Influence of Run Time and Aging on Fouling and Cleaning of Whey Protein Deposits on Heat Exchanger Surface

Adel Fickak & Emma Hatfield

Biotechnology and Food Engineering Group

Department of Chemical Engineering, Monash University

Clayton Campus, Victoria 3800, Melbourne, Australia

Xiaodong Chen

Biotechnology and Food Engineering Group

Department of Chemical Engineering, Monash University

Clayton Campus, Victoria 3800, Melbourne, Australia

&

Department of Chemical and Biochemical Engineering

College of Chemistry and Chemical Engineering, Xiamen University, P R China

E-mail: xdc@xmu.edu.cn & dong.chen@eng.monash.edu.au

Abstract

In the cleaning operations of heat exchange surfaces in dairy processing plants, the effect of heating/run time (HT) and aging on the fouling/cleaning of heat exchangers is not well understood. Longer heating time of the same deposit is expected to result in the formation of stronger and perhaps more cleaning resistant deposit. In this study, the phenomenon of heating and aging on the formation and cleaning of dairy fouling is investigated using the heat induced whey protein gels (HIWPG) produced in laboratory. The processes were also investigated with the whey protein deposits formed in pilot-scale plant trails.

The HIWPGs were produced in tubular capsules for various heating (or run) times (60, 120, 240, 1440 and 2880 min respectively) and then dissolved in aqueous sodium hydroxide (0.5 wt %). The heating process here would have been of 'pure' aging. The dissolution rate was calculated based on the previously established UV Spectrophotometer analysis. The structure and texture of the gels were analysed using Scanning Electron Microscope (SEM) and texture analyser. In the pilot-scale plant study, whey protein fouling layers were generated by recirculating whey protein solution (6 wt %) for various heating periods (30, 60, and 90 min respectively). The deposit layers were then removed by recirculating aqueous sodium hydroxide (0.5 wt %) and the cleaning efficiency was monitored in the form of the recovery of heat transfer coefficient while both fluid electric conductivity and turbidity were recorded as indications of cleaning completion. It was found that increasing the HIWPG heating time significantly increased the gel hardness and dissolution time, implicating the difficulty in cleaning. Similarly to these results on gels, increasing the pilot-scale plant heating/run time increased the extent of fouling. The fouling layer formed next to the metal surface experienced the longest period of aging and the slope of the heat transfer coefficient increase seen at the final cleaning stage is related to this aging effect. The rate of cleaning for deposits formed initially on the metal surface is lower indicating a 'pure' aging effect of the deposit near surface.

Keywords: Cleaning-in-place (CIP), Fouling, Heat induced gels, Dissolution, Dairy processing, Deposit aging

1. Introduction

It is known that heating of dairy fluids is a normal operation for pasteurisation or varying viscosity purposes. Sterilisation and evaporation processes also incur heating. The heating/run time (HT) varies between different

products depending on the specifications. However, severe fouling is frequently associated with prolonging the HT. Longer HT means that the fouling layers on the heating surface will experience aging. Though often mentioned as being an important process, the aging process of the fouled dairy deposits is poorly understood in a quantitative manner. It was early documented by Epstein in 1983 as one of the principal mechanistic stages in the fouling process. However, more recent studies have shown that aging facilitates longer reaction time between the fouling layer matrixes (Liu et al., 2002) and affects the fouling process resulting in a strong and firm fouling layer that is difficult to clean particularly when the fouling layer is aged for a long period (Wilson, 2005).

In studying the fouling and cleaning behaviours of dairy fluids, researchers face numerous difficulties in generating a reproducible fouling deposit due to the variations found in milk systems. To overcome these difficulties, many researchers (e.g. Xin et al., 2002a; Xin et al., 2002b) found heat induced whey protein gels (HIWPG) as a suitable model material to study the fouling and cleaning behaviors of the proteinaceous milk fouling. They found HIWPG to have similar nature as type "A" milk fouling. Furthermore, HIWPG are easy to reproduce (shape, size and concentration, and chemical state) in the laboratory under well-controlled conditions (Xin et al., 2002a; Mercadé-Prieto et al., 2006).

In this study, the effect of heating time and aging on the formation and dissolution behaviours of HIWPG has been investigated. The effect of heating time and aging on the fouling and cleaning behaviours of a pilot scale heat exchanger has also been studied using whey protein solution to mirror the finding made in the HIWPG experiment. Whey protein solution was used as an accepted milk fouling model and has been studied for its heat induced chemical changes (Bird and Fryer, 1991; Tuladhar et al., 2002; Xin et al., 2004; Hooper et al., 2006).

2. Experimental

2.1 Materials

The whey protein concentrate (WPC 85) powder was obtained from a local supplier. The approximate composition of the WPC is given in Table 1. The cleaning reagent, 60 wt % sodium hydroxide solution (NaOH), was purchased from LabServ, Melbourne, Australia.

