introduction

Deposition of undesired materials on heat transfer surfaces are recognized as fouling. At present milk fluid fouling chemistry is qualitatively understood, and mathematical models for fouling at low temperatures exist but the behavior of systems at UHT temperatures is not clearly understood yet. In the dairy industry thermal processing is an energy intensive event as every product is heated there at least once (deJong, 1997). Heat exchanger fouling problem is a serious matter as it retards heat transfer, enhances pressure drop and finally efficiency of heat exchanger drops down which ultimately affects the economy of process plant (Muller-Steinhagen, 1993; Toyoda, Schreier, & Fryer, 1994). Lack of heating due to fouling deteriorates product quality during pasteurization or sterilization. On the other hand deposits when dislodged due to shear stress of flowing fluid and mix with the stream cause contamination (P.J. Fryer, et al., 1996).

In the dairy industry the cost due to interruption in production can be dominating (C. R. Gillham, Fryer, & Hasting, 2000) compared to the cost due to reduction in performance efficiency (Georgiadis, Rotstein, & Macchietto, 1998a, 1998b). Quality issues are also equally important as the cost and in fact shutdown is required many times concerning product quality instead of performance of the heat exchangers (Georgiadis, et al., 1998b).

Milk constituents are: water, solids, fat, lactose, proteins (casein, β -Lg, α -La), minerals and small quantities of other miscellaneous constituents (Bylund, 1995; Delplace, Leuliet, & Tissier, 1994; Gotham, Fryer, & Pritchard, 1992; Lalande, Tissier, & Corrieu, 1985; Lyster, 1970). Microbial growth prevents uninterrupted operation of heat exchanging equipment (Bylund, 1995; Lalande, et al., 1985; Lyster, 1970). Delplace et al. (Delplace, et al., 1994)

conducted experiments and observed that only 3.6% of denatured β -Lg contributes to deposit formation whereas, Lalande et al. (Lalande, et al., 1985) observed around 5%.

Fouling is controlled by the aggregation reaction of protein molecules and the formation of protein aggregates which reduces fouling (deJong, 1997; Delplace, Leuliet, & Levieux, 1997). Denaturation of protein in heat exchangers starts above 70-74°C and the first deposit layer is largely (usually < 5µm) mineral (Fryer & Belmar-Beiny, 1991). Different authors have informed that the major components of milk fouling are calcium phosphate and whey protein (deJong, et al., 1998; Elofsson, Paulsson, Sellers, & Arnebrant, 1996; J. Visser & Jeurink, 1997). Some researchers have identified experimentally that the fouling is caused by denatured and aggregated whey proteins together and primarily influenced by the presence of the denatured (but not the aggregated) proteins in the bulk (X. D. Chen, Chen, & Wilson, 2001). Many research interests highlighted the study of mechanism, deposition, modeling and comparison of deposition on different surfaces to accumulate information for future reference (X. D. Chen, et al., 2001; deJong, Bouman, & vanderLinden, 1992; Toyoda, et al., 1994). Thus the present paper attempts to accumulate information on milk composition, fouling, mechanism along with modeling approach.

2. Factors Affecting Milk Fouling

Milk fouling in heat exchangers are affected by several factors, which can be broadly be classified into five major categories (**Table 1**) (B. Bansal & Chen, 2005; Bipan Bansal & Chen, 2006):

Table 1**2.1 Milk composition**

Milk composition is dependent on its source and hence may not be possible to change (**Table 2**). Variations in milk fouling are also dependent on differences in its composition (Bipan Bansal & Chen, 2006; Belmar-Beiny, Gotham, Paterson, Fryer, & Pritchard, 1993; Burton, 1967; deJong, 1997). Fouling enhances with increase of the protein concentration (Changani, Belmar-Beiny, & Fryer, 1997; Toyoda, et al., 1994). Some researchers reported that the heat stability of milk proteins decreases with the reduction in pH (Corredig & Dalgleish, 1996; deJong, et al., 1998; Foster, Britten, & Green, 1989; Xiong, 1992). Gotham et al. (Gotham, 1990) noted that the effect of pH on fouling was same as the effect of pH on aggregation and thus suggested that aggregation is the governing reaction. The chemistry of the protein reaction in fouling is further discussed by Visser and Jeurink (Changani, et al., 1997; Christian, Changani, & Fryer, 2002; J. Visser & Jeurink, 1997). Roefs and deKruif (Roefs & deKruif, 1994) said increase or decrease of the calcium content of milk lowers the heat stability and causes more fouling in comparison to normal milk. Milk fat has little effect on fouling (J. Visser & Jeurink, 1997).

Additives (mineral, vitamins, enzymes and syntheses additive) may retard fouling by enhancing heat stability of milk but addition of any ingredient in milk may not be permitted in many countries (Changani, et al., 1997; Lyster, 1970; P. J. Skudder, Thomas, Pavey, & Perkin, 1981). 24 hours holding of milk before processing results in less fouling on the other hand further ageing increases fouling (Burton, 1968; Changani, et al., 1997). Burton (Burton, 1968)

and deJong (deJong, 1997) reported that prolonged storage of milk for a few days at 5°C may enhance fouling due to the action of photolytic enzymes.

Table 2

Reduction of fouling in the heat exchangers can be obtained by preheating of milk in holding tubes which causes denaturation and aggregation of proteins before transferring to the heating section (R. W. Bell & Sanders, 1944; Burton, 1968; Foster, et al., 1989; Mottar & Moermans, 1988). Reconstituted milk generates much less fouling as around 25% of fouling generated when β -Lg is denatured during the production of milk powders (Changani, et al., 1997; J. Visser & Jeurink, 1997). Changani et al. (Changani, et al., 1997) reported that the reconstituted milk contains 9% less calcium, which would have resulted less fouling. In contrast, Newstead et al. observed increase of UHT fouling rates of the recombined milk with increase of preheat treatment (preheating temperature X preheating time) (Newstead, Groube, Smith, & Eiger, 1998). Further, they (P.J. Fryer, et al., 1996; Hong Xin, 2003) analyzed the fouling deposits and found high levels of fat (up to 60% or more) compared to that obtained in the deposits formed during fresh milk processing (10% or less) and the difference could be ascribed to changes in fat-globule membranes. Fung et al. (Fung, McCarthy, & Tuoc, 1998) studied the effect of damage to milk fat globule membrane by a pump running in cavitations status (Fung, et al., 1998; Newstead, et al., 1998).

Figure 1 provides some insight into the composition of whey derived from bovine milk, through normal cheese-making processes (H. W. Modler, 2000). The composition of whey is

considerably more variable than milk and is highly dependent on the type of cheese being manufactured, seasonality, culture and rennet selection, manufacturing procedures, and type of equipment used. These differences in composition and functionality are a challenge to the manufacturers. Regester and Smithers (Regester & Smithers, 1991) observed a reduction in the α -La content of milk during the final 3 mo of lactation, concomitant with a rise in β -Lg (W. Modler, 2009; Regester & Smithers, 1991). Proteins react in both the bulk and the thermal boundary layer in the milk. Native protein N is transformed to denaturated protein D, in a first-order reaction. The denaturated protein then reacts to give aggregated protein A in a second order reaction.

Figure 1

A schematic representation of the proposed fouling model is given in **Figure 1**. When the milk is heated to a temperature greater than 65°C , the betalactoglobulin protein is unsteady and becomes the precursor for deposit formation according to two possible mechanisms (Youcef, Abdelkader, & Lounes, 2009):

- 1- The β -Lg natural protein (N-protein) experiences a denaturation process (change of structure) and becomes very reactive due to its SH bond (β -Lg denatured form or D-protein).
- 2- An irreversible polymerisation reaction provides the insoluble particles as aggregations which are noted as protein A (β -Lg aggregated form).

These changes varied with an increase in the casein content of whey protein concentrate prepared during the first 4 mo of lactation. Whey can be divided into 3 distinct nitrogen-bearing

fractions; namely, heat-coagulable (0.44%), non-heatcoagulable proteose-peptone component (0.10%), and nonprotein nitrogen (0.18%). The proteose-peptone content increases by about 0.09% during cheese making due to release of the macro peptide component from k-casein of milk to bring the total to 0.19% (**Figure 1**) (W. Modler, 2009).

Caseins of protein precipitates upon acidification and generates resistant to thermal processing (J. Visser & Jeurink, 1997). The native proteins (β -Lg) first denature (unfold) and expose the core containing reactive sulphhydryl groups with the heating of milk. In denatured state protein molecules react (W. Modler, 2009) with the similar or other types of protein molecules like casein, α -La forming aggregates (Changani, et al., 1997; J. Chen, 2000; Dalgleish, 1990; Treybal, 1981).

The extreme variability of the composition of fouling layers published is highlighted by the selection in **Table 3**.

Table 3

2.2 Effect of whey protein concentration

Deposit layers are often formed on the heat exchanger surfaces exposed to process fluid during thermal processing of the dairy products (Toyoda, et al., 1994). A considerable attention has been given to the fouling and removal of proteinaceous deposits due to their importance in the dairy industries (X.D. Chen, Li, Lin, & Özkan, 2004; Fickak, Al-Raisi, & Chen, 2011; Fryer, Hasting, & Jeurink, 1996; H. Visser, 1997; Wilson, Fryer, & Hasting, 1999; Wilson, Fryer, &

Hasting, 2002). Heat sensitive whey proteins are the major components in dairy fouling deposits (H. Visser, 1997). The fouling deposit structure in the dairy plant is dominated by the aggregated whey protein molecules. Many researchers (Belmar-Beiny, et al., 1993; J. Chen, 2000; Davies, Henstridge, Gillham, & Wilson, 1997; Delplace & Leutiet, 1995; P.J. Fryer, et al., 1996; C.R. Gillham, Fryer, Hasting, & Wilson, 1999; P. Schreier, et al., 1994; H. Xin, Chen, & Ozkan, 2002a, 2002b) have used heat induced whey protein gels (HIWPG) as model system for milk fouling and cleaning operation due to the complex nature of milk deposits and cleaning (Fickak, et al., 2011).

