# EFFECT OF HOMOGENISATION ON MILK FOULING IN A TUBULAR HEAT EXCHANGER

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## *Abstract*

*Fouling of equipment surfaces in milk processing has been a costly problem for many years. In spite of an increasing body of knowledge of the fouling mechanism, the problem is not fully understood yet. Recent investigations suggest that the role of fat in whole milk fouling seems to be very important. The state and form of the fat globules, processing conditions as well as the orientation of heating surfaces may affect the fouling mechanism.*

*Homogenisation of milk is known to cause disruption of fat globules and prevent creaming. The present work aimed to investigate the effect of homogenisation on the rate of fouling, composition and structure of fouling layers.*

*Homogenised and un-homogenised milk were used as test fluids. Milk was heated from 4 °C to 60 °C in a plate heat exchanger then to 70 °C and 80 °C in a double pipe heat exchanger consisted of a horizontal and a vertical tube. The fouling rate in the double pipe heat exchanger was calculated and expressed as the rate of increase of the overall resistance to heat transfer, normalised using the initial heat transfer coefficient at the beginning of the run.*

*Composition analysis of fouling layers was carried out using standard methods of moisture, ash, fat and protein tests. Resistance to deformation analysis was performed using texture tests; coverage measurement was determined by digital image analysis.*

*Within the experimental conditions used in this work, the effect of homogenisation on the fouling rate could not be ascertained conclusively because of large variations in the values obtained but it had a significant effect in the composition of fouling layers. In all experimental runs, the amount of fat in the fouling layer was higher for un-homogenised milk compared to homogenised milk. In fact, the fat contents of fouling layers were found to be very high (between 30%-60% on a dry weight basis), which agrees with observations of other researches in New Zealand.*

*The coverage and thickness of fouling layers were more influenced by the orientation of heated surfaces than by homogenisation. The strength of fouling layers is affected by their thickness, which decreases with increasing milk temperature.*

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## *Table of Contents*

# *List of Figures*

## *List of Tables*

# *Chapter 1 Introduction*

Liquid milk is a major food item in all developed dairying countries, representing 30% of total milk production (Fox and Guinee, 2000). The remainder is processed into one of several thousand products, making dairy products the most diverse and flexible group of food products. The flexibility of milk as a raw material resides in the chemical and physico-chemical properties of its constituents, many which are unique and therefore, there is the need for specific knowledge of milk constituents and the effects of various processing treatments on them.

In fresh whole milk the fat is present in the form of globules that are surrounded and stabilised by their own membranes. This membrane can be altered during processing, e.g. by homogenisation, where the newly created fat surface is coated by adsorbed proteins, mainly caseins (Walstra and Oortwijn, 1982; McCrae *et al.*, 1994).

Homogenisation lies at the heart of dairy processes. It improves the mouth-feel of consumer milk and distributes the cream throughout the milk, thus preventing the formation of a cream line.

Milk fouling in heat exchangers is a very important issue for the dairy industry. Fouling reduces the rate of heat transfer from the heating medium to milk and increases the pressure drop across the heating equipment used. In addition, fouling layer can act as a harbour for bacteria growth, potentially compromising milk product's sterility and safety (Hinton et al., 2002). Plant cleaning involves the use of expensive chemicals and combined with the reduced process run times, means that high rates of fouling can have a significant economic impact for the milk processor.

The fouling deposit mainly consists of fat, proteins and minerals. The composition of fouling layers in most overseas studies is dominated by proteins (β- lactoglobulin) and minerals (calcium phosphate). However, recent studies (Truong *et al,* 1996; Fung, 1998 and Ma and Trinh, 1999) have shown that fouling layers from New Zealand milk contain considerably more fat, between 40% and 60%, than overseas (4% -8%). The reason of this difference is still being studied.

Previous studies have shown that fat plays an important role on fouling. The rate of fouling increases with the increasing amount of fat present in milk (Ma *et al.*, 1999) as well as the amount of damage to the fat globule membrane (Fung, 1998). However it is still unclear how homogenisation could affect the fouling behaviour.

The objective of this work was to investigate whether the changes caused by homogenisation in the structure of fat globules does have an effect on milk fouling. Two areas of study were included: effect of temperature and effect of orientation of heating surfaces. The rate of fouling, composition and strength of fouling layers were used as parameters of measurement of fouling.

This work is divided into five sections. Chapter one outlines the fouling problem and its implications in the dairy industry. Chapter two gives a general assessment of the effect of heating on the different milk components as well as the role of milk fat globules in the formation of fouling deposits. Chapter three describes the resources and methodology used for the development of this research. Chapter four shows and explains the results obtained in this work. Final conclusions and recommendations for further work are given in chapter five.

# *Chapter 2 Literature Review*

## 2.1 INTRODUCTION

The manufacture of almost all milk and dairy-based products involves some type of heat treatment such as pasteurisation or preheating. The main objectives of the heat treatment are:

- To kill microorganisms and inactivate enzymes to improve the products' safety and quality (pasteurisation).
- To modify the product's functional properties (preheating).

### 2.1.1 Composition of milk

Milk is oil in water emulsion; milk fat globules are dispersed in a continuous aqueous phase called milk plasma. The principal constituents of milk are proteins, lipids and salts, lactose and the minor components including vitamins and traces of indigenous enzymes.

#### *2.1.1.1 Milk proteins*

Bovine milk contains eight principal proteins, 6 of which are synthesised in the mammary gland and are milk-specific; the 2 remaining are transferred from the blood, perhaps selectively. The milk-specific proteins are 4 caseins: $\alpha$-$s_1$, $\alpha$-$s_2$, $\beta$- and $\kappa$, representing approximately 38, 10, 36 and 15% respectively of whole casein (Fox and Guinee, 2000) and the principal whey proteins, $\beta$-lactoglobulin and $\alpha$-lactalbumin, which represent 40 and 20% respectively of the total whey proteins (Fox and Guinee, 2000). The minor whey proteins, bovine serum albumin and immunoglobulin are transferred from the blood and each represents 10% of the whey proteins. The remaining 10% is mainly non-protein nitrogen and trace amounts of protein including many indigenous enzymes.

#### *2.1.1.2 Milk lipids*

The lipid fraction of milk is comprised mainly of triglycerides (98%), with 1% phospholipids and small amounts of diglycerides, monoglycerides, cholesterol esters and traces of fat-soluble vitamins and other lipids (Fox and Guinee, 2000). The lipids occur as globules, 0.1-20μm in diameter, surrounded by a layer of emulsifier, referred to as the milk fat globule membrane (MFGM). More detailed information can be found in section 2.1.3.

#### *2.1.1.3 Milk salts*

Some of the salts in milk are fully soluble but others, especially calcium phosphate, exceed their solubility under the conditions that exist in milk and occur partly in the colloidal state, associated with the casein micelles; these salts are referred to as colloidal calcium phosphate (CCP), although some magnesium, citrate and traces of other elements are also present in the micelles.

## 2.2 Effect of heating on milk

### 2.2.1 Protein denaturation

The most notable effect of heating milk is protein denaturation. The conformation of a folded protein molecule is determined by its unique sequence of amino acids and by the effect of environmental conditions on amino acids side chains; this conformation is very stable because it has the lowest free energy (Bloomfield, 1979). This is called the native state of the protein (Mangino, 1984).

Denaturation is a drastic change from the native conformation, where the amino acid sequence is not altered.

Mild heat treatments (up to 60°C) mainly affect hydrophobic bonding within and between proteins. Such effects are important in those milk proteins, which have large hydrophobicities, such as β-casein and β-lactoglobulin (de Wit and Klarenbeek, 1984). The

severity of heating depends on the kind of product made, and milk proteins are affected accordingly.

### *2.2.1.1 Whey proteins*

- **β-lactoglobulin**

The most prevalent protein in whey is β-lg. It comprises 10% of the total milk protein or about 58% of the whey protein. β-lg is very pH sensitive. Its heat stability decreases as pH is increased from 3 to 7.5. When milk is exposed to prolonged heating, the thiol group of the protein becomes available for reaction and the protein can be involved in a series of complex inter and intra-protein thiol-disulfide interchanges reactions or reactions with other proteins (Wong *et al.*, 1988). Heating of β-lg to denaturation temperature (>60°C) allows polymerisation via disulfide bonds and binds to κ-casein (Singh and Fox, 1987).

- **α-lactalbumin (α-la)**

With a denaturation temperature of 62°C, this protein is the least stable of the whey proteins and it requires the largest amount of heat per gram for unfolding. Removal of calcium ions makes unfolding of α-la irreversible.

### *2.2.1.2 Caseins*

The caseins exist in very complex structures, known as casein micelles. Micelles aggregate initially when heated and then dissociate until the onset of coagulation, when rapid and extensive aggregation occurs (Fox, 1981). Strands of protein (β-lg) are formed between casein micelles after heating for 30 min at pH 6.8 and 100°C. Cross- linked β-lg molecules are attached to κ-casein on micelle surfaces by disulphide bonds (Creamer *et al.*, 1978).

The distinguishing property of all caseins is their low solubility at pH 4.6. The common compositional factor is that caseins are conjugated proteins, with phosphate group(s) esterified to serine residues. These phosphate groups are important to the structure of the

casein micelle. The caseins have a high surface hydrophobicity owing to their open structures, that make them extremely heat stable.

Casein is solubilized at UHT (Ultra high temperatures) conditions, up to 154.4°C for 9s (Morgan and Mangino, 1979). Less intense treatments (from 137.8°C for 1 second) cause serum proteins (those found in the liquid in which fat globules and casein micelles are dispersed) to precipitate with the casein during centrifugation.

When casein is heated, inorganic phosphate is released. Lowering the pH of milk to 4.6 solubilizes colloidal calcium phosphate; this removes its neutralizing effect, allowing electrostatic interactions between micelles. Under these conditions micelles coagulate and precipitate from solution.

- **$\alpha$-$s_1$ casein**

This protein contains two hydrophobic regions, containing all the proline residues, separated by a polar region, which contains all but one of eight phosphate groups. It can be precipitated at very low levels of calcium.

- **$\alpha$-$s_2$ casein**

This protein can also be precipitated at very low levels of calcium.

- **β-casein**

β- casein is very hydrophobic and more temperature sensitive than other caseins. Low temperature or removal of calcium causes dissociation of β-casein from the micelle and destabilizes the remaining micelle (Carpenter and Brown, 1985).

- **κ-casein**

κ-casein is very resistant to calcium precipitation, it provides the stability for the casein micelles. Rennet cleavage eliminates the stabilizing ability leaving a hydrophobic portion (Para- κ-casein) and a hydrophilic portion called κ-casein glycomacropeptide (GMP).

### *2.2.1.3 Serum albumin and Immunoglobulins (IgM)*

Its denaturation temperature is 64°C. Serum albumin precipitates between 40°C and 50°C as a result of hydrophobicity-directed unfolding. Some serum albumin remains undenatured even after prolonged heating at 65°C, probably because already denatured albumin is able to protect native proteins from denaturation (Terada, 1980).

Immunoglobulins are very heat unstable, especially below pH 6 (de Witt and Klarenbeek, 1984). IgM, unless heat denatured, acts as a specific agglutinin against some streptococci strains (Mulder and Walstra ,1974).

## 2.2.2 Casein and serum proteins interactions

When milk is heated at temperatures in excess about 70°C, whey proteins are denatured and they associate with casein micelles (Singh and Fox, 1987). Complexes are formed between denatured whey proteins and casein micelles involving κ-casein when milk is heated at 90°C to 140°C and at pH below 6.7. (Figure 2.1).

Depending on the severity and type of heating, different types of associations between casein micelles and whey proteins occur (Joshepson, 1987). κ- casein is involved in the interaction between casein micelles and whey proteins and disulphide interchange reactions are involved in this interaction (Singh and Fox, 1987).

But at higher pH values, whey proteins remain in the intermicellar fluid as fibrous strands (Singh and Fox, 1987). Whey proteins interact with κ-casein in the micelles at pH above 7.3 and this complex tends to dissociate from the casein micelles; however, when milk is cooled and pH is adjusted, the complex is reassociated with the casein micelles (Singh and Fox, 1987).

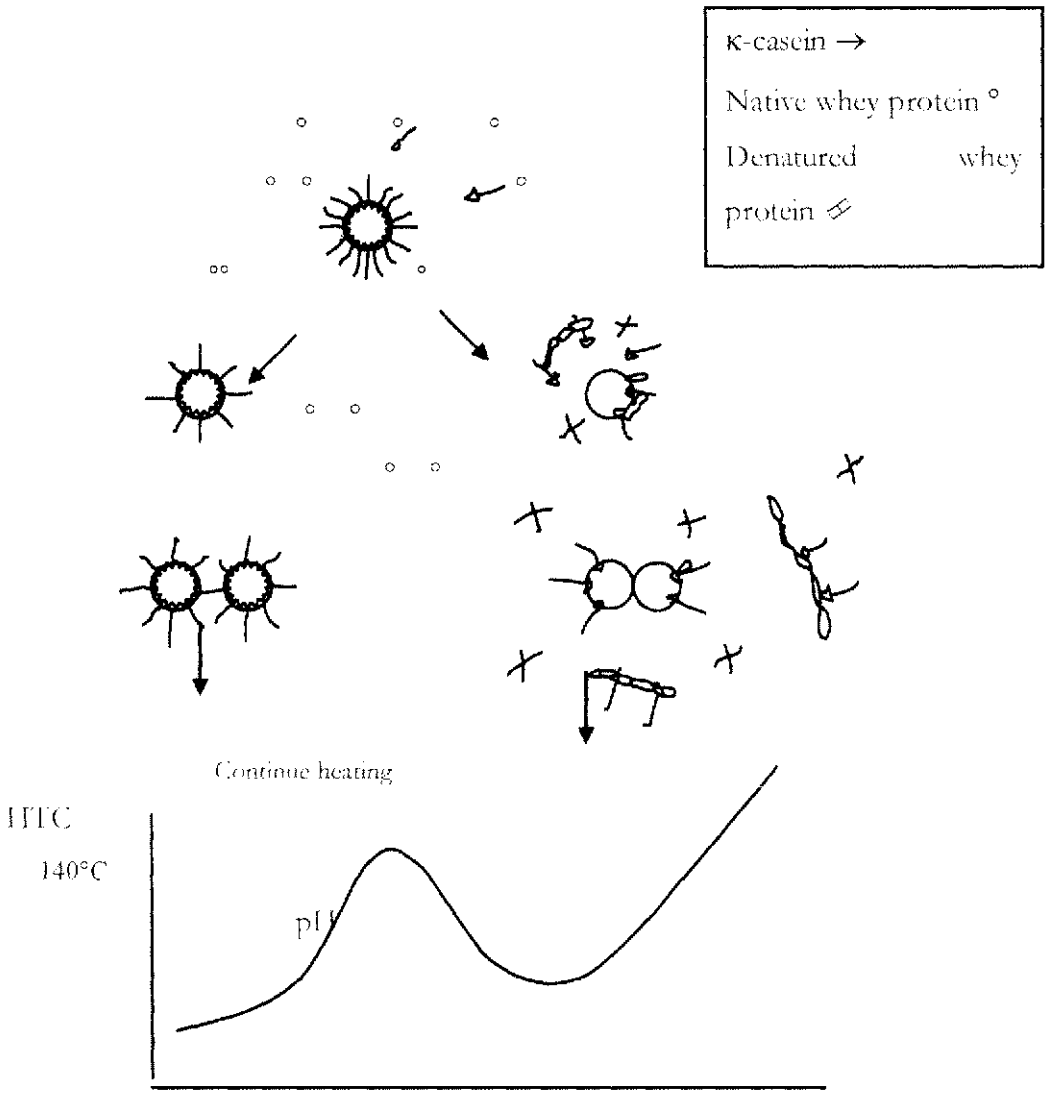



**Figure 2.1 Diagrammatic representation of the effect of heat on the proteins of normal skim milk at different pH values, the whey proteins denature and form a complex with k-casein, which adheres to the micelle surface (whey protein coated micelles). Further heating causes lowering of the pH, desphophorylation of caseins and other reactions. The coagulation is caused by the formation of covalent cross-links between the whey protein aggregates that adhere to the micelle surface. (II). At higher pH values, the whey protein-k-casein complex is largely free in the serum. The micelles have swollen a little and soluble casein is present. The micelles have less k-casein and are sensitive to calcium ion concentrations. (Singh and Fox, 1987. In Advanced Dairy Chemistry Vol 1).**

### 2.2.3 Effect of heating on the milk fat globule membrane (MFGM)

In whole milk, fat is present in the form of globules surrounded by a membrane whose main constituents are phospholipids and proteins. Several researchers have reported changes in the milk fat globule membrane (MFGM) during thermal processing (Mulder and Walstra, 1974; Corredig and Dalgleish, 1996).

Heating whole milk at 80°C for 2.5-20 minutes has been reported to cause interaction between skim milk proteins (β-lg and κ-casein) and milk fat globule membrane (MFGM) components. Such heat treatments result in compositional changes to the membrane system, with the most extensive changes observed after 20 minutes (Houlihan *et al*, 1992).

During heating of un-homogenised milk, denatured serum proteins (β-lg and α-la) bind to the fat globules, either by direct adsorption to the fat or by interaction with the MFGM (McPherson and Kitchen, 1983; Dalgleish and Banks, 1991; Corredig and Dalgleish, 1996).

During heat treatment of milk high density complexes, higher in density than those found in the natural MFGM, are formed. Also losses of natural membrane polypeptides from the medium and low-density lipoproteins are observed upon heating (Houlihan *et al*, 1992).

It is well recognized that homogenisation of milk causes profound physical changes in the size and structure of the fat globules (section 2.4). When milk is homogenised, little or no whey protein may adsorb onto the fat-water interface unless the milk is heated (Sharma and Dalgleish, 1993). Furthermore, the amount of serum proteins found in the homogenised MFGM is affected by the stage at which heating is applied (Sharma and Dalgleish, 1993).

### 2.2.4 Maillard Reactions

Maillard reactions occur when amino acids, peptides or proteins condense with the sugar and act as their own catalysts for dehydration; the reactions of degradation of sugar take place under mild heat treatments and at pH near neutrality. In milk the main potential Maillard reactants are lactose and lysine residues from the proteins.

## 2.3 FOULING

The deposition of a layer of milk components, mainly proteins and minerals, on the surface of the heat exchanger during heating of milk is called fouling. Jeurnink *et al*, (1996) showed that such deposits had undesirable side effects: decreased heat transfer coefficients; increased pressure drop, hence a decrease in pumping efficiency; loss of product remaining on the heated surface; loss of vitamins, minerals and other nutrients present in the fouled layer; increased cleaning costs and environmental load. Fouling may also enhance the adsorption of bacteria to the surface and by encapsulation in the deposit; these microorganisms become protected against cleaning agents and affect the overall quality of the finished products (Hinton *et al.*, 2002).

### 2.3.1 Chemistry of milk fouling

β-lactoglobulin (β-lg) and α- lactalbumin (α-la) are the major constituents of whey proteins. β-lactoglobulin denaturation becomes significant at temperatures above 65°C, showing a strong correlation with the rate of fouling deposition (Hege and Kessler, 1986; Jeurnink, 1995b); that is why whey solution has been used as a model solution in the study of fouling. Although α-lactalbumin also denatures and associates with other proteins during heating, its influence in the fouling process is not very significant.

The presence of calcium phosphate in the fouling deposits is due to its decreasing solubility upon heating. The temperature difference between the bulk and the surface partly influences the precipitation of these minerals. However, part of the mineral precipitates with other proteins. In its ionic form calcium influences the protein deposition by its effect on the aggregation of whey proteins and on the stability of casein micelles (Jeurnink *et al.*, 1996).

### 2.3.2 Factors affecting fouling rate

#### *2.3.2.1 Product Parameters*

- **Composition of milk**

The fouling process can be affected by variations in the composition or the chemistry of the milk components. Factors like feeding regimes, stage of lactation, changes in the concentration of whey proteins can affect the fouling formation and composition (Schraml, *et al.*, 1996).