2.2 Methods

2.2.1 Preparation of HIWPG in the lab and dissolution experiments

The same sort of heat induced gels and the methodologies established previously (Xin et al., 2002a; Mercadé-Prieto et al., 2006; Fickak et al, 2011) were used in the current study. All gels were prepared in triplicate and the errors calculated. Briefly, we introduce the procedures as below.

An experimental apparatus (see Figure 1 drawn after Fickak et al, 2011) was employed to dissolve HIWPG in the laboratory. The apparatus consists of an analytic balance, a digital magnetic stirrer plate with a stirring speed controller, a digital controller of the water bath, a dissolution cell (a 600ml shock bottle and a capsule holder) and a peristaltic pump to recirculate the solutions through a UV spectroscope linked to a computer.

The HIWPGs were formed in 'capsule' shape using the method developed by Mercadé-Prieto and Chen (2006). HIWPGs were formed inside test tubes (inner diameter 12 mm; length 75 mm) using well mixed 17 wt % whey protein solutions (pH 6.4 ± 0.05). The capsules were filled with the whey solution, then covered with plastic stoppers and sealed with foil, and then held vertically in a water bath kept at 80 °C for different heating times ('pure aging') (60, 120, 240, 1440 and 2880 min respectively). After that and before the dissolution, the top 2 mm of the gel was removed with a spatula to ensure an even surface before each dissolution experiment (Mercadé-Prieto et al., 2008). The gels were kept at 4 °C overnight before use. The gel capsules were then dissolved in batch mode and the dissolution rate of each gel was calculated based on the measured increase of the dissolved protein concentration in the NaOH solution over time. The concentration of NaOH solution was verified by titration of an aliquot with HCl (0.01 N). As shown in Figure 1, the gel capsules were held vertically with stainless steel wire that was fixed to a hole at the top of the bottle lid and submerged inside a 600 ml test bottle containing 500 ml of NaOH (0.5 wt %) solution, which was placed in a water bath at 60 ± 1 °C. The solution was stirred at 200 rpm using a magnetic stirrer to ensure the reading of a well mixed solution. A small fraction of the agitated solution was circulated through the cell of a UV spectrometer (HP Agilent 8453 Spectrophotometer, model number: G1103A, Agilent Technologies, Melbourne, Australia) using a peristaltic pump. Using the method established by Mercadé-Prieto and Chen (2006) and also adopted by Fickak et al (2011), the absorbance of the dissolved proteins was continuously recorded at 20s intervals for at least 8000s at wavelength 280nm. The rate of dissolution was calculated by measuring the increase of the concentration of protein (calibrated again UV absorbance) in the NaOH solution over time.

2.2.2 Gel strength measurements

The hardness of HIWPGs is an indicator of the ease for removal during cleaning as shearing is involved in deposit removal in reality. The hardness may be affected by the heating time during the gelation process. Here a depth sensing indentation hardness test, was used which has been widely used for characterizing the consistency of fats and other materials (DeMan, 1983). The test was standardized by the American Oil Chemists Society (AOCS) method 'Cs 16-60' (DeMan, 1983). The test involves the penetration of a sample surface by a metal probe indenter with a known geometry (DeMan, 1969). A parameter referred to as the 'yield value' or 'hardness index' can be calculated by monitoring the penetration force of the probe and the time taken to achieve that penetration depth (DeMan, 1983; Narine and Alejandro, 2001). The sample gels were made in a 50 ml beaker using 10 ml of well mixed whey protein solutions (17 wt %) and held in a water bath at 80 °C for different heating times (60, 120, 240, 1440 and 2880 min respectively). The gels were kept at 4 °C overnight and brought to room temperature (23 ± 2°C) before being tested. The texture analyses were performed at room temperature (23 ± 2°C) using a EZ Graph texture analyser ("EZ Graph 100N", Shimadzu Scientific Instruments, Melbourne, Australia) equipped with a 100-N load cell, and a 12.8 mm diameter cylindrical probe. The test procedure was developed using Shimadzu Instruments texture tests guidelines. Each test was performed by allowing the probe at speed of 50 mm min^{-1} to penetrate 10 mm into the gel, and the force at 5 mm of penetration was taken as gel strength value.

2.2.3 Microstructure analysis

Images of the microstructure of the gel samples may be informative of the fouling formation and were obtained using Scanning Electron Microscope (SEM) (JEOL JSM-840A SEM, 1986, Japan), located at Monash University, Clayton Campus, Melbourne, Australia. Prior to image scanning, 2mm^{-2} gel samples were sliced and placed in a dry oven at 30 ^{0}C for 10 min. The gel samples were then sputtering coated (approximately 1mm) with gold palladium for charge dissipations. The samples were then viewed with the SEM operating at 15KV using back scattering electrons.