Heat induced whey protein gel (HIWPG) which produced deposits containing protein 60% the weight of the deposit. Due to heating the HIWPG deposits are formed from the aggregate of whey proteins in the milk (Langton & Hermansson, 1992; D.M. Mulvihill, Rector, & Kinsella, 1990; Verheul & Roef, 1998a). During formation of HIWPG only a fraction of the whey proteins are aggregated (Burton, 1968; Fickak, et al., 2011; Lyster, 1965; Verheul & Roef, 1998a, 1998b). HIWPG with high protein concentration tend to form faster due to the increasing rate of aggregation and the decreasing coagulation time (Sharma & Hill, 1983). Mleko (Mleko, 1999). It also has been reported that increasing the protein concentration increases the firmness and the aggregate size of whey protein concentrate (WPC) gels which accelerates the gelation process (Mleko, 1999). Furthermore, Puyol et al. (Puyol, Perez, & Horne, 2001) has found that WPI (Whey Protein Isolate, Lacprodan DI-9224) gels with high protein concentration tend to form at lower temperature. Some researchers concluded that high protein concentration can greatly influence the formation mechanisms of whey protein gels; however, the effect of protein concentration on the fouling and cleaning of dairy heat exchange surface has not been

demonstrated quantitatively by them (Bird & Fryer, 1991; Fickak, et al., 2011; C. R. Gillham, et al., 2000; C.R. Gillham, et al., 1999; Mercadé-Prieto & Chen, 2006; Puyol, et al., 2001; H. Xin, et al., 2002b).

Fickak et al. (Fickak, et al., 2011) have studied the effect of whey protein concentration on the fouling and cleaning behaviors of a pilot-scale heat exchanger. They have assessed the influence of the properties of surfaces based on stainless steel on the fouling behavior of different milk components (calcium phosphate and whey proteins), complex milk systems (FMF) and milk related bacteria. In all cases the surface material practically influenced the fouling behavior, although in different ways they influenced the deposition and the cleaning phases (Roxane Rosmaninho, et al., 2007).

2.3 Operating conditions in heat exchangers

The following significant operating parameters could be varied in a heat exchanger: air content, velocity or turbulence, and temperature (Arnebrant, Barton, & Nylander, 1987; Belmar-Beiny & Fryer, 1993; Belmar-Beiny, et al., 1993; Gotham, et al., 1992; Hege & Kessler, 1986; Kessler & Beyer, 1991; Lalande & Reno, 1988; Lalande, et al., 1985; P. J. R. Schreier & Fryer, 1995). Presence of air in milk enhances fouling (Burton, 1968; deJong, 1997; deJong, et al., 1998). It is reported that, fouling is enhanced only when air bubbles are formed on heat transfer surfaces, which then become a nuclei for deposit formation (Burton, 1968; deJong, 1997). Heating of milk decreases its dissolved air content resulting to reduction of the pressure (deJong, 1997; deJong, et al., 1998).

Enhanced turbulence in flow retards fouling deposition (Belmar-Beiny & Fryer, 1993; Belmar-Beiny, et al., 1993; Santos, Nylander, Paulsson, & Trägårdh, 2001; Santos, et al., 2003). Paterson and Fryer (Paterson & Fryer, 1988) and Changani et al. (Changani, et al., 1997) reported that the thickness and subsequently the volume of laminar sublayer decreases with increasing velocity resulting to retardation of foulant deposition on heat transfer surfaces. Other researchers (Changani, et al., 1997; Paterson & Fryer, 1988; Rakes, Swartzel, & Jones, 1986) had informed that the higher flow velocities also promote deposit re-entrainment due to enhanced fluid shear stresses.

Belmar-Beiny et al. (Belmar-Beiny, et al., 1993) concluded that the higher turbulence and different flow characteristics are in fact generate shorter induction period in plate heat exchangers compared to tubular heat exchangers. They reiterated that, the presence of low velocity zones near the contact points between the adjacent plates is responsible for that. Further, they obtained reduced fouling with due to boundary layer agitation pulsating flow. Other researchers (Bradley & Fryer, 1992) noticed that when only the wall region near the heat transfer surface is hot enough to cause the protein denaturation and aggregation reactions then less fouling could be seen due to less exposure time of fluid near the wall for higher mixing rate (Changani, et al., 1997; X. D. Chen, et al., 2001; X. D. Chen, Chen, Nguang, & Anema, 1998; deJong, et al., 1992; Delplace, et al., 1994; Toyoda, et al., 1994; VanAsselt, M.Vissers, Smit, & Jong, 2005).

In a heat exchanger, temperature of milk is probably the single most important factor which is controlling fouling (Belmar-Beiny, et al., 1993; Burton, 1968; Corredig & Dalgleish, 1996; Elofsson, et al., 1996; T. Jeurnink, Verheul, Stuart, & deKruif, 1996; Kessler & Beyer,

1991; Santos, et al., 2003; Toyoda, et al., 1994). Higher fouling is resulted with increase of temperature. The nature of fouling changes from type A to type B (**Table 4**) at temperature exceeding 110°C (Burton, 1968). Milk fouling can be classified into two categories (Burton, 1968; Changani, et al., 1997; Lund & Bixby, 1975; J. Visser & Jeurink, 1997):

1. Type A: protein fouling takes place at temperatures between 75°C and 110°C . The deposits appear white, soft, and spongy (milk film) and their constituents are 50-70% proteins (mainly β -Lg), 30-40% minerals, and 4-8% fat,
2. Type B: mineral fouling takes place at temperatures above 110°C . The deposits are grey in color (milk stone) and hard, compact, granular in structure and their composition is 70-80% minerals (mainly calcium phosphate), 15-20% proteins, and 4-8% fat (B. Bansal & Chen, 2005).

Table 4

Bulk temperature and temperature difference are both important for fouling. Chen and Bala (X. D. Chen & Bala, 1998) studied the effect of surface and bulk temperatures on the fouling of whole milk, skim milk, and whey protein and observed that in initiating fouling, the surface temperature was the most important factor. They had noticed no fouling at the surface temperature less than 68°C , even though the bulk temperature was up to 84°C . Chen et al. (X. D. Chen, et al., 2001) also predicted that mixing caused by in-line mixers can reduce fouling substantially, but no information being provided about precipitation in bulk (X. D. Chen & Bala, 1998; X. D. Chen, et al., 2001).

2.4 Type and characteristics of heat exchangers

In dairy and other food processing industries the plate heat exchangers are extensively used. Delplace et al. (Delplace, et al., 1994) informed that the plate heat exchangers are prone to fouling due to their narrow flow channels. Complete mitigation of milk fouling in a heat exchanger is difficult as the temperature of the heat transfer surface needs to be considerably higher than the bulk temperature for efficient heat transfer. Complex hydraulic characteristics of plate heat exchangers at elevated temperatures made a difficult situation for milk fouling analysis (deJong, 1997; deJong, et al., 1998; Delplace, et al., 1997; Kindle, Busse, Kampa, Meyer-Konig, & Daschner, 1996; Metaxas & Meredith, 1988; Sieber, Eberhard, & Gallmann, 1996; Thompson & Thompson, 1990; Villamiel, Corzo, Martinez-Castro, & Olano, 1996; Zaida, Sarma, Grover, & Heldman, 1987).

Direct resistance heating (Ohmic heating) is a novel heat treatment process where electrical current is passed through the milk for generating heat which pasteurizes or sterilizes milk (deAlwis & Fryer, 1990; Quarini, 1995). In recent years this technology has been in use after being abandoned for a major part of the 20th century. APV International Ltd. (England) developed commercial ohmic heating units for continuous sterilization of food products (Ayadi, Bouvier, et al., 2003; Ayadi, Chopard, Berthou, & Leuliet, 2003; Ayadi, Leuliet, Chopard, Berthou, & Lebouche, 2004; Fryer, deAlwis, Koury, Stapley, & Zhang, 1993; P. Skudder & Biss, 1987).

2.5 Presence of Micro-organisms

The adhesion of micro-organisms to the surface is enhanced by formation of fouling layer resulting to bio-fouling. The deposits become nutrients to microorganisms which ensure their growth (Ayadi, Bouvier, et al., 2003; Ayadi, Chopard, et al., 2003; Ayadi, et al., 2004). Most of the processes in industry are carried out at temperatures below 100°C . For instance, pasteurization is commonly achieved by heating milk at 72°C for 15 seconds in a continuous flow system. Just the pathogenic bacteria along with some vegetative cells are killed at this temperature. A higher temperature of 85°C is required to kill the remaining vegetative cells. Spores resistant to large amount of heat and remain active well beyond this temperature (Bott, 1993).

Bio-fouling in a heat exchanger, either by micro-organisms deposition or biofilm formation raises serious quality concerns. The effect of bio-fouling in dairy plant has been investigated by Flint and co-workers (Bott, 1993; S. Flint, et al., 1996; S. H. Flint, Brook, vandenElzen, & Bremer, 1997; S. H. Flint, Brooks, & Bremer, 2000; S. H. Flint, vandenElzen, Brooks, & Bremer, 1999) and they informed that, bio-fouling occurs as per two different mechanisms: accumulation of micro-organisms on the heat transfer surfaces and attachment of micro-organisms on the deposit layer formed on the heat transfer surfaces.