- **Acidity of milk**

Decreasing the pH of milk (from 6.8 to 6.4) causes a strong increase of fouling due to additional deposition of caseins (Patil and Reuter, 1988; Jeurnink et al, 1996). The deposition of protein and fat is increased but the deposition of minerals is reduced in the case of whole milk. Patil and Reuter (1988) showed that the increased deposit formation of pH-reduced milk is mainly due to the reduced stability of protein to heat.

- **Age of milk**

Aged milk produces more fouling in a heat exchanger than fresh milk does. Experiments with skim milk (Jeurnink, 1991) and whole milk (De Jong *et al.*, 1992) have shown that the action of proteolytic enzymes, produced by psychotropic bacteria, is responsible for the increase of fouling. Recent studies carried out using UHT milk (Datta *et al.*, 2002) showed that the amount of protease required for causing sufficient proteolysis in milk during storage at room temperature is very small. Reliable detection and measurement of such a low levels of activity is difficult.

- **Seasonal Variation**

Grandison, (1988) showed that the rate of fouling varied greatly throughout the year, and the weight of deposit obtained in a laboratory plate heat exchanger also varies with the

season. Burton (1968), followed the seasonal variation in the amount of deposit formed from the milk of single herds; he found that there is a minimum in late spring and early summer (May-July), with a broad maximum in the winter months from September to March in the northern hemisphere.

- **Dissolved air and gases**

Heating decreases the solubility of air in milk. If the local pressure is too low (below the saturation pressure) air bubbles can arise (Burton, 1988). Mechanical forces produced by valves, expansion vessels, etc, could induce the formation of air bubbles. If the bubbles are formed at the heating surface, they act as nuclei for the formation of deposit. Due to evaporation milk is transported from the bulk to the surface where the air-vapour bubble is attached (figure 2.2). Here milk protein accumulates and form a deposit on the surface (Jeurnink, 1995a).

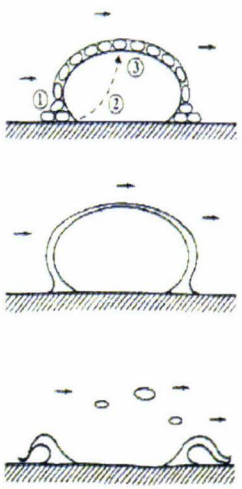



**Figure 2.2 Schematic representation of an air bubble in the fouling process of milk on a hot stainless steel surface. (1) Adsorption/deposition at the vapour/ liquid interface; (2) evaporation; (3) condensation. ➔ Flow direction for the milk. (Jeurnink, 1995 a).**

The composition of the deposit is influenced by the evaporation of the boundary layer of the air bubbles, which are associated with caseins, resulting in increased casein content in the fouling layer (Jeurnink, 1995 a).

- **Calcium**

During milk processing, calcium plays a role in the heat stability of milk as well as in the formation of fouling. This is not only because the solubility of calcium phosphate decreases with heating but also because the denaturation of β-lg and the precipitation of caseins is influenced by the presence of calcium (Delsing and Hiddink, 1983).

Experiments have shown that either increasing or decreasing the calcium concentration in milk leads to lower heat stability and to more fouling in comparison with that in normal milk (Jeurnink and Kruif, 1995). In addition, there is a shift in the protein composition of the deposit from serum proteins to caseins. Obviously, the increased instability of the casein micelles, with denatured β-lactoglobulin at their surface, causes an increase in protein fouling (Jeurnink *et al*, 1996).

### *2.3.2.2 Process parameters*

- **Product Flow velocity**

Flow velocity along with temperature may be the most important variables in the fouling process. As velocity increases, the viscous sub-layer close to the wall becomes thinner thereby reducing the resistance to diffusion and transfer of foulants from the bulk towards the wall, which allows a relatively faster rate of deposition. On the other hand, as velocity increases, the shearing forces, which tend to remove the deposits, are increased. Belmar-Beiny et al. (1993) demonstrated that increasing the Reynolds number lead to less fouling.

- **Temperature**

Increasing the gradient of temperature (ΔT) between surface and bulk liquid, gives more fouling, especially due to additional calcium salt precipitation. Foster and Green (1990) showed that the total amount of deposits increased with surface temperature between 80°C-140°C (figure 2.3). Hegg and Lund (1985) also showed that the fouling behaviour of

β-lg in the temperature range of 50°C-140°C is greatly affected by the gradient of temperature.

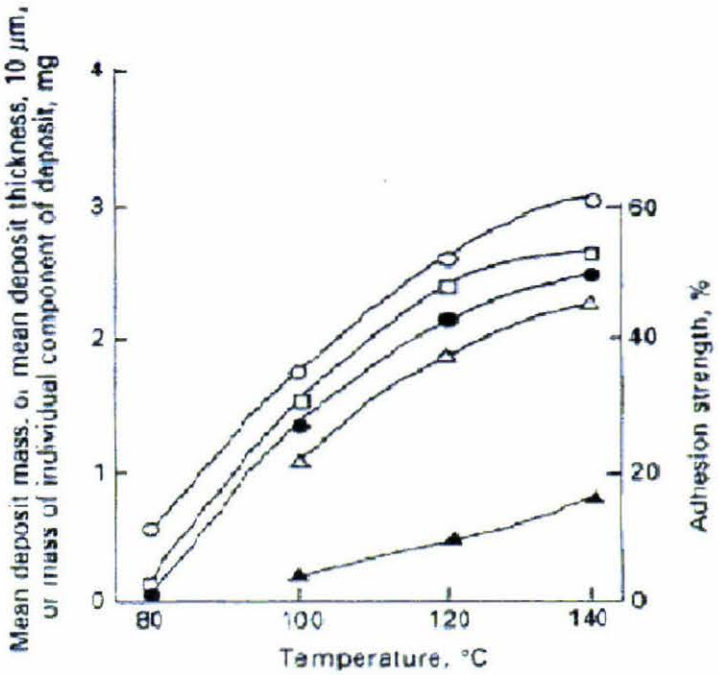



**Figure 2.3 Effect of temperature of surface of formation of deposits. (O), Mean deposit mass, mg: (•), mean deposit thickness, 10μm: (Δ), mass of protein mg: (▲), mass of inorganic phosphate. mg: (□) adhesion strength, %. (From Foster and Green, 1990)**

- **Pre-treatment**

The higher the preheat temperature, the lower rate of deposition formation. If milk passes without delay from the earlier regenerative stage into the final heater, the maximum amount of deposit occurs at a relatively low temperature, and the deposit then decreases to a comparatively small quantity at the heater outlet. (Burton, 1968).

- **Concentration of fouling material**

When the concentration of proteins is increased, an increase in deposit was expected (Schraml *et al.*, 1996). The increase was only found up to 25% total solids. The increased

salt deposition inhibited the formation of a porous structure. The crystallizing salts adhered to groups still reactive on the proteins, result in a reduction of the amount of protein adhering to the deposit (Delsing and Hiddik, 1983; Schraml *et al.*, 1996).

### 2.3.3 Surface and bulk reactions

A fouling process consists of a series of steps involving mass transfer and reaction. If the process occurs as a result of reaction, mass transfer may be the controlling variable. If bulk processes are involved, fouling will occur from a sequence of steps: (i) denaturation and aggregation of proteins in the fluid; (ii) mass transfer to the surface; (iii) incorporation of protein into the deposit, i.e. surface reaction and (iv) possible transfer of proteins back to the bulk (Belmar-Beiny *et al.*, 1993).

If a combination of mass transfer and chemical reaction is involved in the deposit formation, the slowest of these processes will be the rate-controlling step. Belmar-Beiny *et al.* (1993) described two cases to identify the controlling step:

If fouling is mass transfer controlled, the slowest process will be the transfer of reacted protein to the surface. In this case, the rate of deposit formation will not be a strong function of the temperature.

If fouling is controlled by denaturation reactions, deposit formation will be a function on the temperature where the controlling reaction takes place. Reactions in a number of different places could control the process:

*Surface reaction.* If only surface processes control fouling, deposition will occur wherever the wall temperature is hot enough for protein denaturation and aggregation to occur, regardless of the bulk temperature. The process will be a function of the wall temperature.
*Bulk reaction.* If the controlling reaction for fouling takes place in the bulk, two conditions can be proposed:

- If the wall temperature is such that protein denaturation and aggregation will occur, but the bulk temperature is such that native protein is thermally stable, fouling will result

from the deposition of protein which has been denatured and aggregated in the thermal boundary layer adjacent to the wall.

- If both the boundary layer and the turbulent core are hot enough for protein denaturation and aggregation, protein denatured and aggregated in both regions will contribute to deposit formation.

It has been proven by Belmar-Beiny *et al.* (1993) that fouling process is not a mass transfer controlled reaction; it is reaction controlled with both bulk reaction and surface reaction.

### 2.3.4 Structure of fouling deposits

Several studies have been carried out (Tissier and Lalande, 1986; Foster *et al.*, 1990) to describe the structure of the fouling layer. First, the solid heated surface is covered with a sub-layer; then, after a given time, granules of aggregates and fat globules randomly appear on this layer. Finally, the surface is overlaid with a spongy structure that contains minerals and aggregated proteins. Fat globules, casein micelles and microorganisms can be enclosed to this structure.

### 2.3.5 Composition of fouling deposits

Depending of the intensity of heating, Burton (1968) identified two distinct types of deposits (A and B); Type A is a soft, bulky material that is formed at temperatures between 75°C and 115°C (Lalande *et al*, 1984). Owing to the high protein content (50-70%), this type of fouling is known as protein fouling. Type B is formed at temperatures above 110°C; it is hard and has a granular structure with a high mineral content (up to 80%), and is known as a mineral fouling.

Moreover, Ma *et al.*1999, showed that "A" type deposits could be further divided into three different subtypes based on the bulk temperature as seen in table 2.1.

**Table 2.1 Classification of type "A" fouling deposits**

| Subtype | Temperature°C |
|---|---|
| I | 40-65°C |
| II | 65-75°C |
| III | 75-85°C |

Deposit type A tends to restrict the area of the flow passages in the heating equipment and causes an increase in the operating pressure and is therefore of major importance for process optimisation.

Deposit type B has more influence on heat transfer than on the pressure drop across the equipment. Its high concentration of minerals influences the ease of removal of deposits during the cleaning procedures. (Schraml *et al.*, 1996). Both types of deposit contain small amounts of fat (4-12%) and lactose (Foster and Green, 1990). However, higher fat contents in fouling deposits have been reported depending of the treatment conditions and milk treatment, as shown in table 2.2.

**Table 2.2 Summary of fat compositions in fouling deposits**

| Literature | Processing condition/milk treatment | Fat content |
|---|---|---|
| Burton (1968) | Direct heat treatment | 34% |
| Skudder et al. (1986) | Heating 100- 105°C | 15-20% |
| Jeurnink et al. (1996) | Homogenised Recombined milk | 4-8% |
| Fung (1998) | Milk fat globule membrane damage | More than 45% |
| Ma and Trinh (1999) | Milk solutions of different fat contents | 36-68% |

### 2.3.6 Fouling by different milk components

#### 2.3.6.1 Serum proteins

As soon as a serum protein solution, even at room temperature, comes into contact with a stainless steel surface immediately a monolayer of protein adsorbs. (Jeurnink *et al*, 1995).

Protein fouling is controlled by some intermediate of the denaturation of serum proteins taking place in the bulk fluid. The deposition process on top of a monolayer occurs in three steps:

- Formation of fouling intermediate in the bulk of the solution: When whey protein is denatured by heat its sulphydryl groups become highly active and could react with disulphide bonds. (Present in α-la, β-lg, serum albumin, immunoglobulins and κ-caseins).

- Transport to the surface: During this step, a denatured molecule of β-lg can be made unavailable for fouling through aggregation with another activated β-lg molecule in the bulk or with another milk components like fat globules or κ-casein at the surface of the casein micelles.

- Deposition: Not all denatured β-lg molecules are involved in the formation of deposit; only the protein immediately adjacent to the heated surface would be deposited whilst the bulk protein could form polymers of β-lg or react with κ-casein (Skudder *et al.*, 1986).

### *2.3.6.2 Minerals*

The mineral substances in milk are in dynamic equilibrium with the caseins. Their distribution between the serum and colloidal phases of milk is dependent on the ionic environment. Mineral salts affect milk functionality through specific and non-specific interactions with the milk proteins, thus influencing the structure and stability of the proteins (Augustin, 2000).

Calcium phosphate, which is an integral part of the casein micelles, plays an important role in the fouling process due to its decreasing solubility upon heating.

### *2.3.6.3 Caseins*

At room temperature, almost all the casein molecules in milk are associated into casein micelles. Upon heating to 80-90°C, both the size and the mutual interactions of these micelles increase through association with β-lg (Jeurnink *et al*, 1996). There is also an increase in the concentration of κ-casein in the serum phase due to its dissociation from the casein micellar surface.

If the colloidal stability of casein micelles in milk is decreased, the deposition process may be controlled by coagulation of them and not by denaturing serum proteins (Jeurnink *et al*, 1996).

When the colloidal stability of the casein micelles is decreased (by proteolytic action or lowering the pH) there is an increase in fouling as well as a decrease in the heat stability of the milk. Hence, the mechanism for deposition of casein micelles upon heating milk may be similar to that of the heat coagulation of milk (Jeurnink *et al*, 1996).

### *2.3.6.4 Fat*

Jeurnink *et al.* (1996) showed that fat is present in fouling layers (4-8%), and it plays a minor role in the process of fouling. However, researchers in New Zealand found fat contents of more than 45% in dry fouling deposits. (Fung, 1998; Ma and Trinh, 1999; Ma *et al.*, 1999).

Ma and Trinh (1999) studied the role of fat on fouling using milk solutions with different fat contents. They stated that during heating of milk, fat globules tended to migrate towards the wall because of the hydrophobic attraction between the fat globules and the stainless steel surface. The more fat present in the milk, the easier they will migrate towards and attach to the wall.

It is not clear why the fat content in milk fouling layers is much higher in New Zealand milk than those reported overseas. This phenomenon could be linked to different compositions and/or to different handling practices.

#### *2.3.6.5 Lactose*

Lactose is hardly found in milk deposits because it is soluble in water. So even if it were incorporated in a deposit it would dissolve, either in the milk itself or in the water of the first rinsing step of the cleaning. Only at high temperatures (>100°C), when caramelization or Maillard reactions take place, lactose can be found in the deposit (Jeurnink *et al.*, 1996).

### 2.3.7 Stages of fouling

Fouling is a kinetic process; the resistance to heat flow caused by fouling varies with the time. Fryer (1989), found that fouling occurs in three phases (figure 2.4):

- **Induction phase**

During this period conditions in heat transfer and pressure drop remain nearly unchanged; According to the trials carried out by Delplace *et al.*, (1994) in a plate heat exchanger (PHE) using whey protein solutions as a model fluid, the length of this phase was approximately one hour.

Extension of the induction period would improve the operation of industrial plant. It is therefore important to determine the sequence and the rate of events that make up the induction stage (Belmar-Beiny and Fryer, 1993).

- **Fouling phase**

There is a steady growth of deposit on the surface. During this phase, the overall heat transfer coefficient decreases and pressure drop increases linearly with time.

- **Post-fouling phase**

During this period a constant value of the overall heat transfer coefficient was obtained (Delplace *et al.*, 1994).

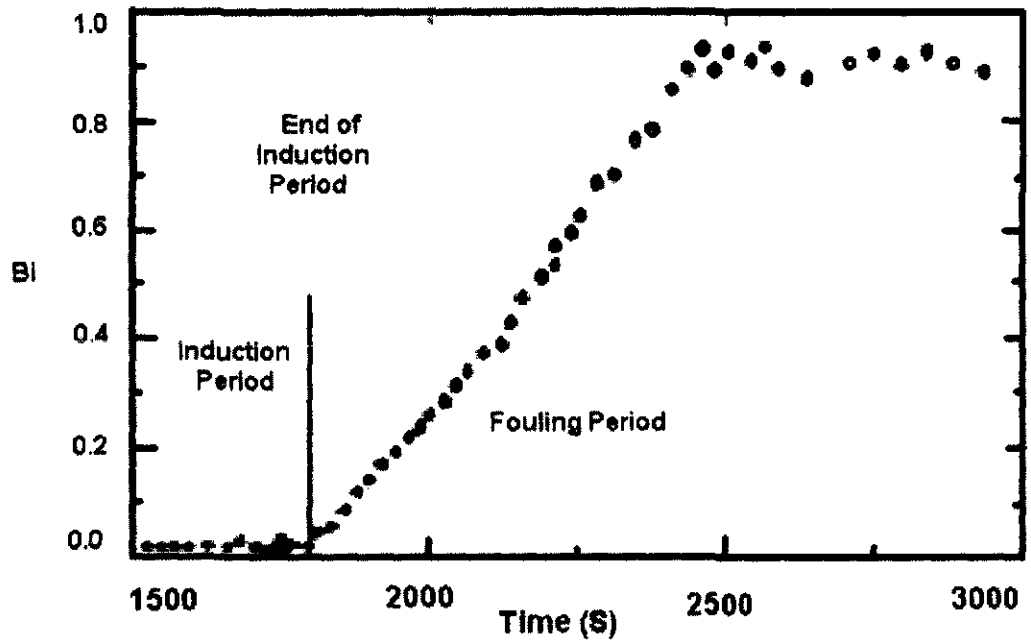



Figure 2.4 Experimental fouling curve (From Belmar-Beiny et al., 1993)

## 2.4 HOMOGENISATION

### 2.4.1 Introduction

Homogenisation is a mechanical treatment of the fat globules in milk which is applied by passing milk under high pressure through a tiny orifice, which results in a decrease in the average diameter of the milk fat globules to 3.5-4 μm or lower (McCarthy, 2001). The smaller globules are equally distributed throughout the milk plasma. As a consequence, there is an increase of 5 to 10 times in the interfacial area between the milk fat and the milk plasma. The purposes of homogenisation are (McCarthy, 2001):

- To avoid the creaming of fat globules (rising to the surface) or to prevent sedimentation of solid particles, for example, particles of denatured proteins in UHT milk (stabilization).

- To prevent partial coalescence (clumping) by improving the stability of fat globules. Three factors contribute to the enhanced stability of homogenised milk: a decrease in the size distribution of the fat globules (causing the speed of rise to be similar to the majority of globules such that they do not tend to cluster during creaming), a decrease in the mean diameter of the globules, given by the Stoke's relation (Senno, 1999. Fig. 2.5) and an increase in their density, bringing them closer to the continuous phase, owing to the adsorption of a protein membrane.

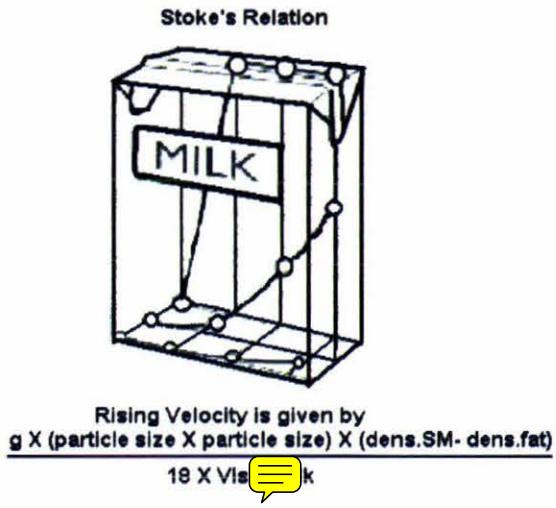



**Figure 2.5 Stoke's Relation. There is a correlation between the fat globule size and the creaming rate. (From Senno, 1999)**

- To obtain recombined milk products, especially recombined whole milk.

- To create desirable rheological properties. The formation of homogenisation clusters can greatly increase the viscosity of a product such as cream or yoghurt.

Homogenisation is usually achieved by applying intense mechanical agitation to the milk using a mechanical device known as a homogeniser (McClements, 1998).