2.2.4 Pilot-scale fouling and cleaning-in-place (CIP) experiments

A pilot-scale plant test rig was designed, constructed to generate and remove whey protein fouling layers (refer to Figure 2, after Fiakak et al (2011)). This system has a 60 L reservoir was equipped with heating coils to preheat the sample solutions. The solution in the holding tank was stirred continuously by re-circulating through the bypass valves. A centrifugal pump was used to ensure an easy circulation of sample solutions. The flowrate in the heating section was kept constant during the fouling and cleaning processes and monitored through a flowrate meter (Model 257-133' from RS Components, Melbourne, Australia) installed at the inlet of the fouling section. The velocity in the heating section (see Table 2) was calculated using the flowrate and the heating section area.

As mentioned by Fickak et al (2011), the heating section consisted of a heater rod (diameter= 17mm, length =160mm) fixed inside a sealed glass tube reactor (ID= 80mm with length= 300mm) with the bottom inlet of ID=20mm. Three outlets (ID=20mm) were uniformly distributed (120° apart) at the top. A turbidity meter (Model TB750G) and conductivity meter (Model DC402G) from YOKOGAWA Electric Corporation, Melbourne, Australia) were installed downstream of the fouling section to monitor the cleaning process. In the current study, the rig operating conditions were kept constant for each run (see Table 2).

2.2.5 Generation of the fouling layer

Similarly in Fickak et al (2011), whey proteins concentrate powder (WPC, 80 wt %) was reconstituted to protein concentration of 6 wt % (pH 6.4 ± 0.05) in 50 litres of RO water. The solution was then transferred to the holding tank and preheated to approximately 70 ± 0.5 ^{o}C and then held for 5 min before being pumped through the fouling section and back to the tank. The recirculation of the solution in the holding tank through the bypass valve results in a continuous stirring of the solution, this help to sustain the required tank temperature. At the start of the experiment, the heater rod surface temperature (the average of the temperature around the outside diameter of the heater rod upper, middle and bottom surface) was obtained at approximately 81 ± 1 ^{o}C by applying the heat flux described in Table 2 using a 5A variac autotransformer. The fouling layers were formed in 30, 60 and 90 min respectively. The fouling layer formed in each run was found to be reasonably evenly distributed along the heater rod surface. The small flow velocity (see Table 2) was employed to exaggerate the heat-to-foul effect as the removal force by the fluid became rather small.

2.2.6 Monitoring of fouling

The most common method for monitoring fouling and cleaning is to record the change in the heat transfer coefficient. A drop in the heat transfer coefficient simply indicates the formation of a fouling layer on the heater surface. The heat transfer coefficient was calculated using the following equation:

$$U \approx \frac{Q}{A(T_S - T_B)}$$

(1)

where U = overall heat transfer coefficient (W.m^{-2}.K^{-1}); Q = power input to the heater rod (W); A = heater rod surface area (m^{-2}); T_S = heater rod surface temperature (^{o}C) (The average of the temperature around the outside diameter of the heater rod upper, middle and bottom surface); T_B = bulk fluid temperature (^{o}C) measured in the fouling section.

2.2.7 Monitoring of the temperatures of the heater rod surface

As mentioned above the average temperature measurement of the heater rod surface was obtained by attaching three thermocouples around the outside diameter of the upper, middle and bottom surface of the heater rod. The bulk fluid temperature was measured through another thermocouple that was placed inside the bulk solution in the heating section of the transparent section leading to the heater rod. The inlet and outlet temperatures of the fouling section were monitored using thermocouples inserted at the inlet and outlet of the fouling section. The tank temperature was monitored using a thermocouple located at the centre of the tank. All measured temperatures (surface, bulk, inlet, outlet and tank) were logged to a computer.

2.2.8 Cleaning of the fouled surface

The fouled surface was cleaned using a three-stage cleaning method. In the first stage, after the fouling layer was formed the whey protein solution was drained and the system was rinsed with water at a velocity of about 0.104 m.s^{-1}, for approximately 10 min, until there were no protein traces left in the rinsing water. The rinsing efficiency was indicated using the turbidity meter by detecting the level originally present in the clean water stream. The rinsing process was stopped once the standard turbidity of drinking water (0.5 to 1NTU) was reached (USEPA, 2001).

In the second stage, similar to that described by Fickak et al (2012) in a study emphasizing the effect of protein concentration, the cleaning solution (50L of NaOH at 0.5 wt %) was used. During cleaning, the cleaning solution temperature was kept constant at 60 ± 0.5 ^{o}C. In the cleaning in place (CIP) process, first the cleaning solution was recirculated through the system. The heater rod surface temperature (approximately 66 ± 0.5 ^{o}C) was attained by applying the heat flux described in Table 2 using enclosed 5A variac autotransformer. The CIP process was monitored visually by observing the change in the heat transfer coefficient and by visually observing the complete removal of the fouling layer through the glass wall of the fouling section (see Figure 6b). An increase of the heat transfer coefficient corresponds to the removal of the fouling layer on the heater surface. The CIP solution was drained when the fouling layer was seen to be completely removed. The cleaning fluid velocity was set at 0.104 m.s^{-1}, which was the comfortable upper limit of the rig.