In the process stream the deposit layer of micro-organisms are not only affect the product quality but also influence the fouling process (Yoo, et al., 2004). Hydrodynamic forces drive the bacteria and release them to the process fluid due to which concentration of bacteria could be increased at the downstream. This may cause the bacterial growth in areas which are not

conducive to bio-fouling. The released pattern of thermophilic bacteria, *Bacillus* spore and other *Stearothermophilus* into the process stream were studied in detail by some researchers (X. D. Chen, Yoo, Benjamin, & Turner, 1998; Yoo & Chen, 2002) and they found the specific bacterial growth inside the fouling layer and later discharge into the flow stream which causes contaminations.

2.6 Location of surface area in equipment

The relative amounts of the denatured and aggregated proteins in processing equipment depend on a number of factors like operating conditions, type and design of heat exchanger, properties of the heat transfer surface etc. The processed milk at the exit of the heat exchanger would have large amounts of the denatured and aggregated proteins which enhance fouling. Thus Petermeier et al. (Petermeier, et al., 2002) noticed controlling of fouling within the heat exchanger may not yield effective results and an overall strategy may need to be developed to study the fouling process over the entire setup.

3. Fouling Deposit of Dairy Materials

Fouling deposit refers to any layer or patches of extraneous material on a heat exchanger surface (Peny & Green, 1985). The composition and structure of fouling vary greatly with the processing conditions, which significantly affect their removal processes as well (Hong Xin, 2003).

3.1 Classification of Milk Fouling Deposits

The formation of fouling, extent and nature of deposits in dairy fluid is influenced by many factors, such as the processing temperature, seasonal changes, age, pH, air content, and pre-treatment (Burton, 1968; Changani, et al., 1997; Lalande & René, 1988; J. Visser & Jeurnink, 1997; Yoon & Lund, 1989).

Milk deposits are classified in many ways depending on the chosen criteria. From the cleaning point of view, it is useful to classify typical soils according to their nature and structure. Based on processing temperature range, Lyster (Lyster, 1965) and Burton (Burton, 1968) classified milk deposits as type A and type B. The typical compositions of type A and B fouling are summarized in **Table 4**. The types and compositions of the fouling are more complex than the ones covered by **Table 4**. However, the two basic types are still useful indicators for characterizing milk fouling deposits (Burton, 1968; Lyster, 1965).

There are significant differences noticed between the deposits and the raw milk. Detailed information can be obtained from the compositional analysis of milk deposits (T. Jeurnink & Brinkman., 1994; Lalande, et al., 1985; Tissier, Lalande, & Corrieu, 1984), and are incorporated in the present study. As fouling results from preferential deposition of certain components, type A are proteinaceous deposit, containing mainly β -Lg proteins. The main protein components in type B are caseins. Lyster (Lyster, 1965) reported that although calcium and phosphate composed only 30% weight of mineral content in raw milk, they typically formed 90% weight of the mineral content of milk fouling deposit. The ratio of calcium to phosphate is found to be 1.5-1.6, indicating the presence of the hydroxy apatite $[\text{Ca}_5\text{OH}(\text{PO}_4)_3]$, the least soluble calcium phosphate complex (Lalande, et al., 1985; Lyster, 1965). The proteinaceous deposits (Type A)

play the dominant role in most food processing plants, and they are normally more difficult to remove compared to the mineral deposits (VanAsselt, et al., 2005). The low density of proteinaceous deposit can induce a high pressure drop and a high thermal resistance across processing equipment (Fickak, et al., 2011; Lyster, 1965).

3.2 Low Temperature Milk Deposits

In dairy processing plants the soil also forms on cool surface. Formation of the low temperature soil is quite different from that of the heat induced fouling deposit. Kane and Middlemiss (Kane & Middlemiss, 1985) informed that a soil with a much more open structure and much larger fat content is formed at low temperature. The difference between the deposits formed on cool (particulate) and heated surfaces (crystallization) are expected to influence the cleaning process, and thus it requires a different design of CIP system and cleaning program.

4. Basic principles of Fouling

The bulk temperatures of hot and cold liquids under turbulent flow extend the boundary layer and generates a good mixing (K. J. Bell & Mueller, 2001).

Heat transfer equipment are often limited by fouling (Bott, 1995). The effect of fouling on the heat transfer surface is accounted in design by overall fouling resistance R_f , into the basic heat transfer Equation (4):

$$\frac{1}{U} = \frac{1}{U_0} + R_f \quad (1)$$

where U_o is clean and U is fouled overall heat transfer coefficients.

Greater surface roughness produces greater turbulence (Bott, 1995). Heat transfer coefficient has been changed while the thermal resistance of foulant, roughness of foulant, and Reynolds (Re) number are changed due to change of foulant.

Figure 2

In **Figure 2**, curve A is showing a linear relation between deposit thickness and time, which represents the enhancement rate of thickness of fouling, with time. In curve B, fouling initiation has occurred similar to curve A and then after progressing linearly up to a significant interval it takes the curved shape. Curve C representing **Figure 2** and it is a general model of fouling progress which could be fitted by equations for the curves. The curves A, B, and C shown in **Figure 2** have initiation period (initiation) and these periods are short and it is difficult to model, so the most mathematical models have ignored it (Bott, 1995).

4.1 Mechanism of Milk Fouling

Calcium phosphate and whey protein are major components in milk fouling deposit. Due to their heat sensitivity, both of the components form insoluble aggregates in bulk solution. The final deposit contains calcium phosphate concentrated at the deposit-metal layer interface. In the milk fouling, more researchers have accepted that in the initial phase of fouling, individual whey

protein molecules are absorbed onto the stainless steel heating surface first, and then the mineral or whey protein aggregates are formed in the bulk solution or sub-layer which is finally attached to that monolayer. There is induction time in fouling phenomena at the initial stage after which the fouling commence and become noticable (Belmar-Beiny & Fryer, 1993; J. Visser & Jeurink, 1997).

Visser and Jeurink (J. Visser & Jeurink, 1997) have pointed out that the whey protein fouling deposit proceeds in the same way as thermally-induced whey protein gelation. When the material, mainly whey protein aggregates accumulates and reaches to the gelation concentration, a surface gel layer (protein deposit) could be formed on the heating surface (J. Visser & Jeurink, 1997).

Visser and Jeurink (J. Visser & Jeurink, 1997) reported about proteins formation in fouling and found the major components in a fouling are calcium phosphate and whey protein. An investigation of fouling for a long period usually shows a higher proportion of minerals in the deposits near the surface caused by the diffusion of minerals through the earlier formed deposits rather than minerals forming on the surface at the beginning (Belmar-Beiny, et al., 1993). Deposits from contact heating time less than 4s were analyzed by Belmar-Beiny and Fryer (Belmar-Beiny & Fryer, 1993) and found that the first layer was made of proteinaceous material. Bulk and surface processes governs fouling in a heat exchanger. A number of stages guide the deposition such as (Belmar-Beiny & Fryer, 1993):

- 1- Aggregation and denaturation of proteins into the bulk fluid
- 2- Migration of the aggregated proteins to the surface

- 3- Surface reactions resulting causing incorporation of protein into the deposit layer and possible re-entrainment or removal of deposits.

Physical/chemical changes in the proteins or the mass transfer of the proteins between the fluid and the heat transfer surface are related to the steps controlling the overall fouling. In some cases, it may be a combination of both. Belmar-Beiny et al. (Belmar-Beiny, et al., 1993) and Schreier and Fryer (P. J. R. Schreier & Fryer, 1995) stated that fouling was dependent on the bulk and surface reactions and not on the mass transfer. On the other hand Schreier and Fryer came with a different observation and (P. J. R. Schreier & Fryer, 1995) reiterated that the fouling rate was independent of the concentration of foulant in the liquid.

Jeurnink et al. (T. Jeurnink, et al., 1996) found that only 0.14% of the denatured β -lg finally ends up in the deposits layer. Build up of minerals and protein aggregates and calcium phosphate in the fouling deposit layer may not proceed independently. Delsing and Hiddink (Delsing & Hiddink, 1983) experimentally obtained that the presence of the calcium ions is essential for the growth of protein deposit layers. Formulation of the fouling problem occurring inside a plate heat exchanger during pasteurization of milk is proposed as follows.

4.1.1 System description

Plate heat exchanger is recommended for milk pasteurization as it provides an important convection coefficient and higher turbulence in comparison to other classes of heat exchangers. The process consists of heating milk at prefixed temperature for a certain time to eliminate the pathological action of any bacteria. This is a necessary treatment that milk must undergo before any further use. In pasteurization process milk from a pre-heating tank, enters into the heat

exchanger where it is heated by hot steam in order to reach the pasteurization temperature (Youcef, et al., 2009).

4.1.2 The mechanism of fouling

A schematic representation of the proposed milk fouling model is given in **Figure 3**. When milk is heated at a temperature greater than 65°C , the β -Lg protein become unsteady and it forms the precursor for deposit formation following two possible mechanisms (Georgiadis & Macchiatto, 2000; Lalande, Tissier, & Corrieu, 1984)

- 1- The β -Lg natural protein (N-protein) experiences a denaturation process (change of structure) and becomes very reactive at higher temperature in presence of SH bond (β -Lg denatured form or D-protein).
- 2- An irreversible polymerisation reaction resulting in insoluble particles as aggregations noted protein A (β -Lg aggregated form).

Induction period is usually extremely short or even instantaneous in plate heat exchangers where high turbulence causes intense mixing of fluid. With increase of turbulence the fouling decreases (Changani, et al., 1997; Youcef, et al., 2009).