### 2.4.2 Homogenisation Mechanism

Milk first enters the valve at a velocity of 4-6m/s, it then moves into the gap between the valve and the valve seat and the velocity is increased to 120 m/s in about 0.2 ms. The liquid

then moves across the face of the valve seat and exits in about 50μs. During this short time, fat globules are affected by hydrodynamic forces, such as shear forces caused by velocity forces; inertial forces caused by turbulent eddies (when these are of similar sizes of fat globules) and cavitations forces caused by shock waves as a consequence of the implosion of cavitations bubbles. The fat globules are affected by these forces causing at first, deformation and then a break-up when the deformation stress exceeds globule strength (McCarthy, 2001).

The homogenisation phenomenon is completed before the fluid leaves the area between the valve and the seat, and therefore emulsification is initiated and completed in less than 50μs.

The product may then pass through a second stage valve, similar to the first one. While most of the fat globule reduction takes place in the first stage, there is a tendency for clustering of the reduced fat globules. The second stage allows the separation of those clusters into individual fat globules.

### 2.4.3 Effect of homogenisation on the milk emulsion

#### *2.4.3.1 Emulsion Stability*

There are a number of physicochemical mechanisms responsible for the breakdown of the milk emulsion, the most important being creaming/sedimentation, flocculation and coalescence (figure 2.6). *Creaming* is the process in which milk fat droplets move upwards because they have a lower density than the surrounding liquid. *Sedimentation* is the process in which droplets move downwards due to gravity because they have a higher density than the surrounding liquid. *Flocculation* is the process in which two or more droplets "stick" together to form an aggregate in which the droplets retain their individual integrity. *Coalescence* is the process in which two or more droplets merge together to form a single large droplet. Partial coalescence is the process in which two or more partly crystalline droplets merge together to form a single irregularly shaped aggregate due to the penetration of a solid fat crystal from one droplet into a region of liquid oil in another droplet.

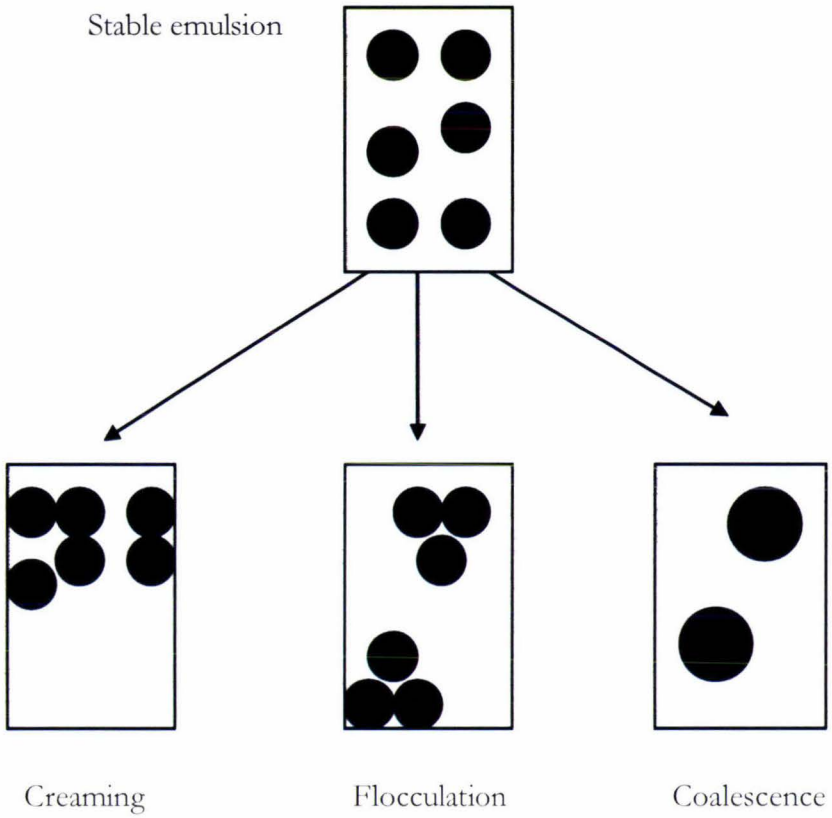



**Figure 2.6 Major mechanisms of emulsion instability (McClements, 1998)**

### *2.4.3.2 The Milk Fat Globule Membrane (MFGM)*

The MFGM lowers the interfacial tension resulting in a more stable emulsion. During homogenisation there is a tremendous increase in surface area and the native MFGM is disrupted. However, there are many surface-active molecules from the milk plasma that readily adsorb and form a new membrane, causing the interfacial tension to fall again. The new emulsion remains stable after homogenisation. The newly formed membrane consists mainly of casein micelles (partly spread) and whey protein (Walstra *et al*, 1999). Refer to figure 2.7.

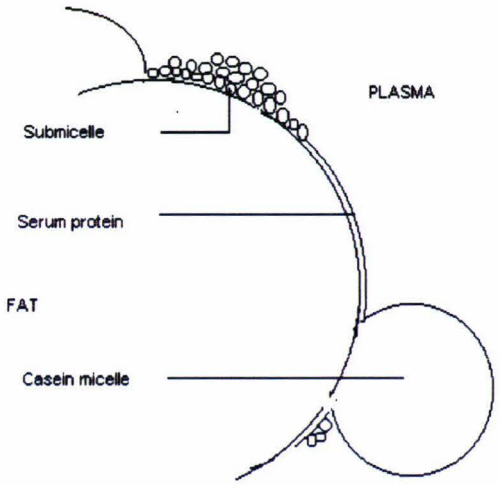



**Figure 2.7 The new fat globule membrane formed during homogenisation. (Walstra *et al.*, 1999)**

### *2.4.3.3 Homogenisation clusters*

A cluster is formed when a casein micelle is attached to two or more globules at the same time as a consequence of the collision of the partly uncovered fat globules during homogenisation. In the case of whole milk, cavitations and/or turbulence forces will break up such a cluster in the homogeniser valve. Clusters may persist if there is not enough protein to cover the new surface completely in the time available, as happens in cream.

Such clusters are called homogenisation clusters, which cannot be broken up by moderate agitation. They are 10-100μm diameter and may contain about a million globules (McCarthy, 2001). The use of a second homogenisation stage (30-50 bar) can prevent clustering.

#### *2.4.3.4 Lipolysis*

The natural fat globule membrane protects the fat globule from the attack of lipolytic enzymes and bacterial lipases. However, the new membrane formed during homogenisation, cannot give this protection because its main components are casein micelles, which are permeable to enzymes and they are bound to lipases that are brought into contact with the fat when casein micelles incorporate into the new membrane. (McCarthy, 2001).

To avoid lipolysis during homogenisation of raw milk, it is essential to pasteurise milk or cream in order to destroy the natural milk lipase. Milk often undergoes homogenisation immediately after pasteurisation.

### 2.4.4 Effect of homogenisation on fouling

During heating of homogenised milk in a tubular heat exchanger small fat globules tend to rise to the surface slower than large fat globules. So, it is expected that homogenised milk will produce less fouling depositions than un-homogenised milk, in which fat globules are larger than those found in homogenised milk.

The amount of fat globules attached to the fouling deposits in a tubular heat exchanger depends on the milk flow rate, composition of milk and bulk temperature (Ma *et al.*, 1999). However, the orientation of the heating surfaces in a tubular heat exchanger could influence the velocity at which, fat globules rise to the surface, and hence affect the fouling deposition.

This aspect will be studied in this project since there is no similar research investigating the effect of homogenisation on fouling.

## 2.5 CONCLUSION

The effect of heat treatment of milk components, especially proteins, is very well understood. However the role of casein micelles at high temperatures is still unclear.

There is evidence that heating of milk causes interactions between whey proteins and casein micelles (β- lacto globulin and κ- casein) and between proteins and milk fat globules. These interactions and the chemical changes to milk upon heating can affect the fouling process. Overseas studies have shown that fat did not play an important role on fouling but recent studies in New Zealand found that the fat present in milk has a major effect on the fouling rate and on the composition and structure of fouling deposits. The rate of fouling increased substantially with both the increased amount of fat in milk and the increased amount of damage done to milk fat globule membrane (MFGM). However, there is no information available on the effect of homogenisation (which, involves changes in the size of fat globules to prevent creaming) on fouling.

# *Chapter 3 Materials and Methods*

## 3.1 Milk

### 3.1.1 Types of milk used

Batches of pasteurised homogenised and un-homogenised milk (mixture of skim milk and cream), with similar fat and protein contents (about 3.3% and 3.4% respectively) were used during this study. Both homogenised and un-homogenised milk were required to have similar fat and protein contents to determine the effect of homogenisation (not of milk composition) on the fouling rate, composition and structure of fouling deposits.

Batches of pasteurised homogenised milk, skim milk and cream from the same factory run, packed in plastic bladders of 10 litres each were purchased from Mainland Products Ltd (Longburn, New Zealand) and then transported from Mainland processing plant to the Massey University pilot plant and stored at 4°C prior to experiments.

Un-homogenised milk (3.3% fat and 3.4% protein) was prepared by mixing skim milk and cream, as shown in appendix A1, in a stainless steel tank fitted with an agitator (Leroy Somer, IEC44-1, Longburn, New Zealand).

## 3.2 Milk powder pilot plant

### 3.2.1 Overview

The experimental work was performed in the milk powder pilot plant of Massey University. The plant is generic and has been used by eight postgraduate research students. Each user has the option to configure the plant or add extensions for his or her specific project. A schematic diagram of the equipment set up used in this project is shown in figure 3.1.

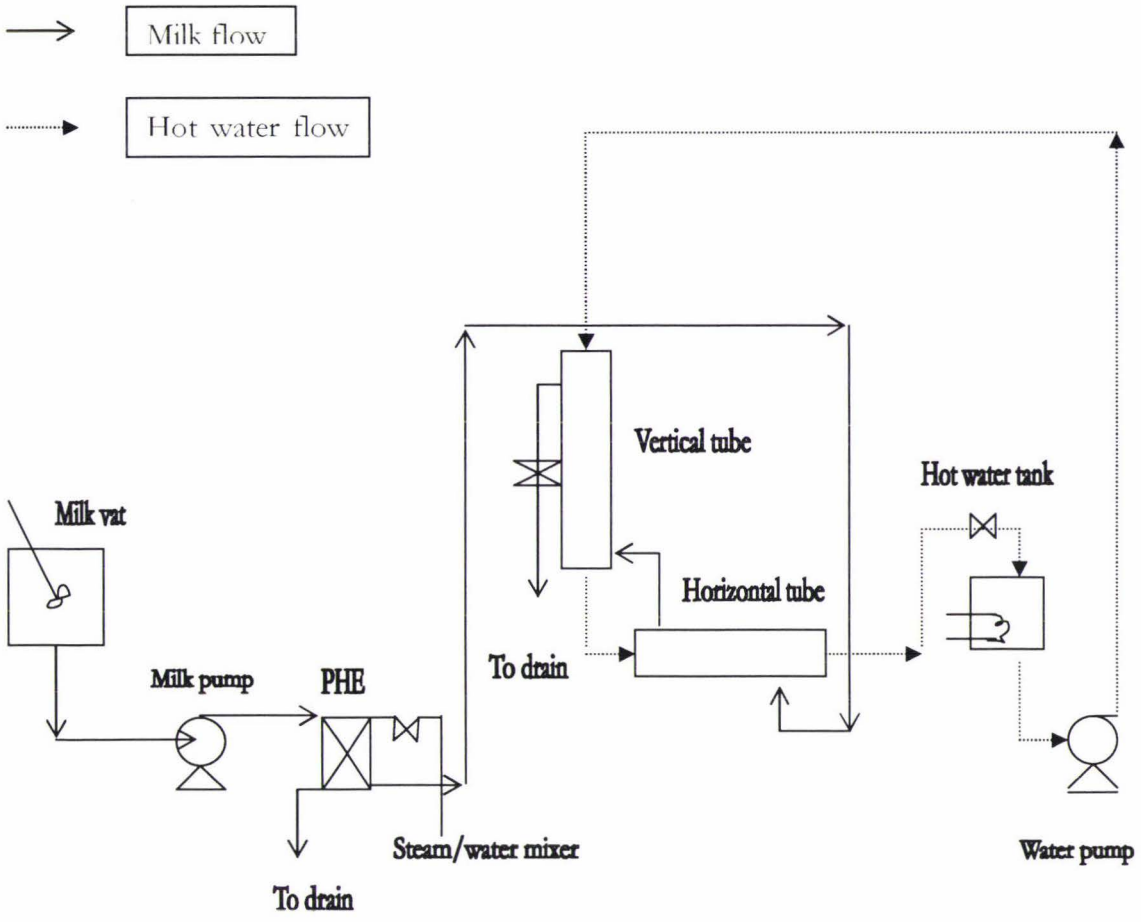



Figure 3.1 Schematic representation of equipment set up

### *3.2.1.1 Milk vat*

Milk was stored in a stainless steel tank of 850 litres capacity (diameter 1.4m, depth 0.61m). The tank was fitted with a refrigeration system to avoid milk spoilage and with an agitator paddle motor (Leroy Somer, IEC44-1, New Zealand) to prevent milk from freezing during storage.

#### *3.2.1.2 Milk pump*

A centrifugal pump (Ebara, CDX 70/05, 0.37kW) was used to circulate milk from the milk vat to the fouling tubes.

#### *3.2.1.3 Plate heat exchanger (PHE)*

A plate heat exchanger (Alpha Laval U-265-R, Denmark) was used to pre-heat the milk from 4°C to the desired temperature (60 °C, 70°C or 80°C) before it entered the fouling tubes. Hot water was used as the heating medium for the PHE, it was obtained by mixing steam and cold water in a steam-water mixer.

#### *3.2.1.4 Flowmeter*

Before entering the double pipe heat exchanger, milk passed through an electromagnetic flowmeter (Endress-Hauser Picomag, 11PM 165333, EMC, Industrial Instrumentation, New Zealand), which was required to measure a flow rate of about 45 litres/hr.

#### *3.2.1.5 Tubular heat exchanger*

The custom-built fouling rig consisted of two tubular heat exchangers: one horizontal tube and another vertical tube (Figure 3.1). The entire heat exchanger was 2 metres long, made up 2 one-metre lengths.

The horizontal tubular heat exchanger used for this research was the existing heat exchanger in the pilot plant, which was also used in other fouling studies (Hinton *et al*, 2002; Bennet, 2000; Ma *et al*, 1999).

The vertical tubular heat exchanger was designed and constructed with the same dimensions as the horizontal tube: inner pipe ID 12.7mm and outer pipe OD 25.4mm and 1 meter long (NZF Stainless Ltd).

A gate valve (Sanwa, 12.7mm) was fitted at the milk outlet of the vertical tube to control the back pressure of the fouling rig, which was kept at 50 kPag for all fouling experiments performed during this study.

Hot water flowed through the inner tube and milk flowed in the annular cross section. The inner tube, which external surface was covered by fouling, could be removed for collection of fouling deposits in both horizontal and vertical tubes at the end of each run.

Hot water was used as heating medium in this research. An electrical heater (Eutro, 3kW) installed in a stainless steel tank of 130 litres capacity was used to produce hot water. The hot water was circulated through the tubular heat exchanger and returned to the hot water tank by a centrifugal pump (Ebara, CDX70/05, 0.37kW).

The milk flow can be described as follows: Milk was stored at 4°C in the milk vat. Then it was pumped to the plate heat exchanger (PHE), where it was heated to 60°C, 70°C or 80°C (depending of the trial conditions). Once milk left the PHE, it passed through the flowmeter and then through the tubular heat exchanger reaching the horizontal tube first followed by the vertical tube, after which milk was drained.

#### *3.2.1.6 Thermocouples*

Eight type T thermocouples were used to measure the inlet and outlet temperatures of milk and hot water in both vertical and horizontal tubes. The thermocouples were installed in the tube socket weld unions and sealed with glue. A SCADA station (section 3.3.2) was used to log data from these thermocouples.

## 3.3 Process control

### 3.3.1 Process control computers

A micro-computer running Windows NT 4.0 (Intel Celeron, Advantage computers Ltd., Palmerston North, NZ) and a programmable logic controller (PLC), (Allen- Bradley SLC 500, Rockwell Automation Ltd., NZ) were used to control the milk powder pilot plant. The micro-computer displayed experimental data in real time which was automatically logged into disk. The software package used to display and log the data was FIX DMACS version 7.0 (Intelluction Inc., Industrial Interface Ltd., New Zealand).

### 3.3.2 SCADA Station

An auxiliary mobile SCADA (Supervisory Control and Data Acquisition) station (Intel Pentium, Advantage Computers Ltd., New Zealand) running Windows 3.1 was used to log data from the thermocouples installed in the fouling tubes because of insufficient thermocouple ports on the main SCADA system. FIX DMACS Version 7.0 (Intelluction Inc., Industrial Interface Ltd, New Zealand) software was used to log and export the temperature data. Thermocouples were calibrated before each trial, using the procedure described in appendix A2, while connected to the SCADA Station.

### 3.3.3 Digital camera

Photographs of fouled tubes were taken using a digital camera (Kodak DC 290 Zoom, USA). The camera was connected to a television monitor to get a magnified preview of the image as well as help for obtaining focused images.

## 3.4 Fouling trials

A number of sighter fouling trials were performed to commission the equipment configuration and develop the experimental protocol. The fouling trials were performed

using the operating conditions described in table 3.1. Each trial is consisted of two runs, one with homogenised milk and one with un-homogenised milk, both performed on the same day; each run lasted 4 hours.

At the end of each run, the fouled tubes were removed from the heat exchanger and rinsed with water to rid off milk residues. The fouling rig was rinsed with water and drained.

### 3.4.1 Operating conditions

**Table 3.1 Operating conditions of fouling trials**

| Trial | Run (type of milk) | Milk temp | Hot water temp | Pressure | Flow rate | Time |
|---|---|---|---|---|---|---|
| 1 | Homogenised | 60°C | 90°C | 50kPa.g | 45 l/h | 4 h |
| | Unhomogenised | 60°C | 90°C | 50kPa.g | 45 l/h | 4 h |
| 2 | Homogenised | 70°C | 90°C | 50kPa.g | 45 l/h | 4 h |
| | Unhomogenised | 70°C | 90°C | 50kPa.g | 45 l/h | 4 h |
| 3 | Homogenised | 80°C | 90°C | 50kPa.g | 45 l/h | 4 h |
| | Unhomogenised | 80°C | 90°C | 50kPa.g | 45 l/h | 4 h |

The plant was operated according to the Bennett-Downey protocol (2001), which included preparation of the pilot plant before runs and operation of the plant during the fouling trials. The detailed procedures of the operating protocol are confidential information of the Institute of Food, Nutrition and Human Health of Massey University.

#### *3.4.1.1 Procedures prior to experiments*

Before each fouling trial, the following procedures were carried out:

- **Cleaning of fouling rig according to cleaning-in-place procedures**

First, the system was flushed with water to wash away milk residues. 1% caustic solution was flushed through the rig and recycled for half an hour. The system was then flushed with water, followed by an acid wash with 1% nitric acid solution. Finally, the system was rinsed with water and drained.

- **Cleaning of heating surfaces (fouling tubes)**

The inner tubes of the tubular heat exchanger were soaked in 1% caustic solution for one hour, rinsed with water and then they were soaked in a nitric solution for one hour. The tubes were rinsed with water and scrubbed with a harsh cloth then rinsed with water again.

Tubes were allowed to dry in air, weigh and installed in the heat exchanger.

### 3.4.2 Collection of fouling deposits

Once the fouling run was finished, fouled tubes were removed from the tubular heat exchanger and photographed with a digital camera for coverage analysis (section 3.6.2).

Once photographs had been taken, the fouled tubes were scrapped using a knife to get long strips of fouling deposits. The wet layers were allowed to dry overnight at room temperature.

The dry fouling deposits from homogenised and un-homogenised milk were kept separately in plastic bags and then weighed in a scale (type BB 2400, Mettler). The weight of the empty plastic bags (initial weight) was taken away from the final weight of bag to determine the total amount of fouling deposits.

Finally, the dried fouling layers were grinded to fine powder to perform composition analysis (Ma *et al*, 2002).

## 3.5 Composition analysis

Fouling layers obtained from vertical and horizontal tubes were analysed separately. Each test of composition was conducted in duplicate using 1 or 2 grams of fouling layer depending on the amount of sample required for each test. Dry fouling strips of 20mm long were used to perform texture analysis (section 3.6.1).