In the third stage, the system was continuously rinsed with water for approximately 10 minutes until there were no NaOH traces left in the rinsing water. The rinsing at this stage was monitored using the conductivity meter. The rinsing was stopped when the typical conductivity of drinking water (<500 μS/cm) (DeZuane, 1990) was reached. The turbidity and conductivity measurements were both used to indicate the efficiency of the cleaning process. After this, an observation of the heater rod was made for the cleanliness of the above processes.

3. Results and Discussion

3.1 Effect of heating time on HIWPG formation

The scanning electron microscope (SEM) images of the HIWPG deposits microstructure (see Figure 3) show that the HIWPG deposit formed at shorter heating time (30 min) contains small and perhaps undeveloped aggregates (see Figure 3a), while the HIWPG deposit formed at longer heating time (1440 min) contained more and larger aggregates (see Figure 3b).

The texture analysis (see Figure 4) shows that the HIWPG deposit formed at shorter heating time (60 min) required a penetration force of approximately 3N to penetrate through the gel, indicating a soft textured gel. In contrast, the HIWPG deposit formed at longer heating time (1440 min) required much higher force of approximately 25N to penetrate through the gel surface, indicating a firm textured gel.

These results can be explained as follows. Upon heating the whey protein solution forms a gel due to the reaction between the proteins molecules in the matrix forming many small monomers. These protein monomers contain a number of cysteine residues (Sawyer et al., 1985; Brownlow et al., 1997) and upon heating the free – SH groups of cysteine residues get oxidized and form disulfide bonds (Verheul and Roef, 1998; Galani and Apenten, 1999). These bonds are involved in cross-linking the protein monomers to form the aggregates (Galani and Apenten, 1999). However, increasing the heating time provides more reaction time between the protein matrixes, increasing the number of disulphide cross-links between the protein molecules. These protein molecules may aggregate more intensively to form a hard and rigid gel. The longer the heating time, the harder the gel becomes.

3.2 Effect of heating time on HIWPG dissolution

The dissolution experiments of HIWPG deposits obtained with different heating times (see Figure 5) show that HIWPG deposit heated for longer time (1440 min) had the lowest dissolution rate ($2.23g \ m^{-2} \ s^{-1}$). Another observation was that the dissolution rate of HIWPG deposit formed in 240 min or longer (1440 min) remain almost the same (0.24 and $0.23g \ m^{-2} \ s^{-1}$) indicating the structure of the gels stabilized after 240 min.

As discussed earlier, longer heating time provides the time for greater extent of the reactions to happen, in particular for the thiol-disulfide exchange reactions (Livney et al., 2003; Jayat et al., 2004). The disulfide bridges are known to be a key factor that affects the gel strength (Hoffmann and Van Mil, 1999; Creamer et al., 2004) and the aggregation process through the polymerization of monomers to form large polymer chains. However, there is only limited availability of the bonds to be formed and hence excessively longer heating time may not be effective anymore. In the dissolution of large polymer chains, or gels, (Devotta et al., 1995; Narasimhan and Peppas, 1996) predicted that the large clusters or the large chains polymers are expected to disengage very slowly from the gel matrix into the solvent. Our results seem to be within the expectation based on previous works.

3.3 The effect of heating/run time on whey protein solution fouling

Figure 6a shows the examples of fouled and cleaned heater rod. The fouling layer formed is similar as type 'A' deposits described by (Lyster, 1965; Burton, 1968). It can be seen that longer heating time resulted in visibly more (denser) fouling (the thickness is not measured here). Figure 7 shows (a) the overall heat transfer coefficient profile during the fouling process; and as an example (b) the temperature difference (ΔT) between the heat exchange surface (average) and the bulk solution during the 90 min run. The decreasing overall heat transfer coefficient is an indication of the fouling layer formation on the heat transfer surface. The formation of fouling layer on the heat exchange surface leads the overall heat transfer coefficient to drop.