Figure 3

4.1.3 Adsorption Mechanism

The adsorption equilibrium analysis is the most important fundamental information used to determine the capacity of adsorbent. Both the adsorption capacity and the kinetic behavior of the adsorbent are of great importance for the lab-scale and large scale applications. The kinetics analysis is a useful tool to get the time required to reach the equilibrium regarding the completion of adsorption. The kinetic process of adsorption is explained by several mathematical models where in case more than one mechanism may be responsible as a rate determining step (Polat, 2009). The optimization and scale up of adsorbent beds requires the equilibrium and mass transfer of adsorbent-adsorbate system fully understood in the formulation of kinetics of sorption in solid-liquid systems. The interaction between the adsorbent-adsorbate is responsible for the nature of equilibrium between them and the interaction is affected by the resistances of mass transfer in the establishment of equilibrium (Polat, 2009).

The interaction between proteins and adsorbents do not occur instantaneously. Heat and mass transfer is controlled by the adsorption rate. The controlling mechanisms of adsorption rate are explained by mathematical models. These models are describing the adsorption data which have been explained by adsorption reaction models and adsorption diffusion models (Polat, 2009).

4.1.4 Causes of protein aggregation

The β -Lg protein has a global structure held together by S-S bonds and one non-exposed internal free SH group. The β -Lg starts to unfold with the rise of temperature. The free thiol group is therefore exposed and the molecule enters into an activated state and making it possible

to react with another β -Lg molecule. Thus, a radical chain may grow to form an aggregate which is able to deposit on the heat transfer surface. The rate of deposition was found related to the concentration of activated molecules in the solution and may be calculated by using the model of denaturation and aggregation of the β -Lg (Delplace, et al., 1994; Lalande & Reno, 1988).

Research has shown that the two major whey proteins, α -La and β -Lg, become unstable at temperatures above 65°C . When heated above this temperature, protein denaturation occurs and forms protein aggregation and precipitation. The response to thermal treatment varies with types of proteins. They precipitate in different proportions and make the separation possible. Therefore, heat-induced aggregation and precipitation is an important treatment in manufacture process of many dairy products, and is used to modify functional properties with the goal of ensuring safety of the food product. The rate at which whey proteins aggregate is controlled by process conditions are protein concentration, pH and temperature and the presence of other components. Different solutions of β -Lg and α -La were considered in the investigation. These proteins are the predominant protein comprises of about 70% of all the proteins in whey and are key to the functional properties and characteristics of whey. Of the whey proteins, α -La has the lowest denaturation temperature, 62°C , followed by Bovine serum albumin (BSA) at 64°C , immunoglobulins (Ig) at 72°C , and β -Lg at 78°C . β -Lg denatures more quickly than α -La at 85°C and at other temperatures in heated milk and whey systems. The overall effect of heating is greater on β -Lg than on α -La. Because α -La can renature upon cooling, it is considered to be the most thermostable whey protein (Polat, 2009).

The kinetics of deposition, the different physical and chemical parameters, quantitative analysis of deposition and fouling resistance are yet to be fully understood (Lalande & Reno, 1988; J. Visser & Jeurnink, 1997; Youcef, et al., 2009).

4.1.5 Mineral deposition

In addition to the protein fouling, deposition takes place also from the calcium phosphate which represents an inverse solubility relation with temperature. During the pre-heating process, the ionic product becomes high following the inverse solubility concentration limits. Jeurnink and Brinkman (T. Jeurnink & Brinkman., 1994) stated that salts deposit in the form of crystals on the surface of the heat exchanger. Calcium phosphate may also precipitate in the core flow. Ultimately in all cases it forms a deposit on the stainless steel wall of the heat exchanger plates as shown in **Figure 1** (Youcef, et al., 2009).

In milk fouling constituent of deposition proceed though a complex process in which both whey protein aggregation and calcium phosphate formation in the bulk fluid are to be accounted.

Details of fouling mechanism could be described in the following steps (Youcef, et al., 2009):

- 1- Straight adsorption of a protein mono-layer occurs on the heat exchanger surface even at room temperature.
- 2- Formation of activated β -Lg molecules in the bulk solution at temperatures higher than 65°C . The β -Lg aggregates (tenths of nanometers) and calcium phosphate particles are formed.

- 3- These foulant particles formed in the bulk are continuously transported to the heated surface. However, some activated molecules could be deactivated during this phase due to some reactions with other molecules in the bulk which transforms the particles to insufficient active [state of creating](#) fouling.
- 4- Deposition of activated molecules by adsorption on the heat exchanger surface. Calcium ions entrapped in the protein deposit may help to stabilize these structures.
- 5- At relatively high temperatures (above 85°C), the main deposit component is calcium phosphate which offers an open network structure where small protein aggregates can be entrapped.

5. Effect of heated material and its surface on milk fouling

It has been found that the deposit growth appears to be dependent mainly on the interactions between the fluid and the surface, and the nature of the surface becomes unimportant once the first layer is formed (Dupeyrat, Labbe, Michel, Billoudet, & Daufin, 1987; Yoon & Lund, 1989). Different coatings on the surface had no effect on the amount of deposit formed, but they had an effect on the strength of adhesion (Britten, Green, Boulet, & Paquin, 1988). The magnitude of the soil-substrate adhesion force, hence the cleaning processes may be altered by changing the nature and conditions of the surface by surface modification methods, such as coating, electrochemical polishing, chemical treatment, magnetic field etc. (Britten, et al., 1988; Kittaka, 1974; Koopal, 1985; Nassauer, 1985; Premathilaka, Hyland, Chen, & Bansal, 2006; Hong Xin, 2003). Ferrous and non ferrous surfaces are describing below:

5.1 Stainless Steel Based Surfaces for Milk Heating Processes

The researcher of dairy industries are facing still a challenge to understand the mechanism and to minimize fouling caused by milk on processing equipment during its heat treatment. The most common types among the materials used in process equipment in the food industry are different grade of stainless steel. Several techniques have been applied to modify its surface properties with the aim of reducing the buildup of unwanted deposits (fouling). The characteristics of some of those techniques were published (Santos, et al., 2004), where the description of their role on fouling caused by several milk components and dairy products (R. Rosmaninho, Rizzo, Muller-Steinhagen, & Melo, 2003, 2005; R. Rosmaninho, Visser, & Melo, 2001) are incorporated. Surface energy is one among several surface parameters affecting and **controlling** the fouling process. Reactive sputtering technique was used to obtain a number of stainless steel materials with similar surface composition and morphology having variable surface energy values for conducting performance investigation of those materials. The objective of some studied were to estimate the calcium phosphate component in the main mineral constituent of deposits from milk (T. Jeurnink, et al., 1996; J. Visser, 1999), the role of the surface energy on fouling build-up and cleaning along with finding out a better way of characterization of fouling caused by milk (R. Rosmaninho, et al., 2005).

The mechanism of deposition can be separated into several steps (Anema & McKenna, 1996; Arnebrant, et al., 1987; Changani, et al., 1997; Gotham, et al., 1992; Hege & Kessler, 1986; Karlsson, Wahlgren, & Trägårdh, 1996; Kessler & Beyer, 1991; Lalande & René, 1988; Lalande, et al., 1985; D. M. Mulvihill & Donovan, 1987; Roefs & deKruif, 1994; Ruegg, Moor, & Blanc, 1977; Toyoda, et al., 1994). In the initial stage, calcium phosphate particles form in the

bulk due to heating. On arrival at the vicinity of the heated surface, these particles could be attached to it depending on the forces established between the foulants and the surface. Deposition on the surface depends on the interaction forces between the particles and the surface, surface properties of the particles and of the metal support (R. Rosmaninho, et al., 2005).

The forces that play a role in the attachment of the calcium phosphate on a heated metal surface are the Lifshitz -Van der Waals interaction forces (LW), the electrostatic double layer interaction forces (EL), the Lewis acid/base interaction forces (AB) and the Brownian motion (Br). Van Oss divided the surface energy of a surface into four components, among them the electron donor (γ^-) component being the most often used for characterizing component of solid surfaces (R. Rosmaninho, et al., 2001; C.J VanOss, 1994; C. J. VanOss, Giese, & Wu, 1997; Wu & Nancollas, 1998). After formation of the initial layer on the solid surface, other particles move from the bulk liquid and adhere on top of the initial layer and develop a more or less structured and compacted deposit. Structure of the deposit depends both on the first layer of construction, which depends mainly on the surface properties of the particles and ions present in the solution which contributes to the growth of the deposition (precipitation/crystallization kinetics). The interactions of these factors also determine the resistance to removal (R. Rosmaninho, et al., 2005).

5.2 Calcium phosphate deposition on different stainless steel-based surfaces

Researchers are living with one of the main challenges in the dairy industry, to understand and minimize fouling of processing equipment caused by milk during heat treatment. Special interest was posed by the researchers to calcium phosphate, the main mineral component

of milk deposits (T. J. M. Jeurink, Walstra, & deKruif, 1996; J. Visser, 1999), and also to the role of the deposition surface on the amount and type of fouling accumulation (R. Rosmaninho & Melo, 2006). Stainless steel is commonly used in processing equipment of food industries. Several techniques have been developed recently to modify the surface of stainless steels with the goal of reducing the buildup of unwanted deposits (fouling) in pasteurizers and sterilizers of dairy industry (Santos, et al., 2004).

Calcium phosphate deposition is a crystallization/precipitation process (Amjad, 1998; Bott, 1994; Krause, 1993; Mullin, 1993), starting with the formation of stable clusters of ions or molecules in the liquid (called nuclei), known as nucleation which occurs either in the bulk liquid (homogeneous nucleation) or at the surface of a foreign material (heterogeneous nucleation) (Mullin, 1993, 2001). Activation energy (Mullin, 1993) is mostly diminished in presence of foreign surfaces due to nucleation process on them. This type of nucleation prevails and becomes the basis for the buildup of a fouling layer (Bott, 1994; Mullin, 1993; R. Rosmaninho & Melo, 2006; R. Rosmaninho, Visser, & Melo, 2002).