### 3.5.1 Moisture determination

The moisture test was performed using the air-oven method, described in appendix A3.

### 3.5.2 Protein determination

Protein analysis of fouling deposits was performed using the Kjeldahl method described in appendix A4.

### 3.5.3 Fat determination

The method used to determine the fat content of fouling deposits was Mojonnier test which is a standard procedure from AOAC (1999) and is described in appendix A5.

### 3.5.4 Ash determination

The mineral content of fouling deposits was determined by combustion of a weighted sample in a muffle furnace (dry ashing). This method is widely used by the dairy industry and it is described in appendix A6.

## 3.6 Physical analysis

### 3.6.1 Texture analysis

The dried fouling samples were tested for resistance to deformation using a TA-XT2 texture analyser (Stable Micro Systems Ltd. UK) and a knife probe (TA 43 knife blade with flat 3mm end). The procedure is described in appendix A7. A computer program (Texture Expert for Windows) was used to record the data. This software allowed running tests as projects. Each project would retain instrument settings and graphical preferences once initially set. Results of force versus distance were saved as spreadsheets. Resistance to deformation of fouling layers was determined by calculating the slope of the force (N) versus distance (mm) curves.

### 3.6.2 Determination of fouling coverage

Photographs of the fouled tubes used in coverage analysis had to cover the whole area of the tubes to ensure accurate results. To achieve this, a background of 1200mm* 1300mm made of custom wood covered by a dark blue fabric was used. Lines of white string were placed on the board every five centimetres to get images with the same size. Two clips placed at both ends of the board held the fouled tube.

Coverage of fouling was determined using image analysis software, Sigma Scan Pro 5.0 that made a surface analysis of the photographs of the fouled tubes by measuring the pixel intensities of each image. The use of Sigma Scan Pro software during this study appeared to be a new method for measurement of coverage of fouling.

The detailed procedure of use of Sigma Scan Pro 5.0 for coverage analysis is given in appendix A8.

The uncovered area of the fouled tubes was calculated in percentage using measurements in pixels:

$$\% \text{ UA} = \frac{\Sigma u_{ai} + \ldots + u_{ai}}{\Sigma T_{ai} + \ldots + T_{ai}} \times 100$$

Where,

% UA = Total percentage of uncovered area of the fouled tube

$u_{ai}$= Uncovered area of each image (pixels)

$T_{ai}$ = Total area of the image (pixels).

# *Chapter 4 Results and Discussion*

## 4.1 Introduction

This chapter is divided into three sections. First, general observations are made on the effect of homogenisation on fouling by whole milk to highlight general trends. Then the effect of temperature as well as effect of orientation of heating surfaces are discussed. The results presented in this study suggested that homogenisation did not affect the fouling rate in the experimental conditions used but it had a major influence in the composition of fouling layers.

## 4.2 Effect of homogenisation

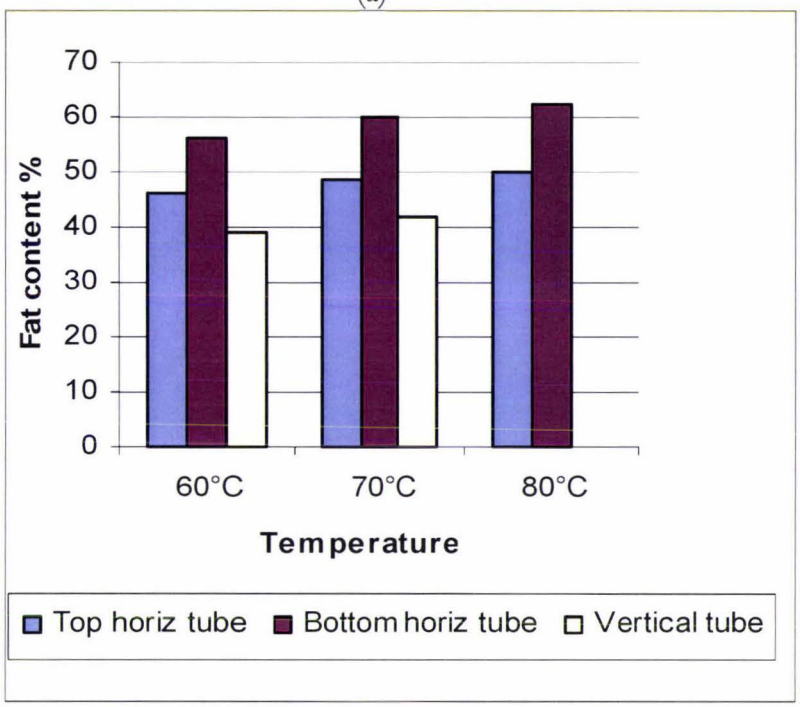
### 4.2.1 Effect of homogenisation on the composition of fouling layers

The fouling layers obtained in this study contained mainly fat (30%-62% w/w), protein (33%-60% w/w) and minerals (1%-11% w/w).

Overall these results agree with previous studies in New Zealand which, indicated that the fouling deposits from New Zealand milk seem to contain much more fat (more than 45%) compared to that reported overseas (4-8%). (Truong *et al*, 1996, Fung, 1998; Ma et al.,1999;) The reason for this difference is still being studied.

In both horizontal and vertical tubes, and at all three temperatures studied in this work, fouling layers from un-homogenised milk had more fat than fouling layers from homogenised milk, as shown in figure 4.1(a and b).

(a)




(b)

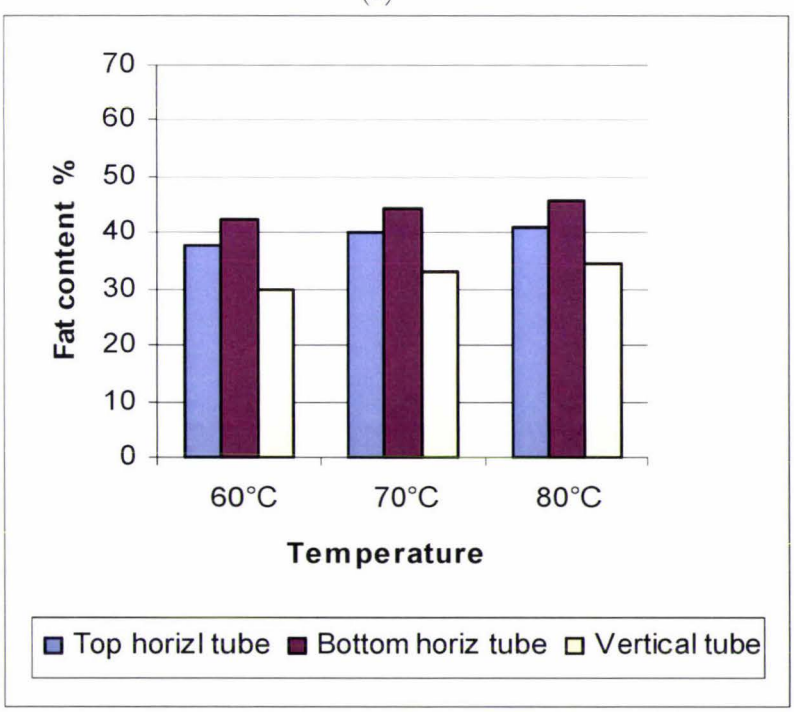
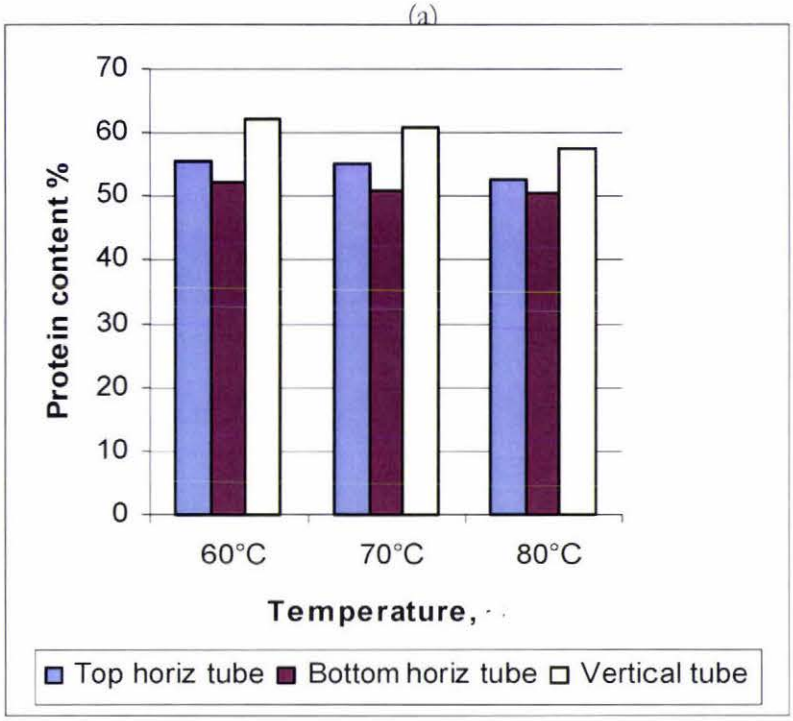



**Figure 4.1 Fat contents of fouling layers from un-homogenised milk (a) and homogenised milk (b)**

As a consequence of lower fat contents in fouling layers from homogenised milk, they had proportionally more protein than fouling layers from un-homogenised milk as shown in figure 4.2 (a and b).

(a)




(b)

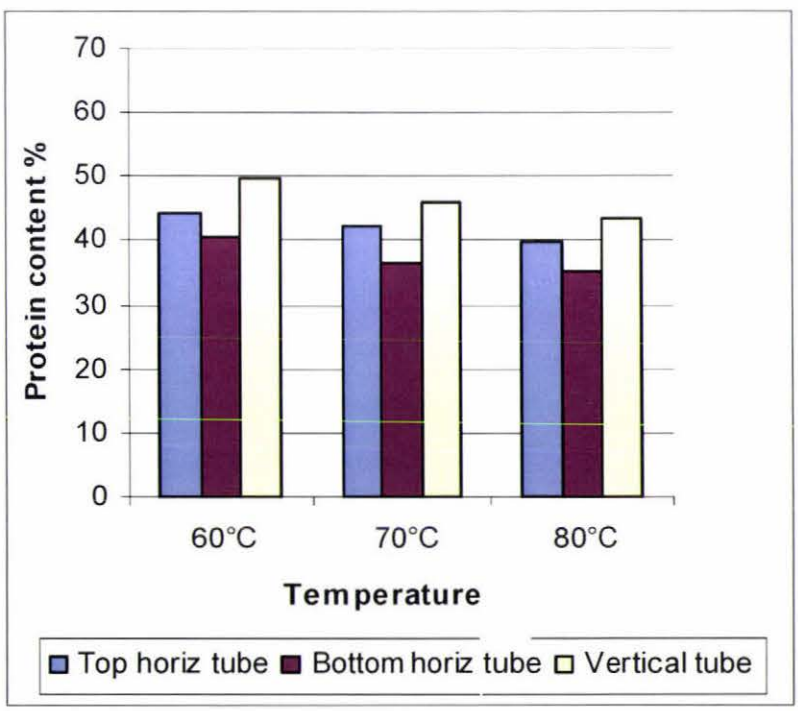
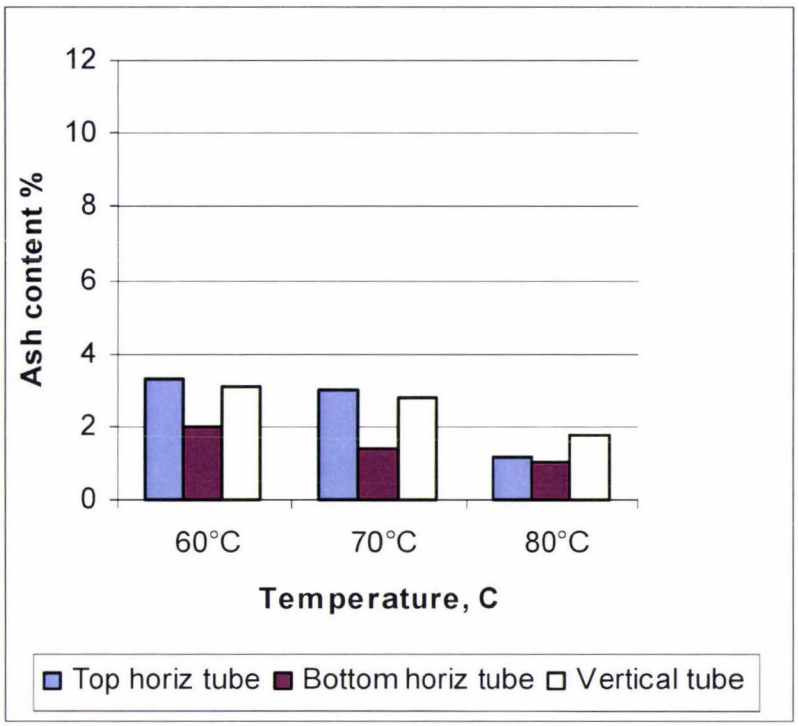



**Figure 4.2 Protein contents of fouling layers from homogenised milk (a) and un-homogenised milk (b)**

At all three temperatures and in both, horizontal and vertical tubes, fouling layers from un-homogenised milk had higher ash content than fouling layers from homogenised milk, as shown in figure 4.3 (a and b).

(a)




(b)

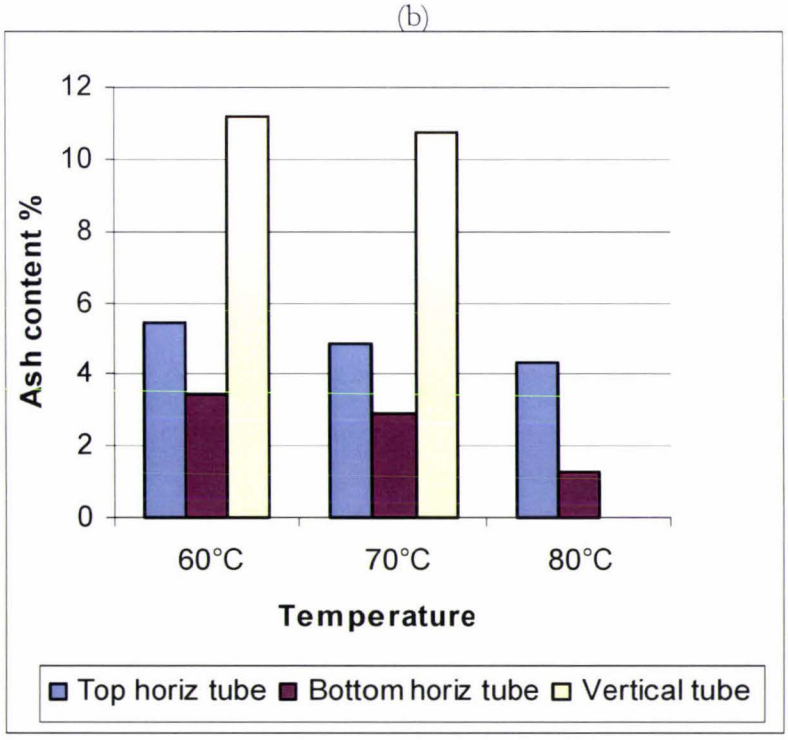



**Figure 4.3 Ash contents of fouling layers from homogenised milk (a) and un-homogenised milk (b)**

### 4.2.2 Effect of homogenisation on the fouling rate

The fouling rate was measured in two ways: By weighing the total amount of fouling at the end of each run, which was always kept at 4 hours duration, and by following the rate of change of the heat transfer coefficient during the run.

#### *4.2.2.1 Weight of fouling*

The weights of dried fouling deposits collected at the end of the runs are summarised in table 4.1.

**Table 4.1 Dry weight of fouling deposits from homogenised and un-homogenised milk**

| Milk temperature C° | Tube | Fouling weight (homog. Milk) g | Fouling weight (unhom. milk) g |
|---|---|---|---|
| 60°C | Top side/horiz. | 5.52g | 7.18g |
| | Bottom side/horiz. | 9.03g | 10.73g |
| | Vertical | 17.81g | 20.02g |
| 70°C | Top side/horiz. | 2.35g | 0.92g |
| | Bottom side/horiz | 5.0g | 4.96g |
| | Vertical | 6.0g | 0.86g |
| 80°C | Top side/horiz | 1.16g | 2.27g |
| | Bottom side/horiz | 6.24g | 11.07g |
| | Vertical | 8.48g | 0.6g |

The difference between fouling deposits of homogenised and un-homogenised whole milk did not show a clear pattern. It appears that other factors, not yet identified and therefore not well controlled in the experiment, affected this weight difference, which was less than two grams in all but two runs. This difference is too small to be significant as shown by the null hypothesis analysis in appendix 9.

### *4.2.2.2 Heat transfer resistance curves*

The rate of fouling was also monitored by following the changes in heat transfer resistances during the runs. A sample calculation of the overall heat transfer coefficient (U) and the normalised heat transfer resistance is given in appendix A10.

An example of heat transfer coefficient traces obtained from the fouling experiment is shown in figure 4.4:

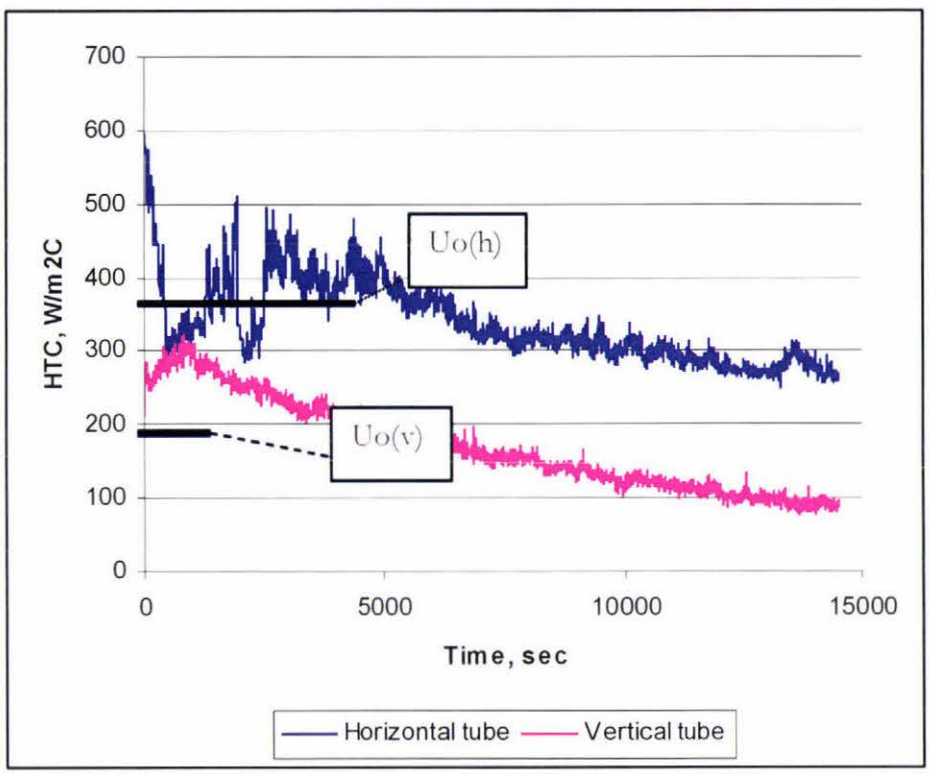



**Figure 4.4 Heat transfer coefficient (HTC) for fouling from un-homogenised milk at 60°C in both horizontal and vertical tubes Uo: Heat transfer coefficient of the clean tube at the beginning of the run.**

An example of a typical normalised heat transfer resistance curve is shown in figure 4.5. More curves can be found in appendix A12.