The fouling behaviour observed in the first 30 min was similar for all runs particularly for the 30 and 60 min runs. The overall heat transfer coefficient decreased continuously with increasing run time (see Figure 7) indicating the formations of more heat resistant fouling on the processing surface. Since the deposit chemical composition would not change during heating so the same deposit experiences 'aging' in essence. The possible explanation for this is that heating the whey protein solutions above 60°C, provokes complex changes of the molecular conformation and undergo denaturation (Aymard et al., 1999). Similarly as discussed in the HIWPG formation, this leads the protein molecules to exhibit hydrophobic areas on its surface (Cantor and Schimmel, 1980; Verheul and Roef, 1998; Galani and Apenten, 1999). The denaturation of whey proteins particularly β-lactoglobulin is considered by many e.g. Grijspeerdt et al. (2004), to be significant in most dairy fouling. The partially denatured proteins aggregate via the exposed covalent disulfide bonds (Galani and Apenten, 1999; Perez-gago and Krochta, 2001) and eventually precipitate on the heat exchange surface. The kinetics of aggregation and the aggregation rate are influenced by the heating conditions (Aymard et al., 1999). With increasing heating time, the formation of strong covalent disulfide bonds may become more extensive within the deposit. The aggregates grow in strength and size, while the fraction of native proteins decreases (Durand et al., 2002). These observations are, in principle, consistent with the observations of Liu et al (2002) on non-dairy product and Wilson (2005) on a general discussion who suggested that increasing the heating time results in increasing the yield and the deposit hardness due to the deposit structural changes. Regardless of the 'structural consolidation', the accumulation of deposit on the heat transfer surface should lead, in any case, to greater thermal resistance hence the overall heat transfer reduction shown Figure 7.

3.4 The effect of heating/run time during fouling on the subsequent cleaning

Figure 8 shows (a) the overall heat transfer coefficient profile during the cleaning in place (CIP) process; and as an example (b) the temperature difference (ΔT) between the heat exchange surface (average) and the bulk solution during the cleaning process of the fouling layer from 4wt % whey protein solution. The time course of

the recovery of the overall heat transfer coefficient is equivalent to the cleaning time required to remove the fouling layer from the heat transfer surface.

Figure 8 shows the recovery of the overall heat transfer coefficient (CIP, or removal time). CIP time of about 1400s was required to remove the deposits formed at the longest (90 min) heating/run time. A CIP time of about 500s and of about 1000s were needed to clean deposits formed at shorter heating/run times of 30 min and of 60 min respectively. The slopes of the first stage of the cleaning curve were not so different, which is an indication of the nature of the deposits that were formed at the final stages of fouling where the temperature at the fouling layer surface would be fairly close to the bulk solution temperature, and the fouling formed at this stage would be weak and more likely interacted via hydrogen bonds which is highly water soluble (Perez-gago and Krochta, 2001) making it easy to remove. The slopes of the cleaning curve in the final stages of the cleaning should indicate more accurately the effect of ageing as this should be the cleaning of the most inner layers of the deposits which would have 'consolidated' for the longest time. The results by comparison are quite dramatic. The slopes marked as $\alpha 1$, $\alpha 2$ and $\alpha 3$ in Figure 8 simply indicates that the aging is most pronounced at the inner layers and that the longer the aging time of the inner layers, the harder it is to remove the fouling. This mirrors the results of the gel strength tests and the gel dissolution experiments (the rate of the recovery of the overall heat transfer coefficient α values for the three different fouling periods are plotted against cleaning time; see Figure 5), described earlier.

4. Conclusions

The HIWPGs, obtained for different heating periods, show dissolution behaviour which mirrors well the cleaning behaviour of whey protein fouling layers formed in the pilot-scale rig as far as the trend is concerned. In the HIWPG experiment, longer heating time ('pure aging' behaviour) produced harder gel materials. The dissolution rate of HIWPG decreased with increasing heating/run (aging) time, and the rate stabilizes after 240 min of heating/aging.

In the pilot-scale experiment, two separate effects are inter-related: The heating/run time and the aging. The prolonged experiment created more fouling. The inner fouling layer experienced considerable ageing. The aging has caused significant reduction on the cleaning rate, this effect may be due to the further chemical bond formation or some kind of structural re-arrangements within the deposit which does not affect much the heat conduction, but clearly affect the difficulty of disentanglements (thus harder to remove). The exterior fouling layer shows similar cleaning rate regardless of run time.

Acknowledgements

Nomenclature

Q   = Power requirement (W)

A   = Heating surface area (m-2)

T   = Temperature (K)

α = Slope (J.m-2.K-1.s-2)

HT   = Heating time (s)

Subscripts

S   Heater surface

B   Bulk

References

Aymard, P., Nicolai, T., & Durand, D. (1999). Static and dynamic scattering of β-Lactoglobulin aggregates formed after heat-induced denaturation at pH 2 Macromolecules, 32, 2542-2552.

Bird, M. R., & Fryer, P. J. (1991). An experimental study of the cleaning of surfaces fouled by whey proteins. *Food Bioproducts Proc*, 69, 13-21.

Brownlow, S., Morais Cabral, J. H., Cooper, R., *et al.* (1997). Bovine β-lactoglobulin at 1.8 A resolution - still an enigmatic lipocalin. *Structure*, 5, 481- 495. http://dx.doi.org/10.1016/S0969-2126(97)00205-0

Burton, H. (1968). Deposits from whole milk in heat treatment plant - a review and discussion. *J Dairy Res*, 35, 317-330. http://dx.doi.org/10.1017/S0022029900019038

Cantor, C. R., & Schimmel, P. (1980). Biophysical Chemistry: Freeman and Company: San Francisco.