The influence of the surface energy of the support on the deposition process was studied by Rosmaninho et al. (R. Rosmaninho, et al., 2005) on different surfaces with a wide range of surface energy values and similar roughness and compositions. Reactive sputtering coating (with Ti and N) technique was applied for preparing all the surfaces of 316 2 R (bright annealed) based. It is a coating technique so all the surfaces became covered by a similar layer of TiN which makes them different mainly on their surface energy. The surfaces were named TiN 10, TiN 11, TiN 12, TiN 13, TiN 14 and TiN 15 (R. Rosmaninho, et al., 2005).

5.3 TiN surfaces

Fouling is dependent on several surface properties like roughness, surface composition and surface energy. Different stainless steel based surfaces with a wide range of surface energy values, having similar roughness and qualitative chemical compositions were used to investigate the influence of the surface energy on the deposition process. The investigated surfaces were 316 2R (bright annealed) based and prepared by using the technique of reactive sputtering coating (with different proportions of Ti and N in an Argon atmosphere). In this technique reactive sputtering of titanium (Ti, 99.7% purity) with an unbalanced magnetron cathode were used for coating on the stainless steel surfaces [where](#) Nitrogen (99.998% N₂ by volume) was used as the reaction gas (R. Rosmaninho, Rochab, Rizzoc, Müller-Steinhagenc, & Melo, 2007).

Rosmaninho et al. (R. Rosmaninho, et al., 2007) have tested all [the modified](#) surfaces in their work under varying operational parameters after obtaining from the same modification technique. Consequently, the different TiN sputtered surfaces had a similar morphology and surface composition although the varying proportions of Ti and N and different surface energy properties. The surface used for investigation could be distinguished and characterized based on their surface energy values (**Table 5**).

Table 5

With reference to **Table 5**, the most distinguishing factor among the surfaces is the electron-donor component (γ^-) and therefore this could be the characterizing parameter for the surfaces considered in works (R. Rosmaninho, et al., 2007).

The mass of calcium phosphate deposits on different surfaces were studied at two temperatures (44°C and 70°C) and 15, 30, 45, 60, 120 and 240 min time interval. The deposition trend of calcium phosphate was qualitatively very similar for all the surfaces and in all individual cases it is similar to an overall linear growth (R. Rosmaninho, et al., 2007). This does not seem to happen always, due to the growth rate and the structuring of the deposits at longer times (Britten, et al., 1988; Changan, et al., 1997). Zhao et al. (Zhao, Liu, Wang, Wang, & Müller-Steinhagen, 2005) have supported the observation and also noticed the long term effect of the surface nature (R. Rosmaninho, et al., 2007; Zhao, et al., 2005).

The trends presented in Figure 4 indicate that more deposit is initially formed on surfaces having higher γ^- . This dependency was informed in previous research with an indication of relation between affinities of the surfaces to nucleation of fouling deposits. Further the combination of size and number of the first aggregates of the foulant, calcium phosphate formed on the surface are dependent on surface energy values (γ^- component) (R. Rosmaninho, et al., 2005; R. Rosmaninho, et al., 2007; Roxane Rosmaninho, et al., 2007).

Figure 4

Effect of whey protein (β -Lg) on the fouling pattern of calcium phosphate of simulated solution (milk) was studied and evaluated by Rosmaninho and Melo (R. Rosmaninho & Melo, 2007). They have mainly considered its dependence on the surface energy of different modified surfaces used for study of deposition of materials. The deposition curves obtained in the presence and absence of protein were considerably different and the investigators have also observed the two growth periods at different time span in presence of the whey protein. Appearance of the second growth period after the delay time was dependent on the type of surface where fouling developed, more precisely on its roughness, surface composition and surface energy values (R. Rosmaninho & Melo, 2007).

Anti-fouling coatings based on nano-composites (**Table 6**) have been used to decrease fouling on plate and frame heat exchangers used in food processing plant (Kananeh, Scharnbeck, & Hartmann, 2009). The low-energy-surfaces led to a hydrophobic and oleo-phobic effect on the heating section of a pasteurizer equipped with four different coated plates were considered for study of fouling. The pasteurizer was operated with a 10% whey protein solution which was heated up to 85°C. Three coated samples of SS accumulated a reduced fouling amount in comparison to the uncoated stainless steel plates. The electro-polished plates showed a dramatic retardation (64% less) of fouling in comparison to the standard stainless steel plates whereas the coated plates showed about 30% less deposition. The time of cleaning in place (CIP) time of all coated plates was reduced (Kananeh, et al., 2009).

Table 6

The influence of the surface energy of the stainless steel substratum at similar roughness was studied by Rosmaninho and Melo (R. Rosmaninho & Melo, 2006) to **observed** the deposition process. In order to change their surface tension the SS 316 2R based materials were subjected to three types of surface modification processes (R. Rosmaninho & Melo, 2006; Santos, et al., 2004) such as Ion implantation, coated by plasma chemical vapor deposition and Nickel-Phosphor-Poly Tetrafluorethylene (Ni-PPTFE) coating (Augustin, Geddert, & Scholl, 2007; Beuf, et al., 2003a; Beuf, et al., 2003b; Kananeh, et al., 2009; Kananeh, Scharnbeck, Kückb, & Rübigerb, 2010; Kück, Hartmann, Manske, Kück, & Rübiger, 2007; R. Rosmaninho & Melo, 2006; R. Rosmaninho & Melo, 2007; Roxane Rosmaninho & Melo, 2008; R. Rosmaninho, et al., 2007; Roxane Rosmaninho, et al., 2007).

5.4 Surface treatment of heat exchangers in dairy industry:

- 1- Ni-P-PTFE (Nickel-Phosphor-Poly-Tetrafluorethylene) surface was the most promising one for non-microbiological deposits (calcium phosphate, β -Lg and fouling model fluid (FMF) milk-based product). It generally accumulates less deposit build up and in all cases, was the easiest to clean.
- 2- Considering food contamination on the basis of the data obtained from adhesion was the most suitable surface appeared as the ion implanted (TiC) surface, which also carries less spores after the cleaning process. Looking only at the number of adhered spores, the Ni-P-PTFE and the Xylan surfaces did not appeared to be as good as previously obtained TiC surfaces (Roxane Rosmaninho, et al., 2007).

To minimize milk fouling effects, Ni-P-PTFE coating seems to be the most appropriate surface treatment. This concluding remark is supported by the characteristics provided by the Ni-P-PTFE surface modification method and they are systematically different from the other methods, as reported by previous authors (Roxane Rosmaninho, et al., 2007; Santos, et al., 2004).

Rosmaninho et al. (Roxane Rosmaninho, et al., 2007) in the previous work have found, the Ni-P-PTFE coating as the only modification method which could clearly change the topography of the unmodified steel surfaces. On masking the grain boundaries at the surface these types of coatings create the thickest layer on the stainless steel surface (10 μm , against 0.2–2.5 μm for all the other surfaces). Rosmaninho et al. (Roxane Rosmaninho, et al., 2007) also reported that the thickness of the Ni-P-PTFE layer is light which failed to create a significant effect on the thermal resistance of the heat transfer wall (Roxane Rosmaninho & Melo, 2008).

Kananeh et al. (Kananeh, et al., 2010) reported that the fouling of gasketed plate heat exchangers in milk production has been reduced by use of nano-composite coatings. On the other hand an antifouling coating with low surface energy (low wettability) led to a hydrophobic and oleophobic effect. They have also investigated the performance of a number of coatings and surface treatments of heat exchanger used in milk processing. Certain polyurethane-coated plates and tubes received thinner deposit layer compared to that of standard uncoated stainless steel plates and tubes. They achieved 80% reduction in cleaning time of coated plates in comparison to the standard stainless steel one. Plates coated with different nano-composites as well as electro polishing were installed in the heating section of the pasteurizer. Polyurethane-coated plates exhibited the thinnest deposit layer due to the lowest total surface energy (Kananeh, et al., 2010).

Electro-polished plates also present the reduced deposit buildup in comparison to the standard stainless steel plates and were almost similar to the coated plates. Kananeha et al. (Kananeh, et al., 2010) observed 70% reduction in CIP time of electro-polished plates in comparison to standard stainless steel plates (Kananeh, et al., 2010).

To decrease fouling on gasketed plate heat exchangers in dairy industry, the nano-composites could be used as anti-fouling coatings. The coatings of different Polyurethane material had reduced fouling in industrial tests, though the PTFE coating showed higher fouling than the standard stainless steel plate. The deposit buildup on the electro-polished plates was lower than the standard stainless steel plates and almost similar to the coated plates. Finally they obtained a CIP time reduction for all coatings: PTFE coated plates curbed the fouling deposition by 90%; nano-composites coated plates reduced deposition by 70%; electro-polished plates diminished by 36% deposition. The results from an investigation in a pilot plant reveals that the coatings must be further developed so that they can withstand the thermal and mechanical stresses which arise in industrial operation (Kananeh, et al., 2010).

6. Milk Fouling Removal

Xin (Hong Xin, 2003) said the cleaning of the milk processing surfaces is an essential stage to remove the undesired materials. The cost of cleaning process is very high due to the high frequency of cleaning, hygienic requirement, the chemicals, energy, water, product losses, labour, and downtime. Pritchard et al (pritchard, et al., 1988) estimated up to 42% of available production time may have spent in cleaning process in dairy industry. Standard cleaning procedures for fouling deposits are well used for cleaning-in-place (CP) system. The procedure is

based on a controlled automatic circulation of formulated detergents in a certain section of a plant. An optimized CIP process can reduce the costs of cleaning, as well as the negative environmental impacts. Currently, most of the CIP processes are designed empirically due to lack of understanding of fundamental chemical and physical mechanisms involved in cleaning processes. Xin (Hong Xin, 2003) reiterated on investigation for developing a mathematical model to optimize cleaning processes.