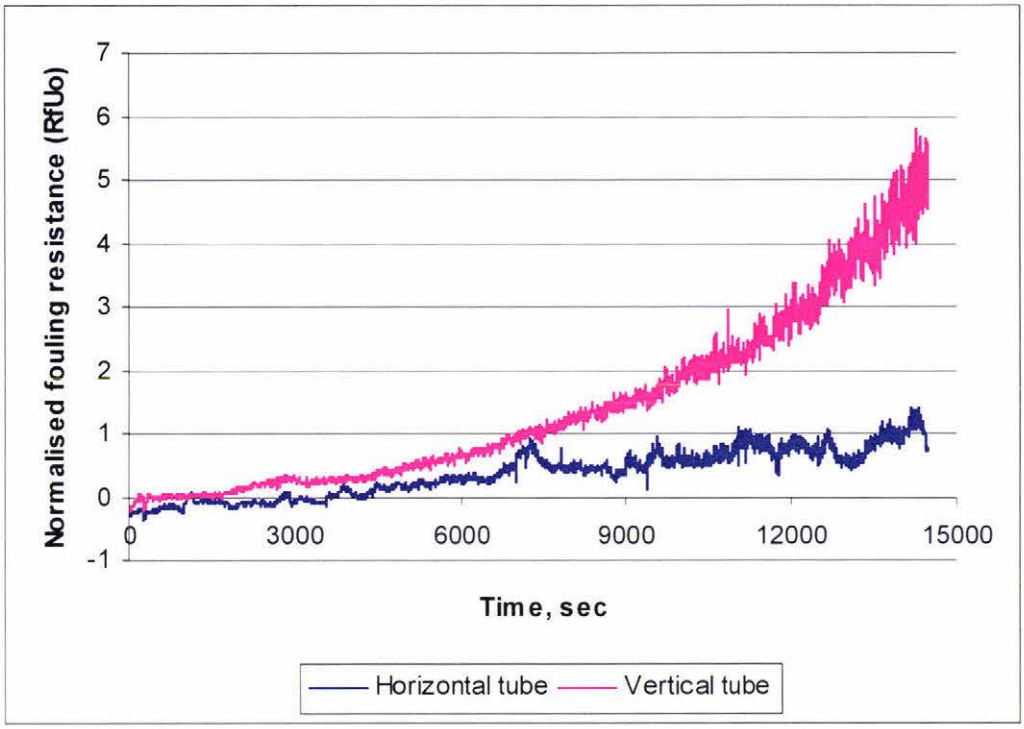



**Figure 4.5 Heat transfer resistance of fouling from un-homogenised milk. Inlet temperature: 60°C**

Following Fung (1998), the slope of the linear portion of the graph was taken as the rate of fouling. The summary of the rate of fouling obtained in this study is presented in table 4.2. Again, no clear trend can be identified.

**Table 4.2 Rates of fouling from homogenised and un-homogenised milk**

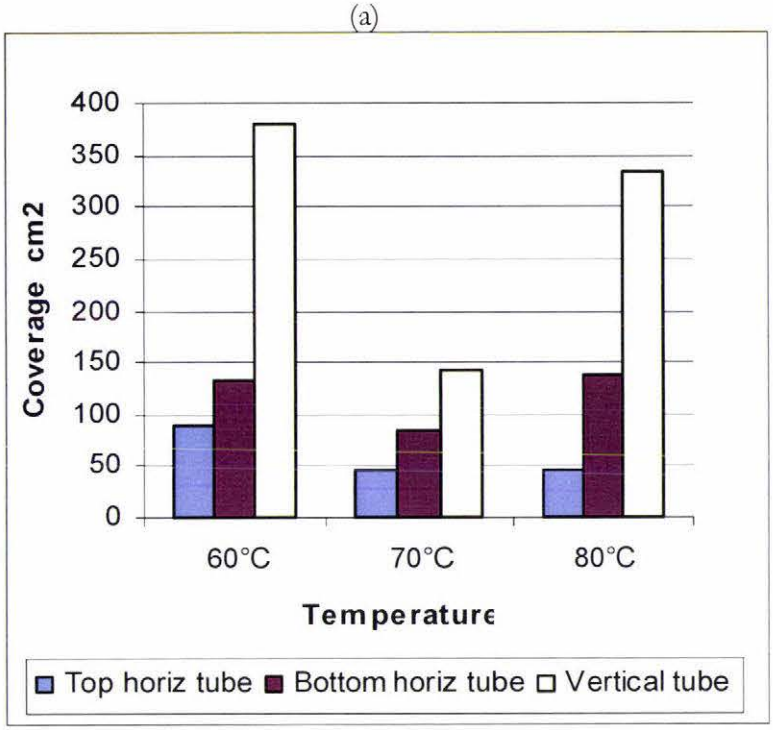
| Inlet temperature (°C) | Tube | Fouling rate Homog. milk ($s^{-1}$) | Fouling rate unhomo. milk ($s^{-1}$) |
|---|---|---|---|
| 60°C | Horizontal | 4.43E-05 | 7.96E-05 |
| | Vertical | 1.8E-04 | 4.12E-04 |
| 70°C | Horizontal | 3.8E-05 | 1.2E-05 |
| | Vertical | 1.82E-05 | 8.25E-05 |
| 80°C | Horizontal | 2.7E-05 | 7.5E-05 |
| | Vertical | 1.3E-04 | 4.0E-05 |

### *4.2.2.3 Coverage and thickness of fouling deposits*

The overall heat transfer coefficient and the final weight of deposits did not give in fact a full picture of the processes of fouling in the heat exchanger. Throughout this work it was noted that the fouling layer did not always cover the entire surface of the heat exchanger. At the beginning of this project, the lack of full coverage occurred because the air pockets, especially on the upper side of the horizontal tube that prevent contact between milk and the hot surface. This was rectified in subsequent runs by withdrawing air periodically from these pockets with a syringe. The air pockets diminished but did not disappear completely. The area covered was measured by digital image analysis using the method described in the previous section (Section 3.7.2) and the results are shown in figure 4.6 (a and b). The thickness of fouling layers which, may be a better indication than the total weight, was obtained by dividing the weight by the covered area (figure 4.7 a and b).

Figures 4.6 and 4.7 show that homogenisation did not have a conclusive effect on the coverage and thickness of fouling layers; rather these two variables seemed to be more influenced by the temperature (section 4.3) and by the orientation of heating surfaces (section 4.4).

(a)




(b)

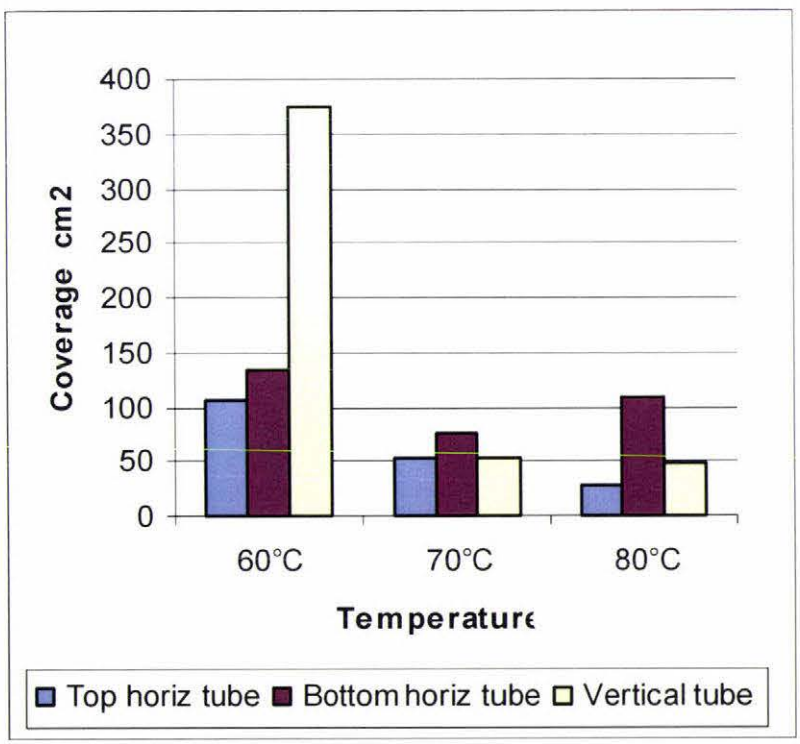
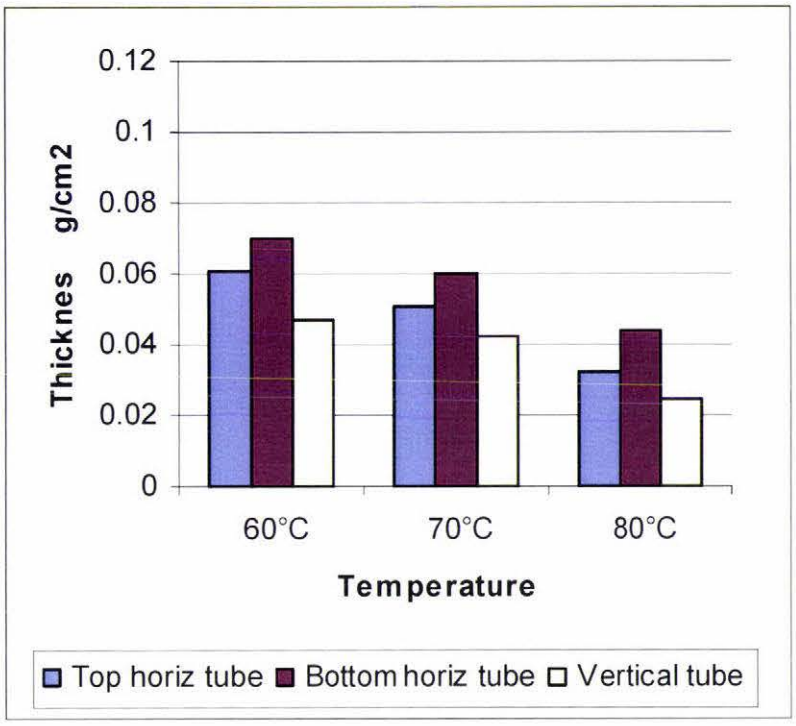



**Figure 4.6 Coverage of fouling layers from homogenised (a) and un-homogenised milk (b)**

(a)




(b)

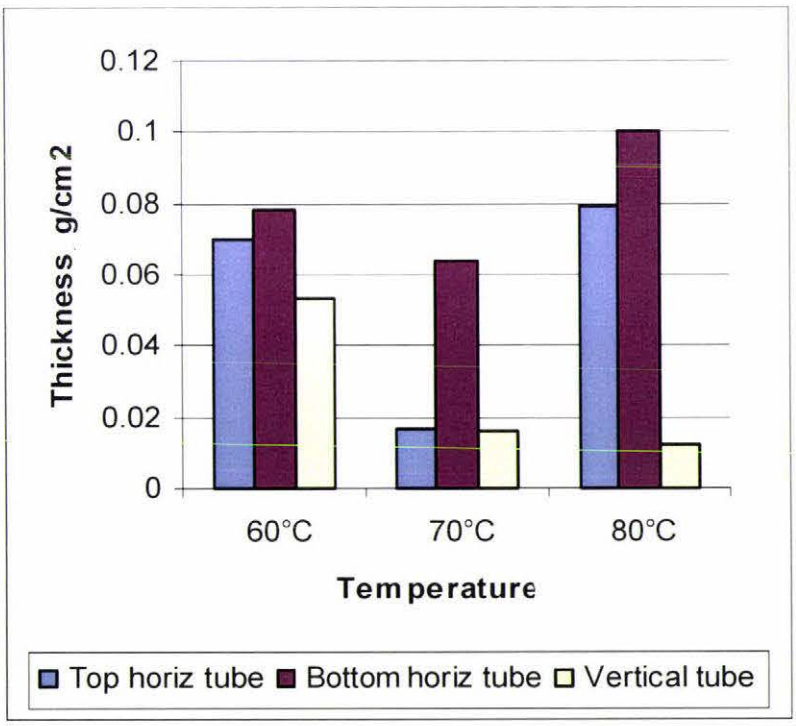



**Figure 4.7 Thickness of fouling layers from homogenised (a) and un-homogenised milk (b)**

## 4.3 Effect of temperature

### 4.3.1 Effect of temperature on the composition of fouling layers

In both horizontal and vertical tubes, fat contents of fouling layers from homogenised and un-homogenised milk tended to increase as milk temperature increased (see figure 4.1 a, b). Simultaneously, protein and ash contents of fouling layers decreased as a natural consequence of the increased fat contents (Figures 4.2 and 4.3 a, b). At higher temperature the density of fat decreases faster than the density of aqueous solutions and the fat globules become more mobile. These fat globules might migrate towards the heated surfaces more readily and, may explain the increase of fat content of the fouling layers at higher temperatures.

### 4.3.2 Effect of temperature on the resistance to deformation of fouling layers

In both, horizontal and vertical tubes, the resistance to deformation of fouling layers from homogenised and un-homogenised milk, measured by the slope of the stress- strain curves (figure 4.8), decreased with increasing milk temperature, as shown in figures 4.9 (a, b). The decreasing strength of fouling layers is a consequence of the decreasing amount of fouling with increasing milk temperature, which results in diminished thicknesses of the fouling samples tested as shown in section 1.3.3 below.

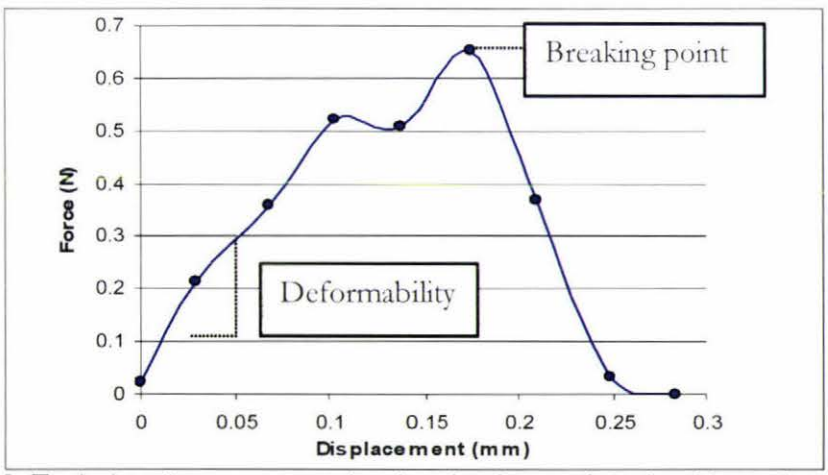
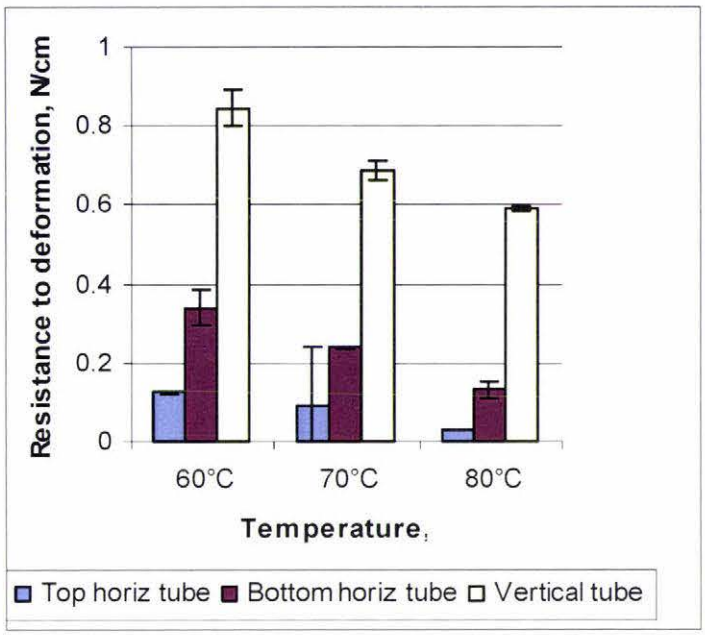



**Figure 4.8 Typical strain-stress curve showing the slope and the breaking point of a fouling sample on the top of the horizontal tube.**

The resistance to deformation of fouling layers from un-homogenised milk in the vertical tube, at 80°C could not be determined due to the little amount of deposits obtained in this run.

(a)




(b)

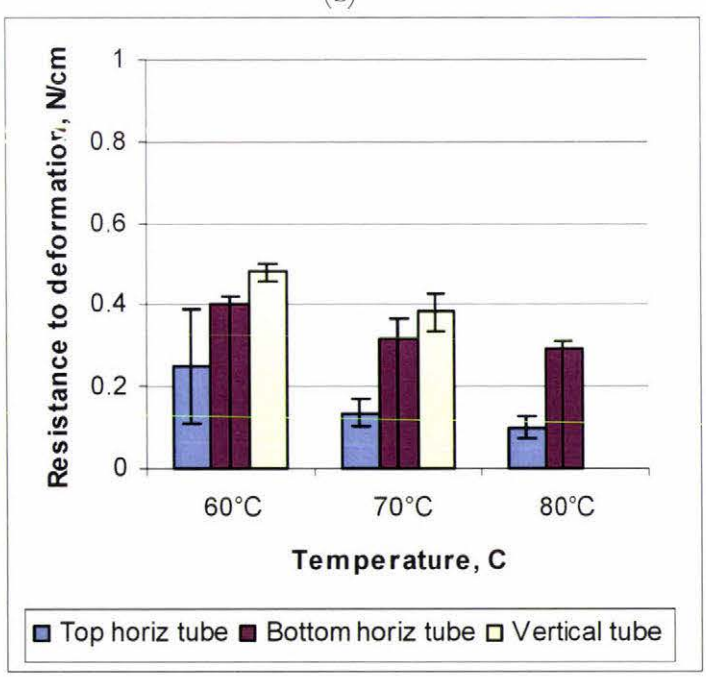



**Figure 4.9 Resistance to deformation of fouling layers from homogenised milk (a) and un-homogenised milk (b). Bars represent standard errors.**

### 4.3.3 Effect of temperature on the fouling rate

At 60°C, in both horizontal and vertical tubes, more fouling was produced by un-homogenised milk than with homogenised milk. At 70°C and 80°C it was not possible to determine which milk (homogenised or un-homogenised) fouled more due to the unclear trend showed by the final weight of fouling (table 4.1).

Fryer (1989) identified two modes of control of milk fouling: surface and bulk. If the wall temperature is very hot proteins are denatured and rendered near the surface and fouling will be a surface control process, especially if the bulk temperature is below the denaturation temperature of the whey proteins, e.g., 60°C.

If the bulk temperature is hot enough to induce denaturation and aggregation, protein denatured and aggregated in the bulk will contribute to fouling. Bulk control of fouling will occur if the concentration of activated proteins (those ready for aggregation with other proteins or fouling surfaces) in the bulk exceeds the concentration of active proteins at the wall.

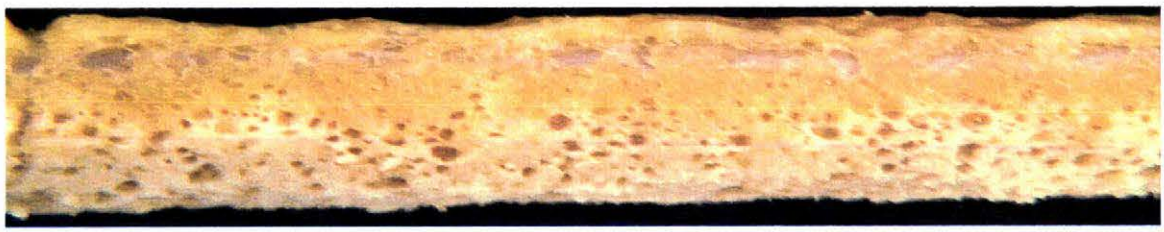
Fryer also suggested that the temperature difference between the surface and the bulk affects the fouling rate. Grandison (1996) stated that a higher differential temperature between milk and heating medium promotes deposition and reducing the temperature differential between the product and the heating medium reduces fouling. During this work, the heating medium temperature remained constant (hot water at 90°C) during all fouling trials while the bulk milk temperature was different in each experiment (60°C, 70°C and 80°C), consequently the temperature difference between milk and hot water decreased as bulk milk temperature increased, resulting in less fouling at 70°C and 80°C than at 60°C.

#### *4.3.3.1 Effect of temperature on the coverage of fouling*

Coverage of fouling from homogenised and un-homogenised milk in the vertical tube was much less at 70°C and 80°C than at 60°C as shown in figure 4.6. Photographs of fouling tubes shown in figure 4.10 a, b and c also confirmed these results.