Creamer, L. K., Bienvenue, A., Nilsson, H., *et al.* (2004). Heat-induced redistribution of disulfide bonds in milk proteins. 1. bovine β-Lactoglobulin. *Journal of Agricultural and Food Chemistry*, 52, 7660-7668. http://dx.doi.org/10.1021/jf049388y

Deman, J. M. (1969). Food texture measurements with the penetration method. *J Texture Stud*, 1, 114-119. http://dx.doi.org/10.1111/j.1745-4603.1969.tb00960.x

Deman, J. M. (1983). Consistency of fats: A Review. *J Am Oil Chem Soc*, 60, 82-89. http://dx.doi.org/10.1007/BF02540897

Devotta, I., Badiger, M. V., Rajamohanan, P. R., Ganapathy, S., & Mashelkar, R. A. (1995). Unusual retardation and enhancement in polymer dissolution - role of disengagement dynamics. *Chem Eng Sci*, 50, 2557-2569. http://dx.doi.org/10.1016/0009-2509(95)00103-C

Dezuane, J. (1990). Handbook of drinking water Quality standards and controls, New York: Van Nostrand Reinhold.

Durand, D., Christophe, J. G., & Nicolai, T. (2002). Aggregation, gelation andphase separation of heat denatured globular proteins. *Physica A*, 304, 253-265. http://dx.doi.org/10.1016/S0378-4371(01)00514-3

Epstein, N. (1983). Thinking about heat transfer fouling: a 5x5 matrix. *Heat Transfer Eng*, 4, 43-56. http://dx.doi.org/10.1080/01457638108939594

Fickak, A., Al-Raisi, A., & Chen, X. D. (2011). Effect of protein concentration on the fouling and cleaning of dairy heat exchangers. *Journal of Food Engineering*, 104(3), 323-331. http://dx.doi.org/10.1016/j.jfoodeng.2010.11.004

Fryer, P. J., Hasting, A. P. M., & Jeurnink, T. (1996). Fouling, cleaning and disinfection in food processing. Luxembourg: Office for official publications for the European Communities, EUR 16894.

Galani, D., & Apenten, R. K. O. (1999). Heat-induced denaturation and aggregation of β-Lactoglobulin: kinetics of formation of hydrophobic and disulphide-linked aggregates. *International Journal of Food Science & Technology*, 34, 467-476. http://dx.doi.org/10.1046/j.1365-2621.1999.00314.x

Grijspeerdt, K., Mortier, L., De Block, J., & Van Renterghem, R. (2004). Applications of modelling to optimise ultra high temperature milk heat exchangers with respect to fouling. *Food Control*, 15, 117-130. http://dx.doi.org/10.1016/S0956-7135(03)00023-9

Hoffmann, M. A. M., & Van Mil, P. J. J. M. (1999). Heat-induced aggregation of β-Lactoglobulin as a function of pH. *J Agric Food Chem*, 47, 1898-1905. http://dx.doi.org/10.1021/jf980886e

Hooper, R. J., Paterson, W. R., & Wilson, D. I. (2006). Comparison of whey protein model foulants for studying cleaning of milk fouling deposits. *Food and Bioproducts Processing*, 84, 329-337. http://dx.doi.org/10.1205/fbp06028

Jayat, D., Gaudin, J. C., Chobert, J. M., Burova, T. V., Holt, C., Mcnae, I., Sawyer, L., & Haertle, T. (2004). A Recombinant C121S mutant of bovine β-Lactoglobulin is more susceptible to peptic digestion and to denaturation by reducing agents and heating. *Biochemistry*, 43, 6312-6321. http://dx.doi.org/10.1021/bi0362469

Liu, W., Christian, G. K., Zhang, Z., & Fryer, P. J. (2002). Development and use of a micromanipulation technique for measuring the force required to disrupt and remove fouling deposits. *Food and Bioproducts Processing*, 80, 286-291. http://dx.doi.org/10.1205/096030802321154790

Livney, Y. D., Verespej, E., & Dalgleish, D. G. (2003). Steric effects governing disulfide bond interchange during thermal aggregation in solutions of β-Lactoglobulin B and α-Lactalbumin. *Journal of Agricultural and Food Chemistry*, 51, 8098-8106. http://dx.doi.org/10.1021/jf034582q

Lyster, R. J. L. (1965). The composition of milk deposits in an UHT plant. *J Dairy Res*, 32:203. http://dx.doi.org/10.1017/S0022029900018549

Mercadé-Prieto, R., & Chen, X. D. (2006). Dissolution of whey protein concentrate gels in alkali. *AIChE Journal*, 52, 792 - 803. http://dx.doi.org/10.1002/aic.10639