At operation temperature lower than 100°C , during pasteurization process, protein is the main constituent of milk fouling deposits as a result of the heat-induced protein denaturation and aggregation reaction (deWit, Klarenbeek, & Adamse, 1986; Grasshoff, 1989; Lalande, et al., 1985). Due to the high heat transfer resistance and the difficulty of removal, proteinaceous fouling is a major concern for cleaning processes (Hong Xin, 2003).

In a typical cleaning study, a test set-up has to be used to develop fouling deposits and then it is cleaned by selected cleaning solutions. Concentrated whey protein is used to study fouling formation but the preparation is still not precisely controlled. In similar fouling procedure, significant variations of water content and amount of fouling have been reported (C.R. Gillham, 1997; Gotham, 1990), which makes the comparison of cleaning results very difficult due to the sensitivity of the cleaning rate on the composition and properties of fouling (Hong Xin, 2003).

Many monitoring methods have been developed for cleaning research. There are still lack of accurate on-line methods for study of suitable cleaning kinetics. Exploration of the fundamental cleaning mechanisms and optimization of the cleaning process are still a big challenge for researchers in this field (Hong Xin, 2003).

6.1 Chemical Cleaning Mechanisms

The resultant effect of three competing forces, namely the mechanical force exerted fluid flow, the adhesion force between soil and substrate and the cohesion force of the soil (strength of the deposits) governs removal of deposit from a hard surface in a chemical cleaning process. The introduction of the cleaning chemicals can change the adhesion and cohesion force of deposits with time. The diffusion and reaction of cleaning chemicals with the deposit can cause the swelling of the deposit. The cohesion force in the swollen deposit layer could be significantly reduced within some formed cracks.

Figure 5 depicts the three competing forces and possible chemical cleaning procedures. Normally, if the mechanical force is larger than the adhesion force, significant amount of deposits may be removed from the surface, which could be the most efficient cleaning procedure. The soil may be removed as small lumps, when the mechanical forces become larger than the cohesion force but smaller than the adhesion force. In these two consequences, the mechanical action controls the cleaning process. However, the dissolution process would be the controlling mechanism of cleaning when the mechanical force is smaller than both the adhesion force and the cohesion force. Xin (Hong Xin, 2003) stated that the concept of forces would help to differentiate the basic mechanisms of fouling removal due to the chemical reactions.

Figure 5

Milk protein is made of long molecule chains and is recognized as a biopolymer (Biddle, 1986). During a fouling process cross-linking reactions or chain reactions can produce protein aggregates with even larger dimensions. In the milk fouling the final surface gelation process indicates that milk protein is a form of polymer (Narasimhan, 2001; Hong Xin, 2003), which differentiates the dissolution character of milk proteinaceous fouling deposits from that of the mineral fouling deposits, which is crucial for understanding the chemical dissolution controlled cleaning process (Hong Xin, 2003).

In place cleaning of whey protein deposits by using alkali based cleaning solutions of 0.5 wt% NaOH over a range of surface temperatures ($20\pm 80^{\circ}\text{C}$) and flow rates of ($Re\ 500\pm 5000$) was investigated by Gillham et al. (C.R. Gillham, et al., 1999). They quantified cleaning by measuring both the **changes** in the thermal resistance of the deposits and the mass removal. The results confirmed that cleaning involved three stages, namely deposit swelling, uniform erosion and a final decay phase. Simultaneous measurements of thermal resistance and protein removal did not show a simple correlation and it can be informed that the existing models for cleaning require further development (C.R. Gillham, et al., 1999).

The mechanisms involved in the alkali-based cleaning of whey protein deposits on stainless steel surfaces during steady pipe flow have been investigated using mass removal rate, thermal resistance and surface imaging techniques (C.R. Gillham, et al., 1999).

During the uniform cleaning stage several of the observations are inconsistent with previous quantitative models for the protein removal rate. The fouling resistance results are not consistent with the models of some researchers (Bird & Fryer, 1991; Gallot-Lavallee & Lalande, 1985; C.R. Gillham, et al., 1999), suggesting that further development of quantitative models is

required before conducting rigorous optimization of the cleaning process (Bird & Fryer, 1991; Gallot-Lavallee & Lalande, 1985; C.R. Gillham, et al., 1999; Usepa, 2001).

The fouled heat exchanger surfaces were cleaned by three-stage cleaning method. After fouling run is over, in the first stage, the whey protein solution was drained and the system was washed with water at a velocity of 10.423 cm s^{-1} (for approximately 10 min), until there were no protein traces left in the stream water. The rinsing efficiency was estimated using information of the turbidity of flowing stream. The rinsing process was stopped at 0.5-1 NTU (Number of Transfer Units Method) turbidity of cleaning water (Fickak, et al., 2011; Usepa, 2001).

A cleaning solution (50 L of NaOH at 0.5 wt%) was used in second stage. During cleaning, the cleaning temperature of the cleaning solution was maintained constantly at 60 ± 0.5 °C. In the process of cleaning-in-place (CIP), the cleaning solution was recirculated through the system (Fickak, et al., 2011). The CIP solution was drained after complete removal of the fouling layer (Fickak, et al., 2011).

Fickak et al. (Fickak, et al., 2011) reported that the formation and dissolution rate of heat induced whey protein gels (HIWPG) is influenced by the whey protein concentration. It was noted that the structure of HIWPG became more rigid with the increasing protein concentration. The dissolution rate of HIWPG decreased with the increasing protein concentration and the decreasing rate is almost linearly against concentration (in the gel) increase (Fickak, et al., 2011). Fickak et al. (2011) also found the faster build up of fouling whey protein deposition with increasing fluid bulk whey protein concentration. They reiterated that the fouling under higher protein concentration will take much longer time to clean.

Hooper et al. (Hooper, Paterson, & Wilson, 2006) stated that the whey protein solutions could be a model system to generate consistent and reproducible fouling layers in the study of cleaning mechanisms. They have studied fouling in forced flows over heated surfaces and natural convective flows subjected to thermally induced gelation in sealed vessels. Specifically, deposits were generated from the same whey protein powder (30% WPC).

The cleaning behavior was observed by using fluid dynamic gauging (FDG) in three modes (Hooper, et al., 2006):

- 1- quasistagnant mode,
- 2- quasi-stagnant mode with fluid sampling,
- 3- duct flow with thermal resistance measurements.

Larger differences between the surface and deposits were observed under cleaning-in-place conditions within the same occasions (Hooper, et al., 2006). Hooper et al. (Hooper, et al., 2006) have investigated three deposit types and have shown variations in deposit structure and protein–protein interactions. They observed differences in cleaning behavior of deposits under all conditions.

7. Model for Milk Fouling

Mathematical models for fouling at low temperatures are existing but the behavior of systems at UHT temperatures is still unclear. Heat transfer coefficients and pressure drops were measured during fouling in all sections of the heat exchanger (P.J. Fryer, et al., 1996).

Nema and Datta (Nema & Datta, 2005) have developed a model, which can be used to control steam temperature or pressure in a helical triple tube heat exchanger to overcome the

drop in milk outlet temperature due to fouling. They raised the temperature of the wall gradually to counter heat losses due to fouling (X. D. Chen & Bala, 1998; X. D. Chen, et al., 2001). It is reported that the major contribution to the overall cost due to interruption of production is coming from cleaning. Thus the duration of the heating cycle tends to be maximum at the optimal solution (Toyoda, et al., 1994). Their proposed model may be useful for predicting the steam temperature or increase of pressure required for compensating the drop in milk outlet temperature as affected by fouling in a tubular heat exchanger. It could be implemented for commercial UHT milk sterilizers with suitable modifications (Nema & Datta, 2005).

Georgiadi and Macchiatto (Georgiadis & Macchiatto, 2000) used a fouling model which relies on the β -Lg reaction scheme as shown in **Figure 1**. The model was adopted from the work of Toyoda and Fryer (Toyoda & Fryer, 1997) and was first proposed by de Jong et al. (deJong, et al., 1992). Above 65°C , β -Lg of milk becomes thermally unstable and (i) transforms to molecular denaturation and exposes to reactive sulphydryl (-SH) groups and (ii) polymerizes irreversibly to produce insoluble particles in aggregated form (deJong, et al., 1992; Georgiadis & Macchiatto, 2000; Toyoda & Fryer, 1997). In the study of fouling, the key step is to understand the interrelationship between the chemical reactions which give rise to deposition and the fluid mechanics associated with the heat transfer equipment. The reaction scheme is described as follows (Georgiadis & Macchiatto, 2000):

- 1- In both the bulk and the thermal boundary layer in the milk the proteins reacted. In a first-order reaction the native protein N is transformed to denaturated protein D. The denaturated protein is then reacts to form aggregated protein A in a second order reaction.

- 2- From each protein, the mass transfer between the bulk and the thermal boundary layer takes place.
- 3- The aggregated protein is specifically deposited on the wall. The concentration of aggregated protein in the thermal boundary layer is proportional to the deposition rate.
- 4- The thickness of the deposit is proportional to the fouling resistance to heat transfer (Georgiadis & Macchiato, 2000).

The reaction rate constants are expressed in the common form as:

$$k = k_0 \exp(-E/RT) \quad (2)$$

For the two reactions the pre-exponential factors k_0 and the activation energies, E , are taken from de Jong et al. (deJong, et al., 1992) and presented in **Table 7**.