At 60°C, coverage of fouling by both homogenised and un-homogenised milk was very similar. However, at 70°C and 80°C coverage of fouling was bigger at the bottom of horizontal tubes than at the top as shown in figure 4.6 a, b and appendix A11.

(a)



(b)

(c)

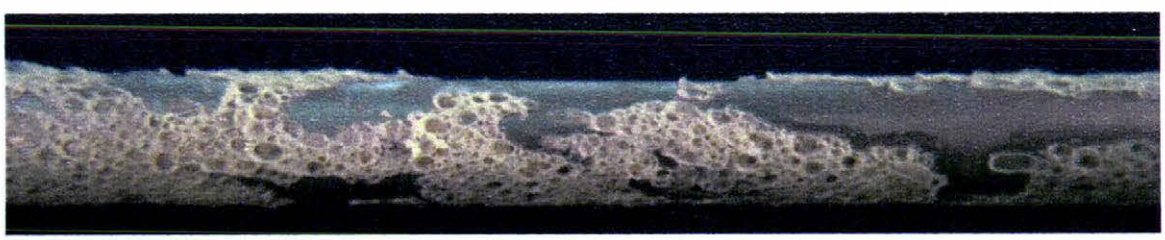


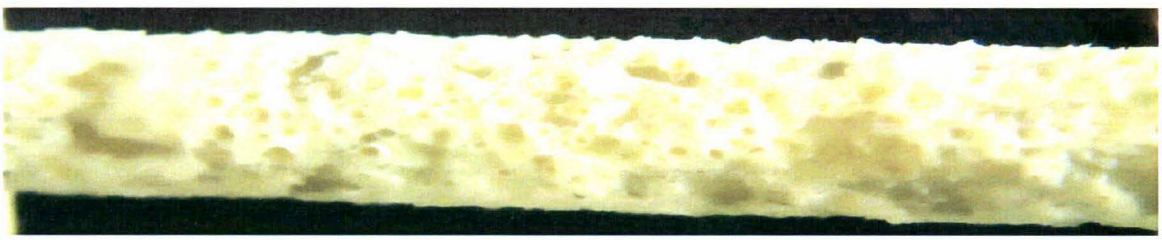
**Figure 4.10 Fouling layers from homogenised milk in the vertical tubes formed at 60°C (a) at 70°C (b) and at 80°C (c), showing decreased coverage with increasing temperature for a constant hot water inlet temperature of 90°C.**

### *4.3.3.2 Effect of temperature on thickness of fouling layers*

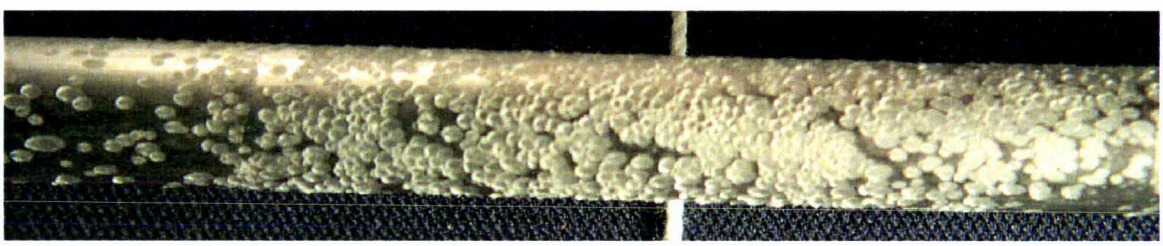
As milk temperature increased, the thickness ($g/cm^2$) of fouling layers from homogenised milk in both horizontal and vertical tubes, tended to decrease. But fouling layers from un-

homogenised milk did not show a clear trend on the horizontal tubes (Figure 4.7 a and b), probably because the tubes were not totally covered by fouling. This is not the case for fouling from un-homogenised milk in the vertical tubes, where the thickness trend was similar to that of fouling from homogenised milk. This is shown in figure 4.7, a and b and figure 4.11 a, b and c.

(a)



(b)



(c)

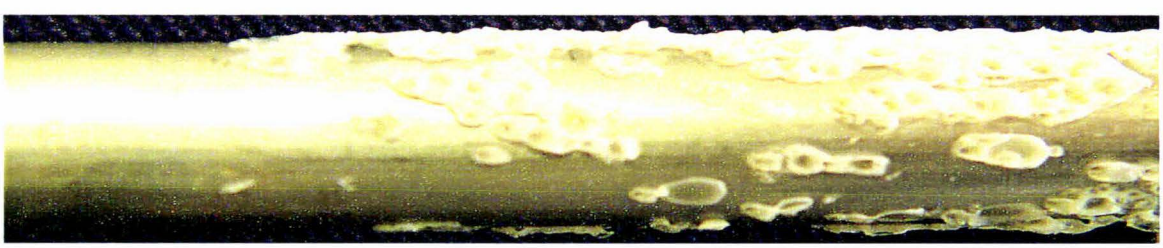


**Figure 4.11 Fouling layers from un-homogenised milk in the vertical tubes formed at 60°C (a), 70°C (b) and 80°C (c), showing decreased coverage with increasing temperature for a constant hot water inlet temperature of 90°C.**

Since the weight of fouling decreased as the temperature increased, this trend was also reflected in the changes in area covered by the fouling deposits and in their thickness. The greater thickness found on the underside (bottom) of the horizontal tube is probably due to the creaming effect discussed in the next section.

## 4.4 Effect of orientation of heating surfaces on fouling

### 4.4.1 Effect of orientation of heating surfaces on the composition of fouling layers

Fouling layers from homogenised and un-homogenised milk in the bottom had more fat than fouling layers on the top of horizontal tube. Consequently, proteins and ash contents of fouling deposits on the bottom were lower than those found on the top of horizontal tube (figure 4.1, a, b).

At all three temperatures fat contents of fouling layers from homogenised and un-homogenised milk in horizontal tube were higher than those of fouling layers in vertical tube (Figure 4.1 a, b). At the same time, protein and ash contents of fouling deposits were higher in vertical tube than in the horizontal tube as shown in figures 4.2 (a, b) and 4.3 (a, b).

These results suggested that the amount of fat attached to the wall depended of the orientation of the heating surface, explained as follows:

#### *4.4.1.1 Fat migration mechanism in the horizontal tube*

Creaming of milk fat globules (separation of fat from the aqueous phase) is due to the difference in densities between the fat and the plasma phases of milk (Walstra and Jenness, 1984). Homogenisation of milk causes disruption of fat globules to prevent creaming (Section 2.4).

Migration of milk fat globules to the wall has also been demonstrated by Yamamoto et al., (1986), who performed experiments of milk flow in capillary tubes at low flow rates. Ma and Trinh (1999) suggested that fat globules tended to migrate towards and adhere to the stainless steel wall because of its hydrophobic attraction, which in turn promoted fouling. Fung (1998), also made similar observations when working with different levels of milk fat globule membrane damage and its effect on fouling.

Large fat globules (e.g. un-homogenised/native milk fat globules) migrate towards and attach to the wall easier than smaller fat globules (e.g. homogenised milk). In addition, when milk was heated in a horizontal tube, the fat globules movement towards the bottom of the inner tube was increased by a creaming force resulting in higher fat content in fouling layers at that position. The top side of the inner tube was somewhat shielded from the creaming effect by the presence of the tube itself and the creaming effect actually worked in a opposite direction to the hydrophobic attraction as shown in figure 4.12. The results obtained in this research confirmed this theory.

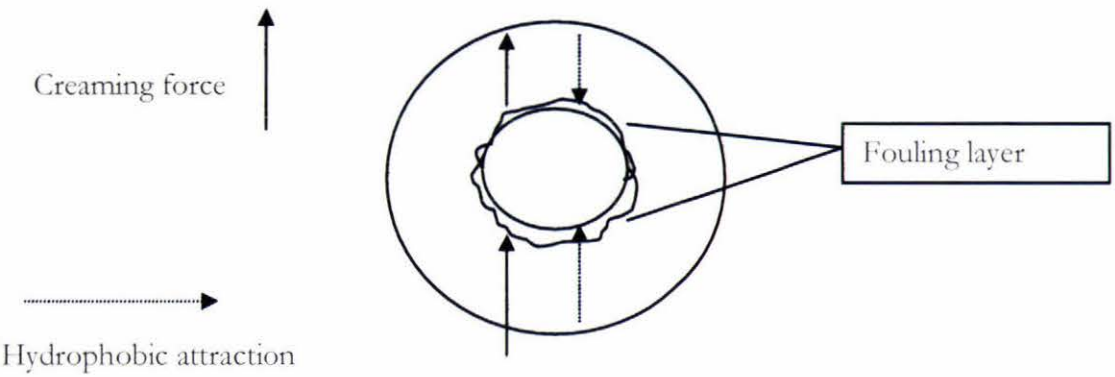



**Figure 4.12 Mechanism of fat migration in a horizontal tubular heat exchanger**

### *4.4.1.2 Hypothesis of fat migration mechanism in the vertical tube*

In the vertical tubes, the creaming effect tended to push the fat globules to move faster in the direction of the milk flow. This does not contribute to the movement of fat globules towards the wall that occurs along a horizontal plane. If anything, the amount of time available for the transport of the fat globules towards the heated wall by hydrophobic attraction is decreased by the acceleration of vertical flow of fat globules parallel to the heated surface by creaming forces. Hence, the fat content of fouling layers on vertical tubes is smaller than that on the bottom side of the horizontal tubes.

As Reynolds Number increases, the relative importance of fat diffusion towards the wall compared to convection along the wall decreases and, the difference in composition between fouling layers from homogenised and un-homogenised milk is expected to decrease.

However, even at the high Reynolds numbers encountered in the industry, there are pockets of slow flow. These so-called recirculation regions occur near disturbances to the main stream introduced, for example, by sudden pipe expansions (Truong *et al.*, 1996) and/or by abrupt changes in flow direction, and involve even stagnation points where the linear velocity of the milk is zero (Schlichting, 1960). These recirculation zones, where fouling is more serious, will exhibit differences between fouling layers of homogenised and un-homogenised milk similar to those found in this study.

It should also be noted that incorporation of fat into the fouling layer takes place through the intermediary of the proteins (whey and caseins) that coat of fat globules when the native MFGM is disrupted, as happens in homogenisation. This protein coating can become more reactive upon heating and initiate the incorporation of fat into the fouling layer. The "activation" of the fat globules is thus an issue of protein behaviour during heating, and as such is not dependent on the Reynolds.

### 4.4.2 Effect of orientation of heating surfaces on the strength of fouling layers

In vertical tubes, at all three temperatures, fouling layers from homogenised milk had more resistance to deformation than fouling layers from un-homogenised milk (Figure 4.9 a, b). These results confirmed the findings of previous study (Ma *et al.*, 1999) which showed that rigidity of fouling layers was reflected by their protein content; as more protein was present in fouling deposits, the resistance to deformation of fouling layers increased.

Fouling layers on the top of the horizontal tubes, had less resistance to deformation than fouling layers in the bottom of horizontal tubes as shown in figure 4.9 a, b.

This is in disagreement with Ma *et al.* (1999) who found that fouling layers on the top of horizontal tubes had more resistance to deformation than fouling layers on the bottom because of their increased protein content.

The fouling layers obtained in this work were not uniform as shown, for example, in figures 4.13. It is therefore quite difficult to ensure that samples used for textural strength tests are fully representative of layers over the entire surface. This may explain the difference with Ma et al's result where the fouling layers were more uniform.

**Figure 4.13 Example of fouling layers on the top of a horizontal tube. Squares show different thicknesses along the surface. Fouling from homogenised milk at 60°C**

### 4.4.3 Effect of orientation of heated surfaces on the fouling rate

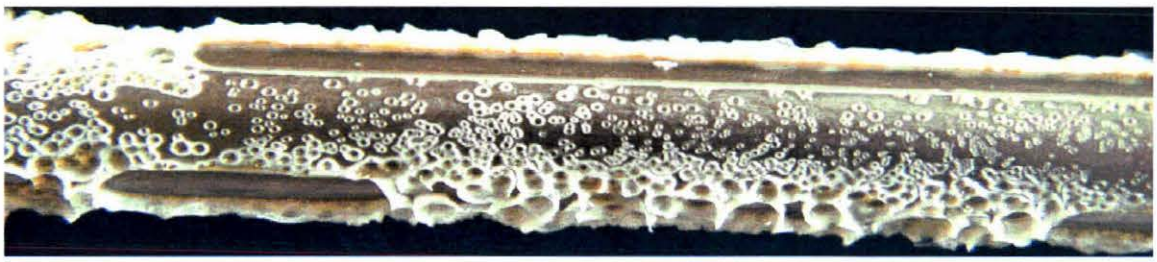
In horizontal tubes, at all three temperatures, the amount of fouling from both, homogenised and un-homogenised milk, appeared to be larger on the bottom side than on the top side of the pipe (table 4.2). As discussed in section 4.4.1, the creaming process aided the migration of fat globules, mainly, to the bottom of the pipe, increasing the amount of fouling at this point.

At all three temperatures, the amount of fouling obtained from both, homogenised and un-homogenised milk did not show a clear trend in none of the tubes (table 4.2).

#### *4.4.3.1 Effect of orientation of heated surfaces on coverage of fouling*

At all three temperatures, the top-side along horizontal tube was less covered by fouling than the bottom side for both homogenised and un-homogenised milk. An example of this observation is shown in figure 4.14. On the other hand, coverage of fouling layers from homogenised and un-homogenised milk in the vertical tube tended to be more uniform (section 4.3.3). More photographs of coverage of fouling at different temperatures can be found in appendix A11.

(a)



(b)

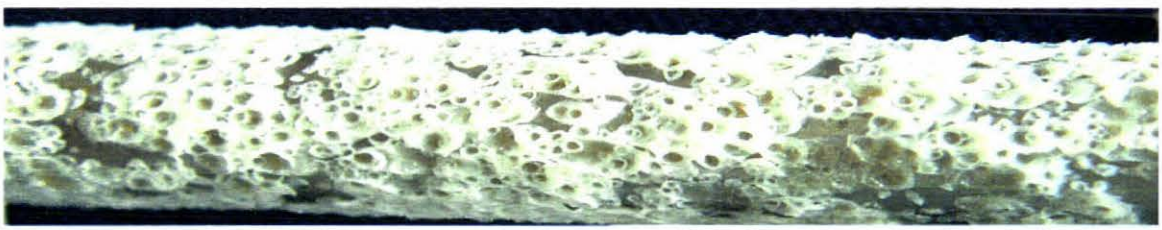


**Figure 4.14 Fouling from homogenised milk in the horizontal tube at 60C, showing partial coverage on the topside (a) and total coverage on the bottom side (b)**

### *4.4.3.2 Effect of orientation of heating surfaces on thickness of fouling layers*

At all three temperatures, fouling layers from homogenised and un-homogenised milk on the bottom of horizontal tubes were thicker than fouling layers on the top, as shown in figure 4.7 (a and b). This is also confirmed by the fouling photographs in appendix A11.

At all three temperatures and with both homogenised and un-homogenised milk, fouling layers on vertical tube were thinner than those in horizontal tube (top and bottom) as shown in figure 4.7 (a and b).

# *Chapter 5 Conclusions and Recommendations*

## 5.1 Conclusions

The results of this research showed that:

- Homogenisation strongly affected the composition of fouling layers. The fouling deposits from un-homogenised milk were found to have higher fat contents than deposits from homogenised milk at all three milk inlet temperatures. The highest protein contents were found in fouling layers from homogenised milk.

- Homogenisation did not affect the fouling rate in a clear trend within the conditions used in the present experiments.

- Fouling was heaviest when the milk was heated to 60°C in the heat exchanger with hot water at 90°C, in this setting, fouling is directed by the surface control mode. In all fouling cases except one of the 18 data points, fouling at 70°C and 80°C were substantially lower. The effect of homogenisation on the fouling rate was unclear possibly because of the different temperature difference between the hot and cold side, which had a different effect on homogenised and un-homogenised milk.

- The resistance to deformation of fouling layers was related to their thickness, which was affected by the temperature and the orientation of heating surfaces.

- Coverage of fouling was more strongly affected by the orientation of the heated surface than by homogenisation. Coverage was more uniform in the vertical tubes than in the horizontal tubes. The topside of the horizontal tubes was less covered than the bottom side. As a consequence thickness of fouling layers was more influenced by the orientation of the heated surface than by homogenisation. Fouling layers were thicker at the bottom than at the top of the horizontal tubes.

In summary, while we have not detected a definite relationship of fouling with homogenisation because of insufficient control in the runs, it is clear that the fat content of fouling layers from un-homogenised milk was much higher than that of fouling from homogenised milk and that the strength of the fouling layer is affected by the thickness of the deposit, which decreases with the milk inlet temperature.

The results imply that a different CIP protocol should be formulated for removing fouling deposits from homogenised and un-homogenised milk due to the different strengths and compositions which may affect the effectiveness of CIP.

## 5.2 Recommendations for further work

- Further experiments should be conducted with $\Delta T$ as another added controlled variable and it may be necessary to change the heating medium to reach temperature above 100°C.

- To ensure complete removal of air from the system, it may need to be redesigned to avoid air pockets.

- Study CIP effectiveness of standard formulations for fouling layers for different fat contents and devise more optimal formulations.

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# *Appendices*

## *Appendix A1 Calculation of the amount of skim milk and cream used to prepare un-homogenised milk*

The amounts of skim milk and cream were calculated based on their fat and protein contents, using the following equations:

$S+C=200$

$Sf_1+Sf_{2=}200f_3$

$Sp_1+Sp_2=200p_3$

Where,

S Amount of skim milk in litres

C Amount of cream in litres

$F_1$ Actual fat content of skim milk in ratios

$F_2$ Actual fat content of cream in ratios

$F_3$ Target fat content in ratios (0.033)

$P_1$ Actual protein content of skim milk

$P_2$ Actual protein content of cream

$P_3$ Target protein content in ratios (0.034)

The average fat and protein contents of skim milk and cream used for all trials is summarised in table below:

**Average fat and protein contents of skim milk and cream**

| Type of milk | Fat content % | Protein content % |
|---|---|---|
| Skim milk | 0.096 | 3.6 |
| Cream | 40.06 | 1.96 |

After un-homogenised milk was prepared, it was stored at 4°C prior to experiments.

## *Appendix A2 Thermocouples calibration procedure*

The thermocouples were calibrated with ice-water and boiling water, following the method developed by Bennet (2000). Thermocouples were placed in a flask containing ice-water slurry. The temperature was recorded every second for a period of 5 minutes. The data obtained were averaged and this value was used as the temperature reading of the probes for the ice point (0°C). The thermocouples were then immersed in boiling water, which remained boiling throughout the calibration exercise. The process of data collection and analysis was reported resulting in the probe readings for boiling point (100°C). A linear regression of the two reported temperatures was carried out for each thermocouple that provided a calibrated value for each thermocouple:

$\phi_c = a\phi_{cj} + b$

Where,

$a = 100/\phi_{100} - \phi_0$.

$b = a\phi_0$.

$\phi_c$, Calibrated temperature.

$\phi_{cj}$, Raw measured temperature.

$\phi_{100}$, Raw measured temperature in boiling water.

$\phi_0$, Raw measured temperature in ice-water.

# *Appendix A3 Moisture analysis*

## Air-oven Method

### Apparatus

- Aluminium moisture dishes, diameter 55mm, height 40 mm, provided with well fitting slipover covers. Previously dry at $108 \pm 5\,^{\circ}C$ and keep in a dessicator at room temperature.
- Desiccator must be airtight, containing dry silica gel or phosphorus pentoxide.
- Air oven capable of being accurately maintained at 100-130°C and provided with openings for ventilation. A thermometer should pass into the oven in such a way that the tip of the bulb is level with the top with the moisture dishes and it is not directly exposed in currents of escaping water vapour.

Note: Always use metal tongs when handling metal dishes and lids. Never use hands, as moisture and oils from hands can cause a significant error in the measurements.