Mercadé-Prieto, R., Falconer, R. J., Paterson, W. R., & Wilson, D. I. (2006). Probing the mechanisms limiting dissolution of whey protein gels during cleaning. *Food and Bioproducts Processing*, 84, 311-319. http://dx.doi.org/10.1205/fbp06021

Mercadé-Prieto, R., Paterson, W. R., Wilson, D. I. (2008). Comparison between the dissolution of whey protein gels and of synthetic polymers. *Journal of Polymer Science*, Part B; Polymer Physics, 46, 1007-1021. http://dx.doi.org/10.1002/polb.21434

Narasimhan, B., & Peppas, N. A. (1996). On the importance of chain reptation in models of dissolution of glassy-polymers. *Macromolecules*, 29, 3283-3291. http://dx.doi.org/10.1021/ma951450s

Narine, S. S., & Alejandro, G. M. (2001). Elastic modulus as an indicator of macroscopic hardness of fat crystal networks. *Lebensmittelwissenschaft und Technologie*, 34, 33-40. http://dx.doi.org/10.1006/fstl.2000.0719

Perez-Gago, M. B., & Krochta, J. M. (2001). Denaturation time and temperature effects on solubility, tensile properties, and oxygen permeability of whey protein edible films source. *Journal of Food Science & Technology*, 66, 705-710.

Sawyer, L., Papiz, M. Z., North, A. C. T., & Eliopoulos, E. E. (1985). Structure and function of bovine β-lactoglobulin. *Biochemical Society Transactions*, 13, 265-266.

Tuladhar, T. R., Paterson, W. R., & Wilson, D. I. (2002) Investigation of alkaline cleaning-in-place of whey protein deposits using dynamic gauging. *Food and Bioproducts Proc*, 80, 199-214. http://dx.doi.org/10.1205/096030802760309223

Usepa. (2001). 'United states environmental protection agency'. Office of water (4606). EPA 816-F-01-011.

Verheul, M., & Roef, S. P. F. M. (1998). Structure of whey protein gels, studied by permeability, scaning electron microscopy and reology. *Food Hydrocolloids*, 12, 17-24. http://dx.doi.org/10.1016/S0268-005X(98)00041-1

Wilson, D. I. (2005). Challenges in cleaning: recent developments and future prospects. *Heat Transfer Engineering*, 26 (1), 51-59. http://dx.doi.org/10.1080/01457630590890175

Xin, H., Chen, X. D., & O Zkan, N. (2004). Removal of a model protein fouling from metal surfaces. *AIChE J*, 50, 1961-1973. http://dx.doi.org/10.1002/aic.10149

Xin, H., Chen, X. D., & Ozkan, N. (2002a). Whey protein based gel as a model material for studying the initial cleaning mechanisms of milk fouling. *J Food Sci*, 67, 2702–2711. http://dx.doi.org/10.1111/j.1365-2621.2002.tb08802.x

Xin, H., Chen, X. D., & Ozkan, N. (2002b). Cleaning rate in the uniform cleaning stage for whey protein gel deposits. *Trans IChemE*, 240-246.
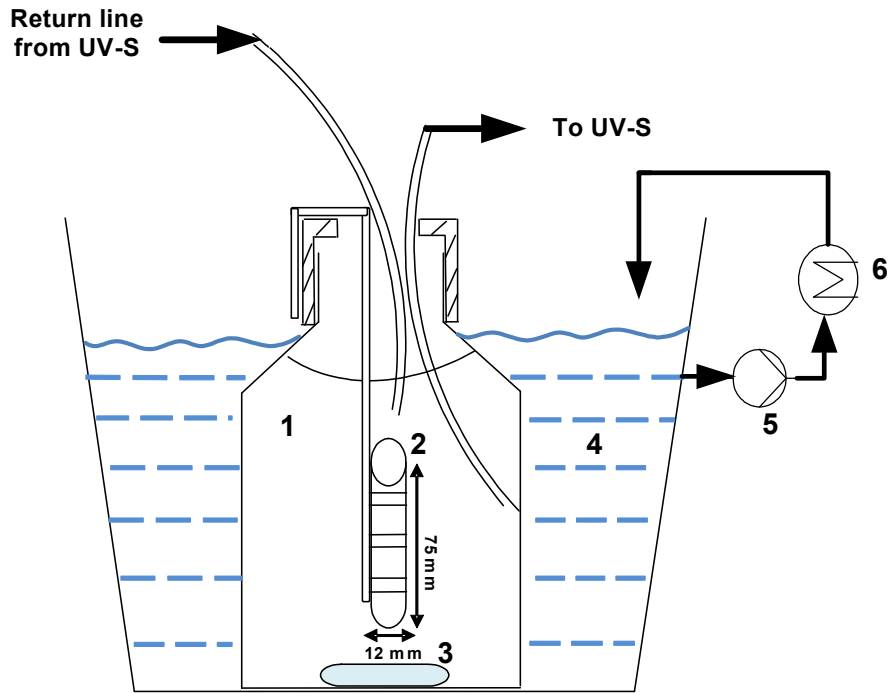
Table 1. Whey Protein Concentrate (WPC) powder composition