Table 7

8. Summary

Thermal processing of fluid in dairy industry is an energy concern matter. Heat exchanger scaling there diminishes heat dissipation, enhances pressure drop and reduces efficiency which ultimately degrades the economic operation of the plant. Product quality could possibly be deteriorated due to fouling and lack of proper heating. Milk fluid fouling chemistry is qualitatively understood. Now it is needed to intensify research on gathering quantitative information on milk fouling. Heat transfer coefficient and pressure drop study was conducted by many researchers during fouling in all types of the heat exchangers. Many research works were conducted to explore mitigation of fouling on heated surfaces. Among them, surface modifications and changing of heat exchanger configurations are notable. The present review highlighted information about recorded works on fouling and influencing parameters of the fouling factor including mitigation approach. Further research on fouling will enable to find ways on fouling mitigation in milk and food processing industries.

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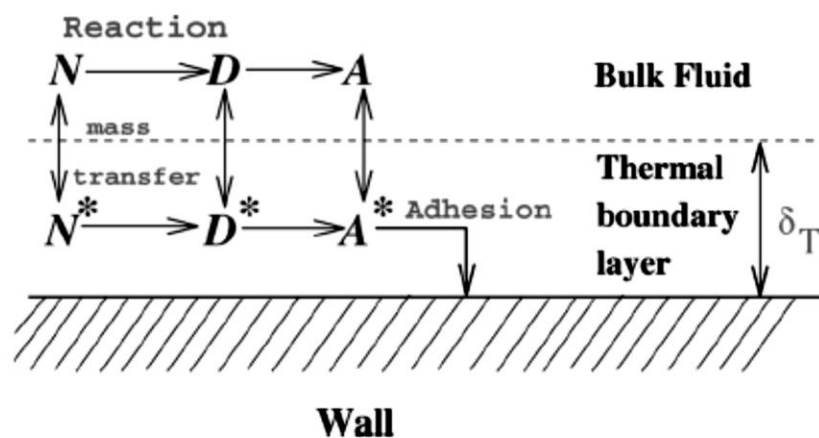


Figure 1 Model of proteins and salts deposition on the heat exchanger surface (Georgiadis and Macchiatto, 2000; Youcef et al., 2009)

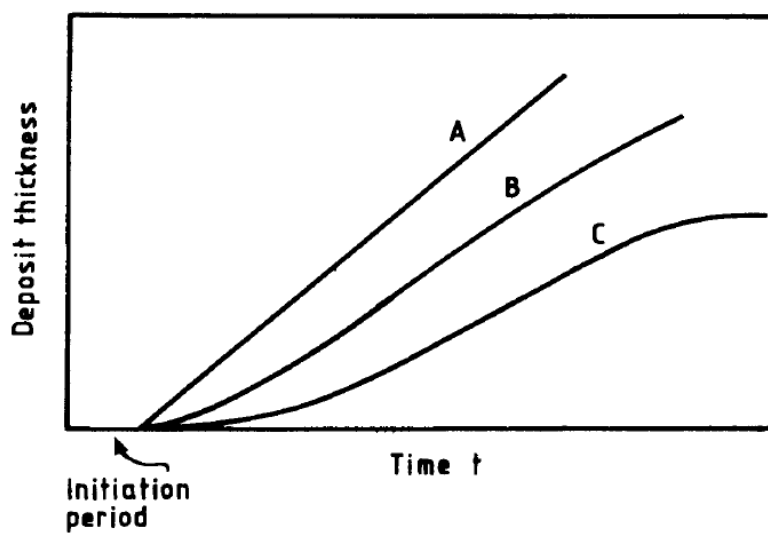


Figure 2 Idealized deposition curve (Bott, 1995)

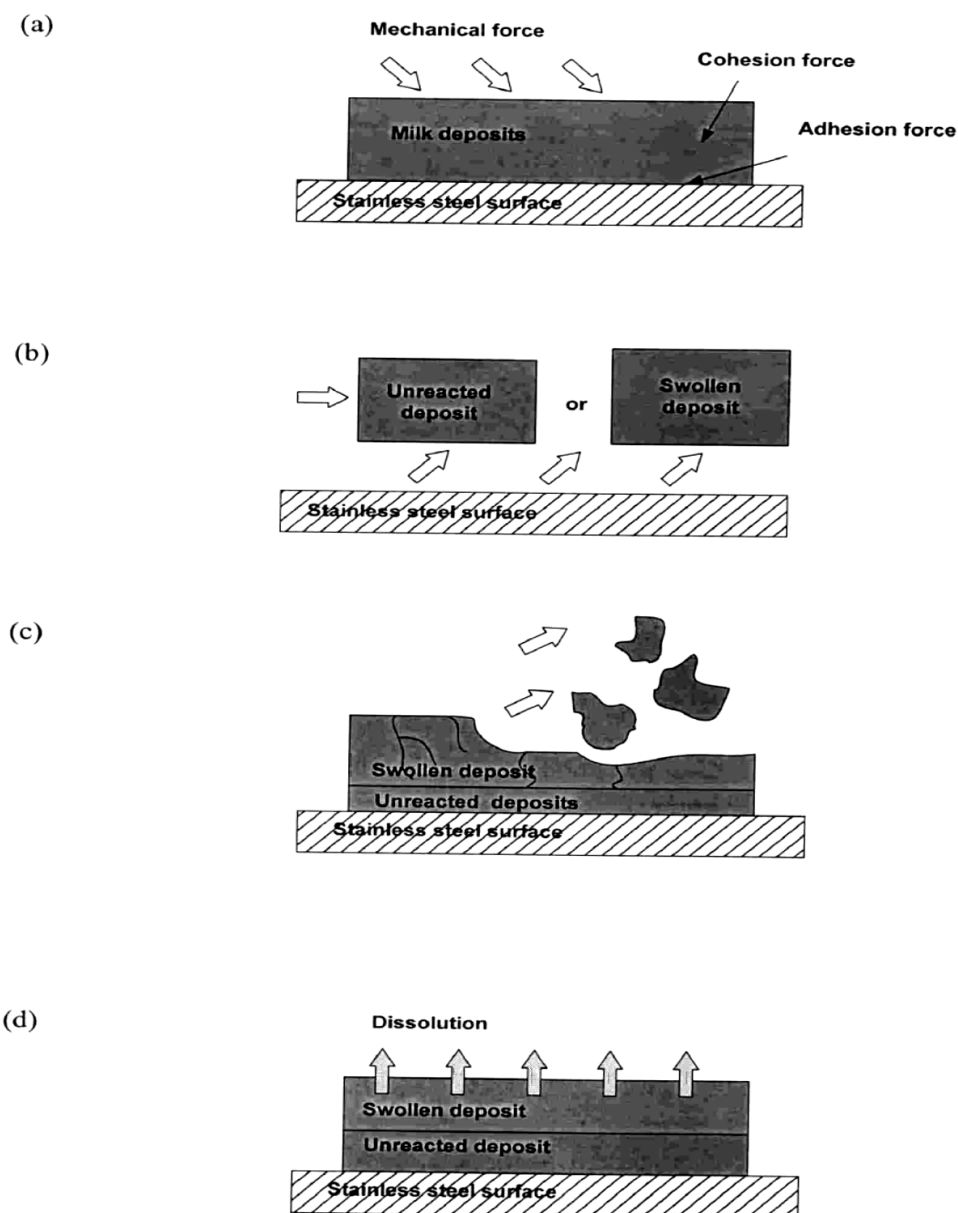


Figure 3 Schematic illustrations of the forces involved in cleaning and related cleaning procedures (a) forces involved in cleaning; (b) detachment of large pieces of swollen and/or unreacted deposits; (c) removal of small lumps; (d) dissolution process (Xin, 2003).

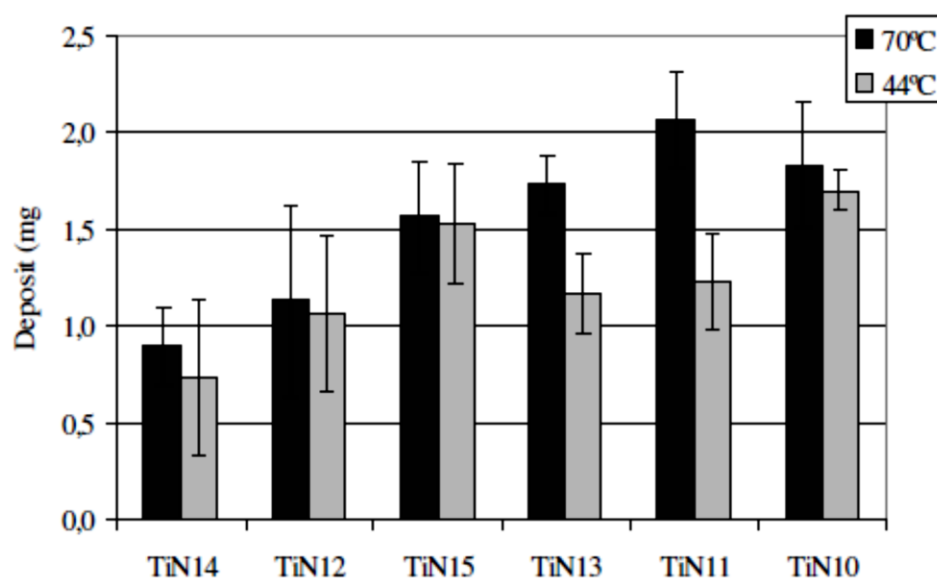


Figure 4 Deposit mass formed after 15 min of deposition which is the first mass detected at 44 and 70°C. Surfaces are placed from left to right in an increasing order of their surface energy, more precisely their γ^- parameter (Rosmaninho et al., 2005)