### Procedure (AOAC, 1999)

- Accurately weight a dry, cooled aluminium moisture dish and lid. Add approximately 2g of sample to the dish, replace the lid and quickly re-weight.

- Place the dish, lid and contents in the air oven at the prescribed temperature for three hours. Remove the lid and place it underneath the dish to avoid confusion later. Before removing from the oven, cover the dish with the lid and transfer rapidly to the desiccator.

- Once cool, weight accurately.

- Return to oven and repeat steps b and c. Repeat until the weight in step c is constant.

**Calculation**

$$\% \text{ total moisture and volatile matter} = \frac{(A-B)x100}{A}$$

$$\% \text{ total solids} = \frac{B}{A}x100$$

Where:

A= original weight of sample (g)

B= final weight dry sample (g)

# *Appendix A4 Protein analysis*

## Kjeldahl Method

### Apparatus

- Kjeltec 1026 system (Tecator Sweden)
- Kjeldahl tubes
- Distillation unit
- Digestion unit
- Scale for weighing
- 250 ml conical flask

### Reagents

- Concentrated $H_2SO_4$
- Kjeltabs
- 4% boric acid solution
- 0.1 M HCl

### Procedure (AOA)

### Digestion

- Accurately weigh about 0.5 g sample into the digestion tube.

- Add two kjeltabs (each containing 3.5g $K_2SO_4$ and 0.0035g Se) and then 15 ml concentrated $H_2SO_4$.

- Carry out a blank digestion at the same time (no sample but all the other reagents). Set up block digestor unit and digest samples at 420°C for 40 minutes or until clear.

- Remove the tubes carefully from the heating unit, leaving the exhaust manifold in place and water aspirator about half on. Allow cooling until the tops of the tubes are cool to touch.

- Add approximately 70 ml hot distilled water to each tube and shake gentle to mix. Ensure all solids have been dissolved.

**Distillation and titration**

- Add 25 ml 4% boric acid solution to 250 ml conical flask.

- Set up the distilling unit.

- Connect the digestion tube with the first sample to be distilled in position.

- Place the receiver flask and boric acid solution on the platform and raise to its upper position. To avoid contamination do not touch the glass outlet tube with your fingers. Hold it by its plastic tubing.

- Close the safety door. The distillation automatically starts.

- When distillation is complete, the machine will beep several times. Remove the digestion tube and the receiver flask.

- Titrate the sample with 0.1 M HCl to grey-mauve end point.

**Calculation**

$$\%N = \frac{(AxB)x14x100}{1000xC}$$

Protein= %N x 6.38

Where:

A= mls HCl used

B= exact molarity (normality) of HCl

C= weight of original sample taken (g)

# *Appendix A5 Total fat content*

## Monjonnier Method for milk fat content (AOAC, 1999)

### Apparatus

- Water bath
- Flaks and boiling chips
- Mojonnier fat extraction tubes
- Steam bath
- Drying oven
- Centrifuge, Monjonnier type
- Rubber bungs (stoppers)
- Scale

### Reagents

- Ethyl alcohol (95% )
- Diethyl ether, free from residue on evaporation
- Petroleum ether, boiling point below 60°C
- 35% w/w ammonium hydroxide
- 2% phenolphthalein

### Procedure

- Weigh accurately sufficient sample to give between 0.3-0.7g of extracted fat into a dry Mojonnier tube (e.g. 10ml of milk; 1-2g of milk powder). Make up to 10ml if necessary with water, and shake to dissolve or blend.

- Add 2ml ammonium hydroxide and mix well in the lower bulb. Place in 60°C water bath for about 5 minutes and swirl occasionally. Cool. Add 2-4 drops of phenolphthalein

- Add 10 ml of ethyl alcohol and mix by allowing the liquid to flow backwards and forwards between the two bulbs; avoid bringing the liquid too near the neck of the tube. The complete extraction of the fat is dependent on satisfactory mixing at each stage.

- Add 25 ml of diethyl ether, close the tube with the stopper and shake gently for about a minute.

- Remove the stopper and add 25ml petroleum ether, using the first few ml to rinse the stopper and the neck of the tube, allowing the rinsing to run into the tube.

- Replace the stopper, again wetted with water, and rock carefully for 30 seconds.

- Centrifuge Mojonnier flask for 2 minutes at 600 RPM.

- Examine the tube to see if the interface of the liquid is in line with the upper junction of the neck of the tube. If it is below, it should be raised by the addition of distilled water down the side of the tube.

- Remove the cork and carefully decant as much as possible of the organic solvent layer into a pre-weighed short-necked flask by gradually bringing the cylindrical bulb of the tube into a horizontal position.

- Add 5 ml of ethyl alcohol and mix. This helps to prevent emulsions forming and is in accord with the AOAC (1999).

- Repeat the extraction using 15 ml of diethyl ether and 15 ml of petroleum ether (step d to I). Add second extract into the same flask as used in the previous step.

- Distil carefully the solvents from the flask and dry the flask in the oven at 100°C for 90 minutes, taking precautions to remove all traces of solvent vapour, prior to placing in the oven.
- Allow the flask to cool at room temperature. Do not use a desiccator.

- Weigh the flask and record the fat content of the sample.

- At the same time as the above procedure is carried out, make a blank determination with 10ml of water instead of sample. Use a similar extraction apparatus, the same reagents and the same technique throughout. Correct the apparent weight of fat for the change, if any, in the weight of the flask used for the blank determination.

**Calculation**

Crude fat content is the final weight of residue remaining in the flask, expressed as a percentage of the weight of the original sample.

$$\%\text{Fat} = \frac{w2 - w1}{w3} x100$$

Where:
w1= weight of empty flask (g)
w2= weight of flask +fat (g)
w3= weight of sample taken (g)

## *Appendix A6 Ash analysis*

### Dry Ashing Method

The easiest and most direct method for determining ash is by combustion of a weighted sample in a muffle furnace. This method is widely used by the dairy industry.

### Apparatus

- Silica or platinum dish (or crucible)
- Forceps
- Bunsen burner
- Muffle furnace
- Dessicator
- Scale for weighing

### Procedure (AOAC, 1999)

- Heat a silica or platinum dish (or crucible) for 60 min in the muffle furnace at 525-550°C. Cool in a dessicator for at least 60 minutes.

- Using forceps, remove a cooled crucible and accurately weigh.

- Accurately weight about 1-2 g of sample into a crucible spreading the sample uniformly in the crucible before weighing.

- Char over a Bunsen burner, taking care that sample does not ignite.

- Place the dish in muffle furnace, cool thoroughly and weigh.

Note: Ash content is the final weight of residue remaining in the crucible.

**Calculation of % ash**

$$\% \text{ ash (dry basis)} = \frac{w2 - w1}{w3} x100$$

Where:

w1= tare weight of the crucible (g)

w2= weight after ashing (g); w3= original sample weight (g).

# *Appendix A7 Texture analysis*

## TA XT2 Texture Analyser

### Apparatus

- Texture analyser machine
- Computer
- Probes (TA43 knife blade with flat 3mm end)

### Procedure

- Turn on the TA-XT2 analyser using the switch at the rear of the instrument.
- Switch on the computer, start windows and start the texture expert program in the texture expert 2 group.
- Scroll down to LAB user name and click ok. If a project starts up from the last time the program was used, left click at the icon and select " close".
- Create a new project. Select "file new project". Click restart.
- Set up and save graphs preferences. Select "File-preferences-graph.
- Set up and save the TA-XT2 settings. Select "T.A settings".
- Click save and name the file.
- Calibrate the probe before any testing is carried out, by selecting "T.A calibrate probe" from menu.
- Place sample on the base.

- Run the test by selecting " T.A: run a test"

- After the test, select "go to" menu and set the measurements to perform on the graph.

- To save, select "File-save as" and enter the file name.

## *Appendix A8 Fouling coverage analysis by Sigma Scan Pro*

### Apparatus

- Computer
- Photographs of fouled tubes

### Procedure

- Turn the computer on and open the Sigma Scan pro program. Select "open image" from the file menu.

- Select " calibrate image, distance and area" from the image menu. This will bring up a dialog box with three alternatives of calibration. Select two-point rescaling calibration. The cursor will show a letter "C" associated to it and click one side of the image rectangle.

- Move the pointer to the opposite side and click a second time. (This will tell the program the number of pixels in a straight line from one side of the image to the other.

- Click ok. This is the "old distance" in pixels. The new distance is known since it is possible to have the dimensions of the image in millimetres; enter this value into the "new distance" box.

- Finally enter the words "mm" and "sq mm in the two boxes at the bottom of the dialog box and click ok.

- Select "measurements settings" from the measures menu and tick the measurements you want to perform. From the same dialog box find source overlay and select one colour. (When you make measurements overlays will deposit a coloured area on the image); select "measure objects" from the measurements menu. It will appear a red area on the image and a data worksheet; save the data as an excel file.

## *Appendix A9 Null hypothesis analysis for the weight of fouling*

$D=X_1-X_2$ (difference fouling weights; $X_1$ and $X_2$ are the weight of fouling from homogenised and un-homogenised milk respectively).

$d=\Sigma (X_1-X_2)/n = 0.33$ (sample mean of the difference fouling weights)

$S_D= \sqrt{\sum (D-\bar{d})/(n-1)}$ =3.95 (sample standard deviation)

df= n-1= 8 (degrees of freedom)

$s_d= SD/\sqrt{n} = 1.316$ (standard error)

$\alpha= 0.05$ level of significance

$\mu_{Dhyp}= 0$ (hypothesized mean for all difference weights in the population)

n= 9 (sample population)

**Research problem:**

Is the weight of fouling from homogenised milk different to that of un-homogenised milk?

**Statistical hypothesis:**

Ho: $\mu_D=0$ (Null hypothesis)

$H_1$: $\mu_D \neq 0$ (Alternate hypothesis)

**Decision rule:**

Reject Ho at 0.05 level of significance if $t_c \pm 2.306$ (from $t_c$ distribution table, given that df=8)*.

**Calculations:**

The null hypothesis was tested with the following ratio:

$$t=\frac{d-\mu Dhyp}{sd} = 0.25$$

**Decision/Interpretation:**

Retain Ho/ The weight difference between fouling from homogenised milk and un-homogenised milk is not statistically significant; there is not evidence that show homogenisation affects the weight of fouling.

***Critical values of t ■(Witte and Witte, 1997)**

| df | $t_c$ at $\alpha$=0.05 | df | $t_c$ at $\alpha$=0.05 |
|---|---|---|---|
| 1 | 12.706 | 18 | 2.101 |
| 2 | 4.303 | 19 | 2.093 |
| 3 | 3.182 | 20 | 2.086 |
| 4 | 2.776 | 21 | 2.080 |
| 5 | 2.571 | 22 | 2.074 |
| 6 | 2.447 | 23 | 2.069 |
| 7 | 2.365 | 24 | 2.064 |
| 8 | 2.306 | 25 | 2.060 |
| 9 | 2.262 | 26 | 2.056 |
| 10 | 2.228 | 27 | 2.052 |
| 11 | 2.201 | 28 | 20.48 |
| 12 | 2.179 | 29 | 2.045 |
| 13 | 2.160 | 30 | 2.042 |
| 14 | 2.145 | 40 | 2.021 |
| 15 | 2.131 | 60 | 2.000 |
| 16 | 2.120 | 120 | 1.980 |
| 17 | 2.110 | $\infty$ | 1.960 |

■Two-tailored or non-directional test

## *Appendix A10 Calculation of the overall Heat Transfer Coefficient and Normalised Heat Transfer Resistance*

***Sample calculation:***

For un-homogenised milk, inlet temperature: 80°C (horizontal tube)

$Q'_{milk}$= 44.8 L/hr

$t_m$= 79.11°C (milk inlet temperature)

$T_m$= 84.27°C (milk outlet temperature)

$t_w$= 91.32°C (water inlet temperature)

$T_w$= 90.48°C (water outlet temperature)

$\rho_{milk}$= 1.035 Kg/$m^3$

$\mu_{milk}$= 0.5Cp

$Cp_{milk}$ (80°C)= 3.930 KJ/Kg°C

$d_e = D_i - d_o$= 0.0095m

$S' = \pi/4 (D_i^2 - d_o^2)$= 2.86 x $10^{-4}$ $m^2$ (4.1)

V' = Q' /(1000x 3600S') = 0.175m/s

$Re = \rho_{milk} \times V' \times d_e / \mu_{milk}$ = 984 (4.2)

$\Delta T_1 = t_w - T_m$= 7.05°C (4.3)

$\Delta T_2 = T_w - t_m$= 11.37°C (4.4)

$\Delta T_m = \Delta T_1 - \Delta T_2 / \ln (\Delta T_1/\Delta T_2)$= 9.03°C (4.5)

$Q_m = m^* Cp_{milk} * (t_m - T_m)$= 256.53 W (4.6)

$A = \pi d_o L$= 0.068$m^2$ (4.7)

$Q_m = Q_w = UA\Delta T_m$ (4.8)

$1/U = A\Delta T_m / Q_m$; U=419.85W/$m^2$°C (4.9)

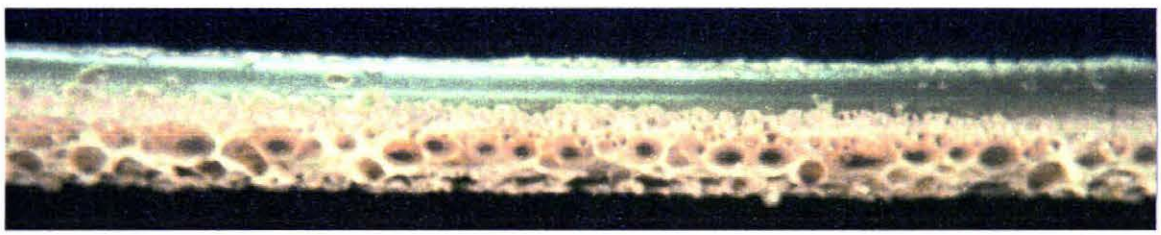
$R_f^* = (1/U) - (1/U_o^{**}) = (U/U_o - 1)$= -0.00072 $m^2$°C/W (4.10)

$R_f U_o = (U_o/U) - 1$ (Biot Number) = 0.24 (4.11)

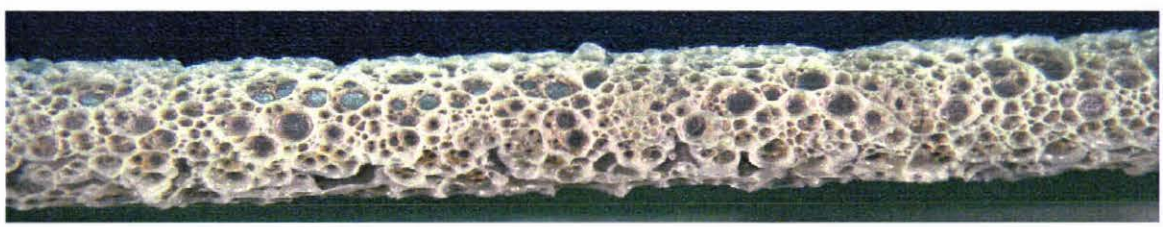
## *Appendix A11 Coverage of fouling from homogenised and un-homogenised milk on the horizontal tubes at different temperatures.*

**Fouling from homogenised milk in the horizontal tube at 70°C. Top side (a) and bottom (b)**

(a)



(b)



**Fouling from homogenised milk at 80°C. Top side (a) and bottom side (b).**

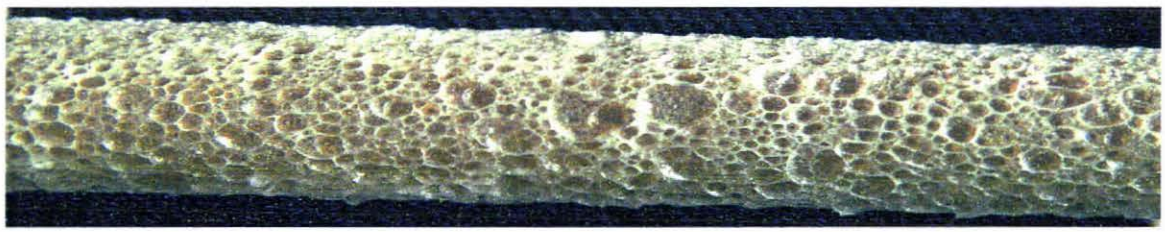
(a)

(b)

**Fouling from un-homogenised milk at 60°C. Top side (a) and bottom side (b).**

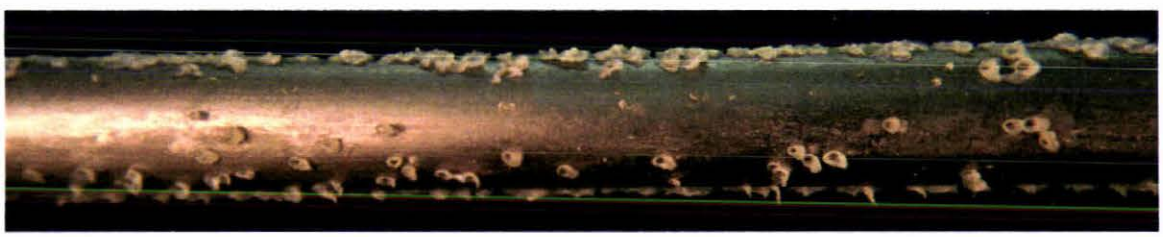
(a)

(b)

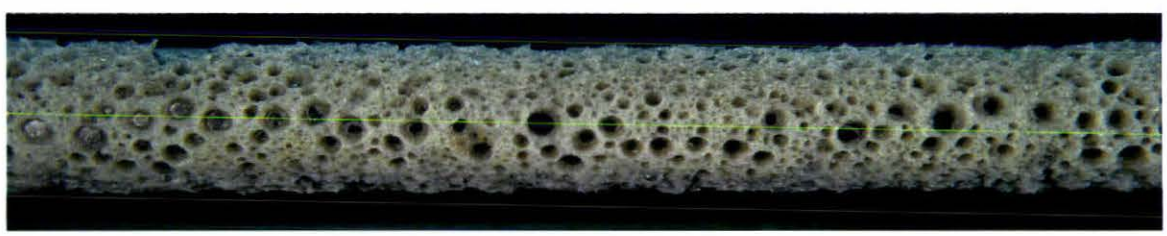


**Fouling from un-homogenised milk at 70°C. Top side (a) and bottom side (b).**

(a)

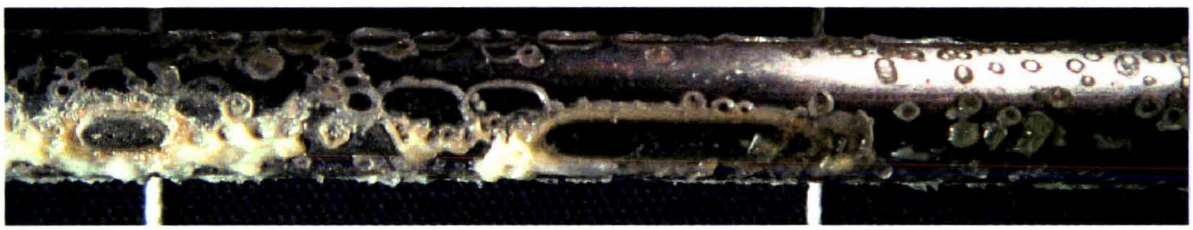


(b)

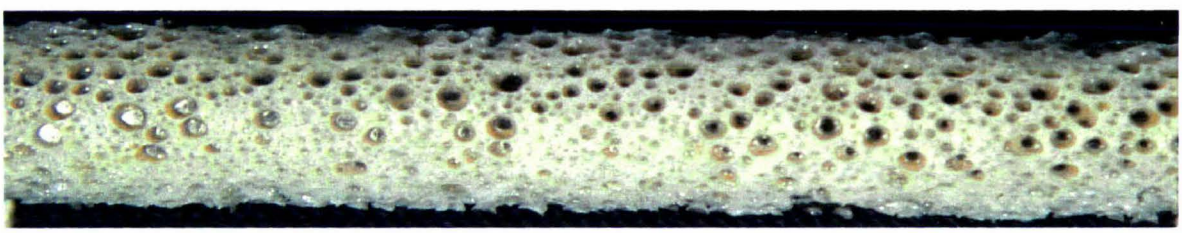


**Fouling from un-homogenised milk at 80°C in horizontal tube. Topside (a) bottom side (b).**

(a)