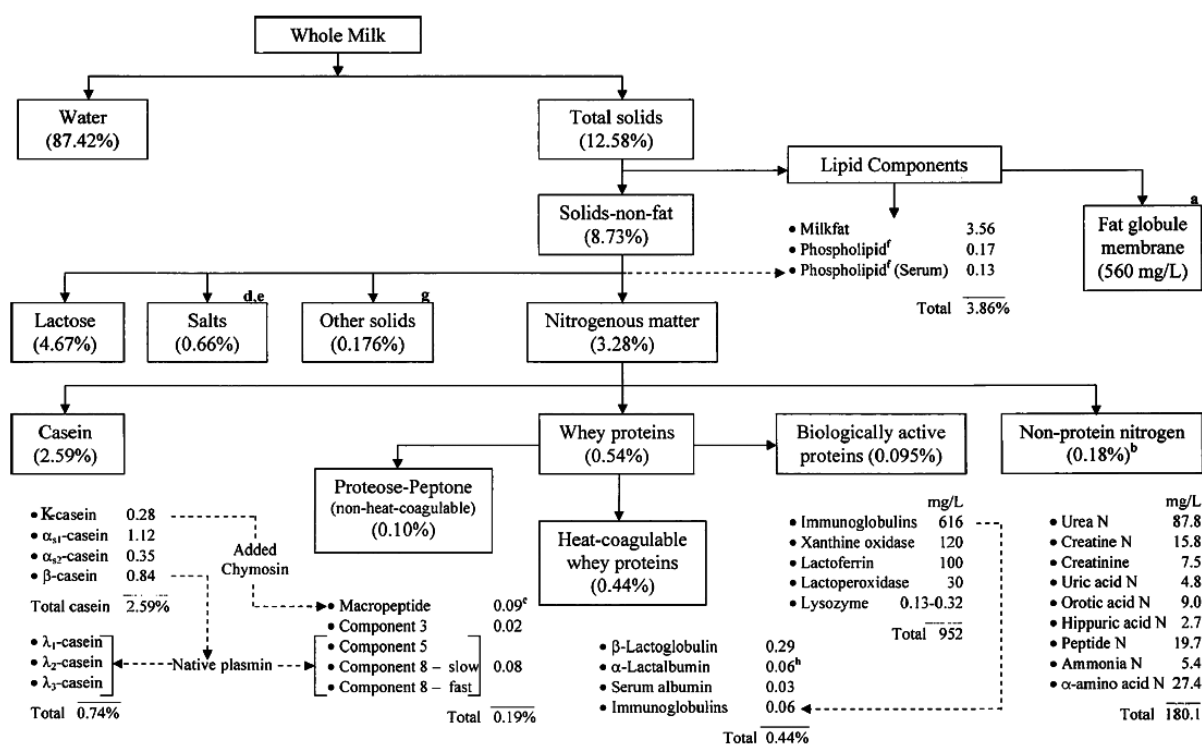


Figure 5 Approximate distribution of the components in milk. Reproduced with permission from Wiley-VCH, Verlag GmbH, Germany (Modler, 2000)

Table 1 Important aspects of fouling mechanisms (Bansal and Chen, 2006)

Aspects	References
Protein denaturation is reversible	de Wit and Swinkles (1980), Anema and McKenna (1996), Changani and others (1997), Chen and others (1998), Polat, Z. (2009)
Protein denaturation is irreversible	Ruegg and others (1977), Lalande and others (1985), Arnebrat and others (1987), Gotham and others (1992), Roef and de Kruif (1994), Karlsson and others (1996), Polat, Z. (2009)
Protein aggregation is irreversible	Mulvihill and Donovan (1987), Anema and McKenna (1996), Changani and others (1997), Chen and others (1998)
Protein denaturation is the governing reaction	Lalande and others (1985), Hege and Kessler (1986), Arnebrant and others (1987), Kessler and Beyer (1991), de Jong and others (1992)
Protein aggregation is the governing reaction	Lalande and René (1988), Gotham and others (1992), Delplace and others (1997)
Formation of protein aggregates enable to reduce fouling	de Jong and others (1992), Delplace and others (1997), van Asselt and others (2005)
Only protein aggregates cause fouling	Toyoda and others (1994)
Fouling is considered to depend on protein reactions only	de Jong and van del Linden (1992), de Jong and others (1992), Belmar-Beiny and others (1993), Delplace and others (1994, 1997), Schreier and Fryer (1995), Grijspeerdt and others (2004), Sahoo and others (2005), Nema and Datta (2005)
Fouling is considered to depend on protein reactions as well as mass transfer	Toyoda and others (1994), Georgiadis and others (1998), Georgiadis and Macchietto (2000), Chen and others (1998a, 2000, 2001), Bansal and Chen (2005), Bansal and others (2005), Bansal and Chen (2006)

Table 2 Average composition of milk (Bansal and Chen, 2006)

Constituents	Average concentration (%)
water	87.5
Total solids	13
Proteins	3.4
Lactose	4.8
Minerals	0.8
Fat	3.9
Proteins	3.4
Casein	2.6
β -Lg	0.32
α -La	0.12

Table 3 Composition of fouling layers from a selection of studies (Bennett, 2007)

Reference	Milk type	Milk temp (°C)	Equipment	Composition (% dry basis)
Lyster (1965)	Whole	85	Plate heat exchanger (regenerative section)	Protein: 60 Mineral: 25 Fat: 12
Lalande et al. (1984)	Whole	65-70	Plate heat exchanger (regenerative section)	Protein: 50 Mineral: 40 Fat: 1
	whole	120-138	Plate heat exchanger (regenerative section)	Protein: 15 Mineral: 75 Fat: 3
Fung (1998)	Whole	4-90	Tubular heat exchanger	Protein: 32 Mineral: 5 Fat: 50
	Whole (damaged)	4-90	Tubular heat exchanger	Protein: 32 Mineral: 4 Fat: 49
Tissier et al. (1984)	whole	72	Pasteurizer	Protein: 50 Mineral: 15 Fat: 25
	whole	90	Sterilizer	Protein: 50 Mineral: 40 Fat: 1
	whole	138	Sterilizer	Protein: 12 Mineral: 75 Fat: 3
Yoon & Lund (1989)	whole	88	Plate heat exchanger (preheat)	Protein: 43 Mineral: 45 Fat: ND
	whole	120	Plate heat exchanger (steriliser)	Protein: 45 Mineral: 40 Fat: ND
Calvo & Rafael (1995)	whole	80	Plate heat exchanger (heating)	Protein: 52 Mineral: 9 Fat: 23
Grandison (1988)	whole	110-140	Plate heat exchanger	Protein: 19-44

			(regenerative and heating)	Mineral: 57-20 Fat: 1-28
Jeurnink et al. (1989)	whole	85	Tubular heat exchanger	Protein: 64 Mineral: 18 Fat: 15
	whole	120	Tubular heat exchanger	Protein: 43 Mineral: 49 Fat: 3
Delsing & Hiddink (1983)	Skim	76	Tubular heat exchanger	Protein: 78 Mineral: 17 Fat: -
Jeurnink & Kruif (1995)	Skim	85	Plate heat exchanger	Protein: 44 Mineral: 45 Fat: -
Skudder et al. (1986)	Whole	80-110	Plate heat exchanger (regenerative)	Protein: 51 Mineral: 20 Fat: 6
	Whole	110-140	Plate heat exchanger (heating)	Protein: 22 Mineral: 53 Fat: 5
Ma et al. (1998)	Whole	85	Tubular heat exchanger	Protein: 20 Mineral: 4 Fat: 45
	Skim	85	Tubular heat exchanger	Protein: 64 Mineral: 13 Fat: -
Johnson & Roland (1940)	whole	82	Tubular heat exchanger	Protein: 35 Mineral: 5 Fat: 52
Truong (2001)	Whole	110	Downstream from direct steam injector (rig)	Protein: 39 Mineral: 8 Fat: 39
	whole	105	Downstream from direct steam injector (plant)	Protein: 63 Mineral: 20 Fat: 3

Table 4 Types of milk deposits formed at different processing temperature ranges (Xin, 2003)

Classification	Temperature	Process	Composition (wt%)		
			Protein	Mineral	Fat
Type A	75-110°C	Pasturisation	50-70	30-50	4-8
Type B	110-140°C	UHT treatment	15-20	70-80	4-8

Table 5 Surface energy components of the TiN sputtered surfaces (Rosmaninho et al., 2007)

surface	γ^{LW} (mJ/m ²)	γ^+ (mJ/m ²)	γ^- (mJ/m ²)	γ^{TOT} (mJ/m ²)
TiN 1	43.2 (0.1)	0.7 (0.0)	55.3 (0.0)	55.70 (0.1)
TiN 2	43.6 (0.2)	1.3 (0.0)	23.0 (1.8)	54.3 (0.2)
TiN 3	43.4 (0.1)	1.0 (0.2)	46.2 (4.6)	56.7 (0.4)
TiN 4	43.4 (0.1)	1.3 (0.1)	18.3 (2.7)	53.2 (0.4)
TiN 5	42.80 (0.9)	1.0 (0.9)	26.0 (2.2)	53.0 (0.6)

Table 6 Fouling surface sample specifications used in the experiments conducted by Kananeh et al. (Kananeh et al., 2009)

Substrate	Material	Thickness (µm)
SS	Stainless steel	-
EP	Electrically-polished stainless steel	-
A2	Epoxy-resin based coating of INM	83.7
A9	Polyurethane based coating of INM	53.0
A10	Polyurethane based coating of INM	85.2
A67	Polyurethane based coating of INM	27.6
PTFE	Teflon	22.5

Table 7 Kinetic data for the reactions of β -Lg (deJong et al., 1992; Georgiadis and Macchiatto, 2000)

E_N (kJ/mol)	K_{No} (1/s)	E_D (kJ/mol)	K_{Do} (m ³ /kg s)
261	312	$3.37 \cdot 10^{37}$	$1.36 \cdot 10^{43}$