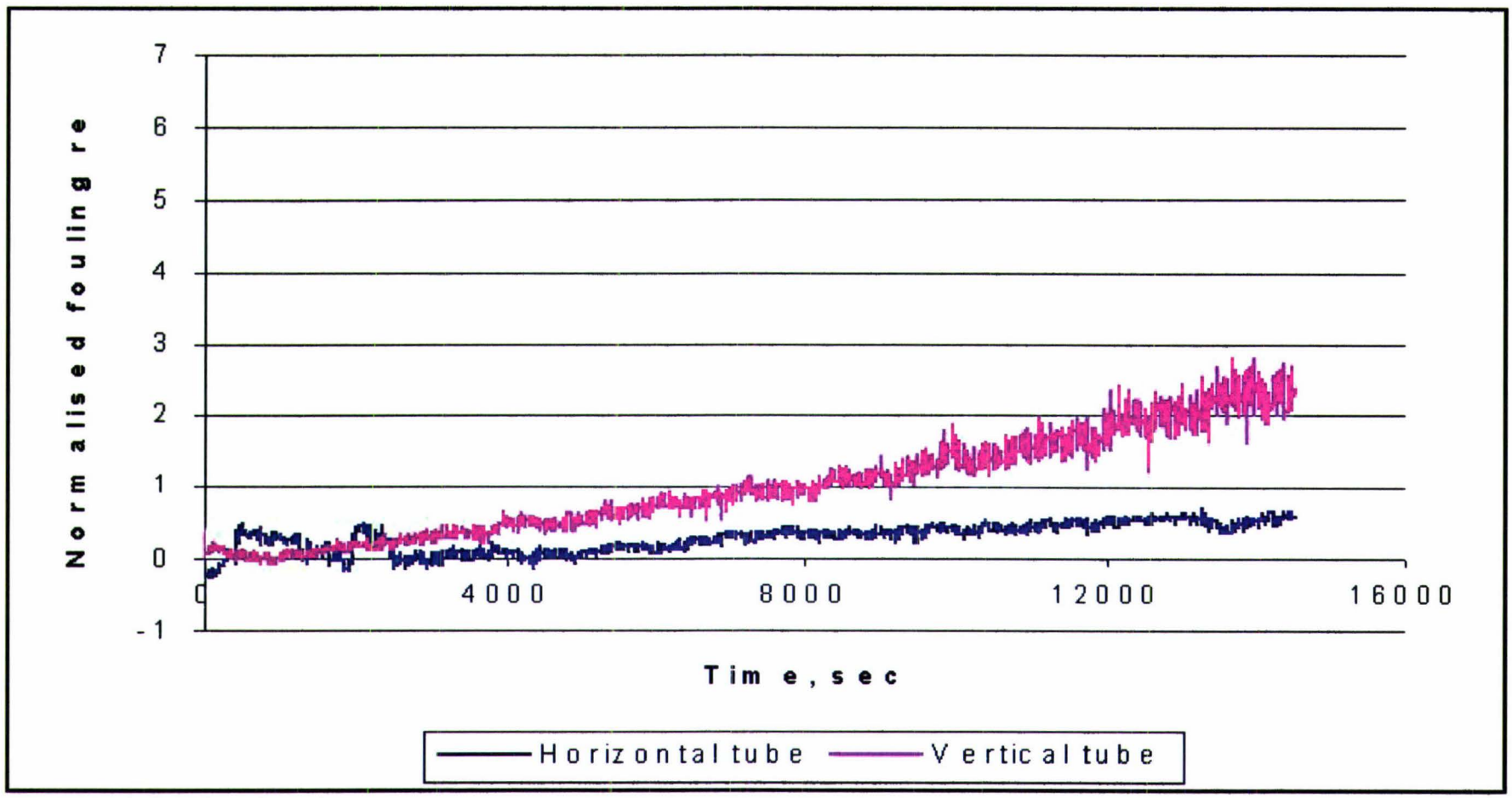
(b)



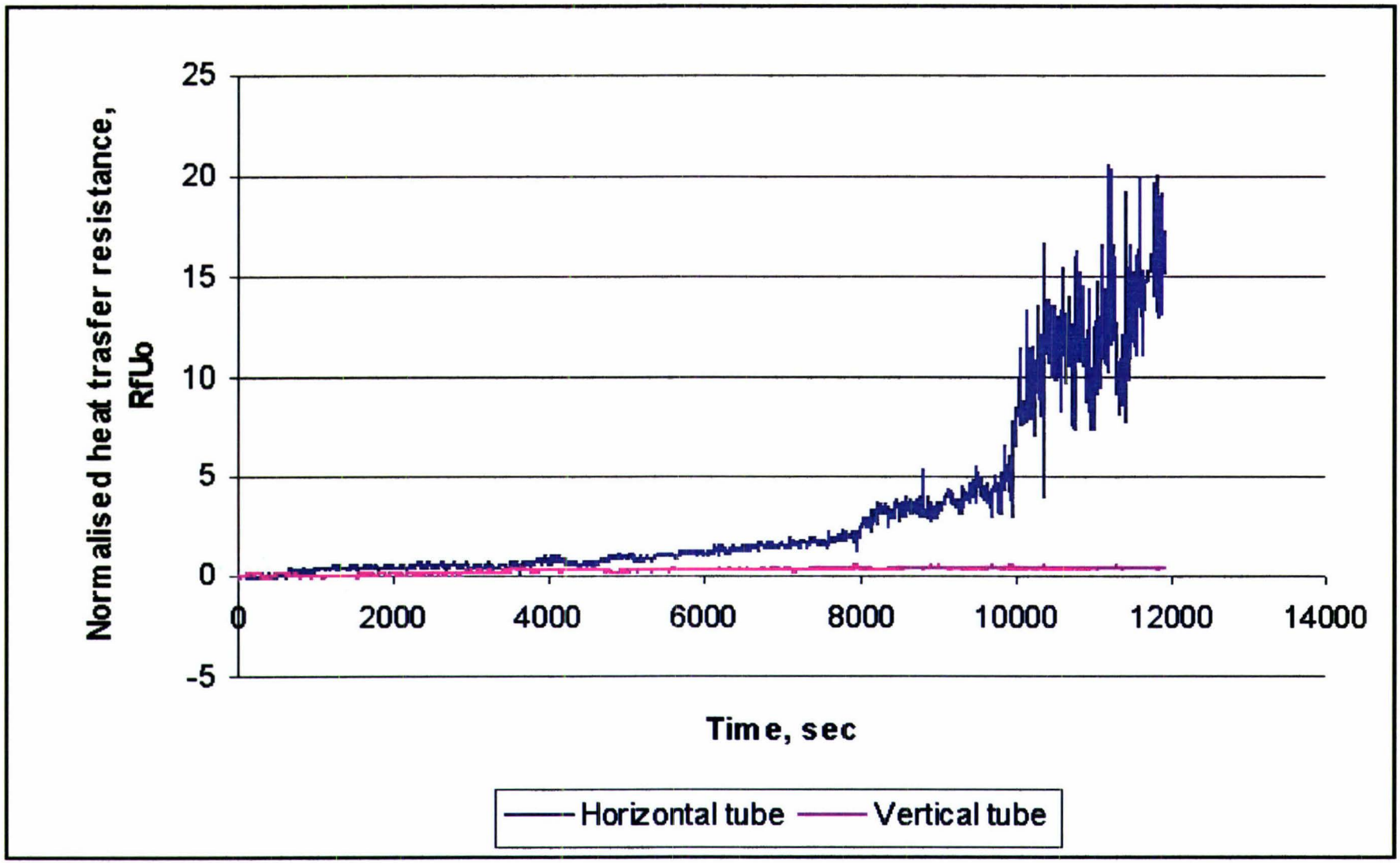
## Appendix 12 Normalised heat Transfer Resistance curves

**Fouling from homogenised milk. Inlet temperature: 60°C, (a), 70°C (b) and 80°C (c)**

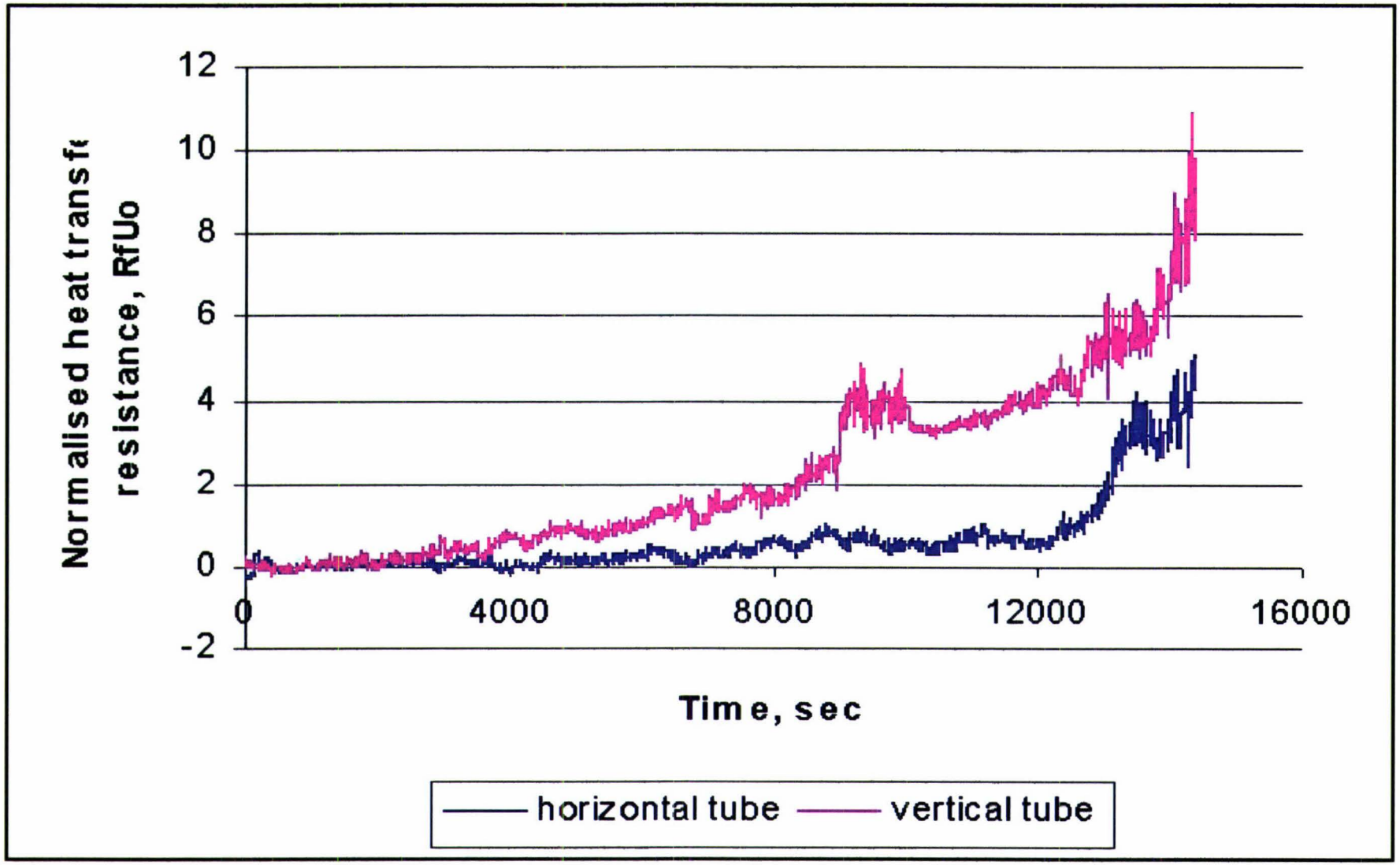
(a)



(b)

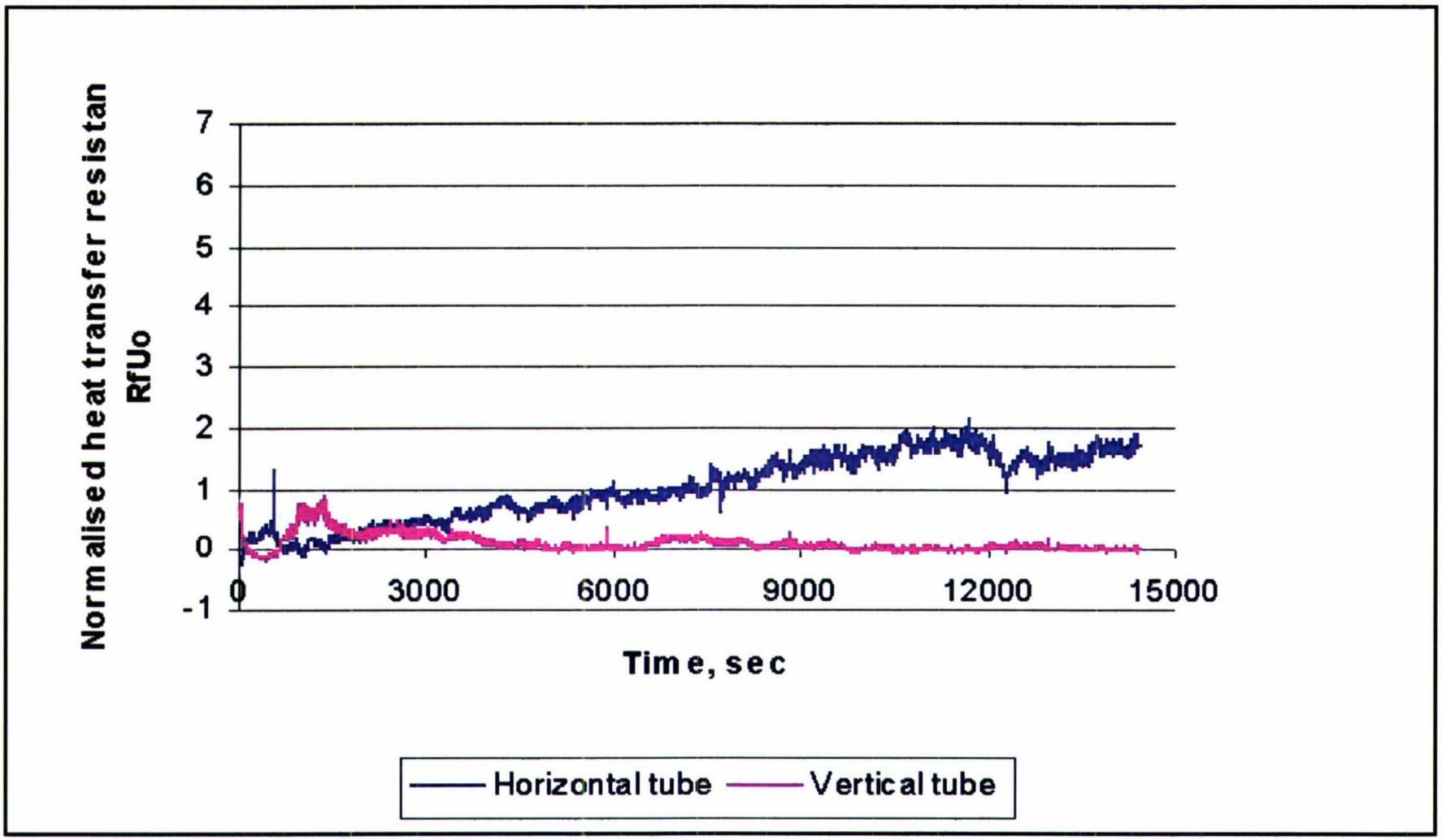

Normalised heat trasfer resistance, RfUo
25
20
15
10
5
0
-5
0
2000
4000
6000
8000
10000
12000
14000
Time, sec
Horizontal tube
Vertical tube

(c)

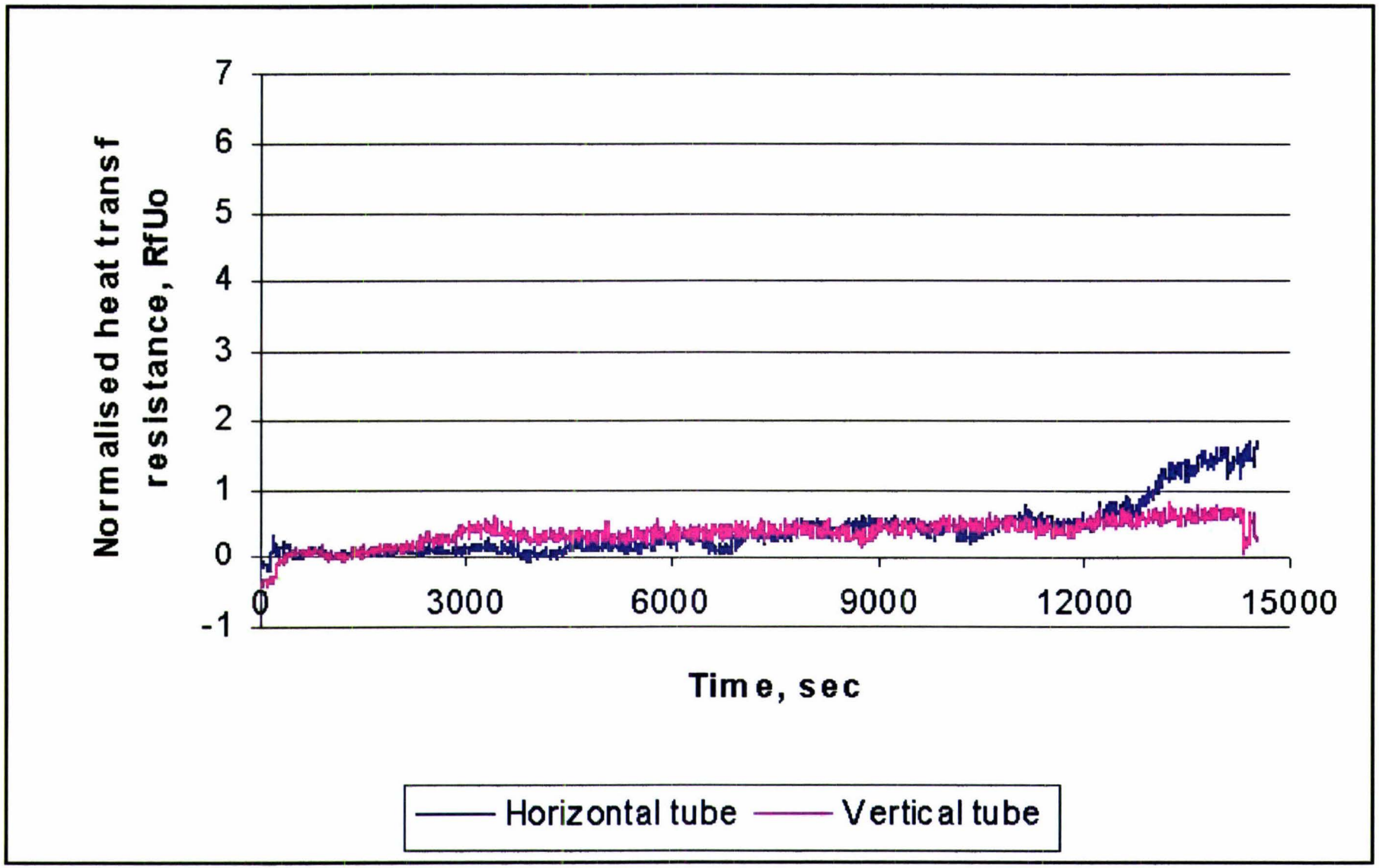

Normalised heat transfe
resistance, RfUo
12
10
8
6
4
2
0
-2
0
4000
8000
12000
16000
Time, sec
horizontal tube
vertical tube

Fouling from un-homogenised milk. Inlet temperature: 70°C (a) and 80°C (b)

(a)



(b)


Normalised heat transf resistance, RfUo
7
6
5
4
3
2
1
0
-1
0
3000
6000
9000
12000
15000
Time, sec
Horizontal tube
Vertical tube