Component	Content (wt %)
Proteins	82.0
Fat	6.2
Moisture	3.5
Ash	8.3

Table 2. Pilot plant operation conditions during fouling and CIP processes

Operation variable	Fouling	CIP
Feed tank liquid volume	50 L	50 L
Feed tank temp	$70 \pm 0.5^{o}C$	$60 \pm 0.5^{o}C$
Velocity	0.001 m.s^{-1}	0.104 m.s^{-1}
Heat flux	6.560 kW.m^{-2}	1.098 kW.m^{-2}

1. Dissolution solvent (0.5 wt %) NaOH
2. Capsule containing heat induced whey protein gel
3. Magnetic stirrer
4. Water bath
5. Pump
6. Heater

Figure 1. Apparatus for the dissolution of heat induced when protein gels
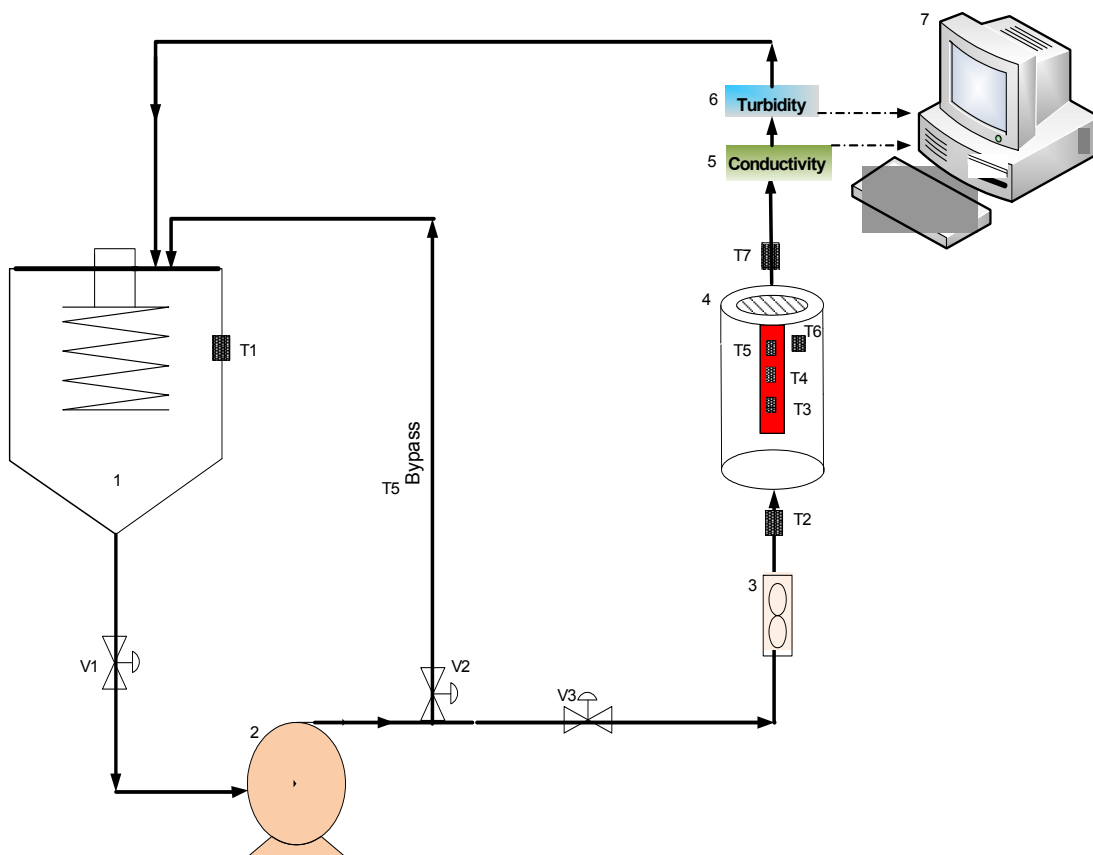
1 Tank including coiled heater
2 Centrifugal pump
3 Flow meter
4 Fouling section (include a heater rod)
5 Conductivity meter
6 Turbidity meter
7 Computer for temperature, turbidity and
conductivity recording

TI-1: Tank temperature sensor
TI-2: Fouling section inlet temperature sensor
TI-3: Heater rod bottom surface temperature sensor
TI-4: Heater rod middle surface temperature sensor
TI-5: Heater rod top surface temperature sensor
TI-6: Bulk solution temperature sensor
TI-7: Fouling section outlet temperature sensor
((TI-3) + (TI-4) + (TI-5)) / 3: Surface temperature

Figure 2. Fouling and cleaning test system

(a)   **(b)**
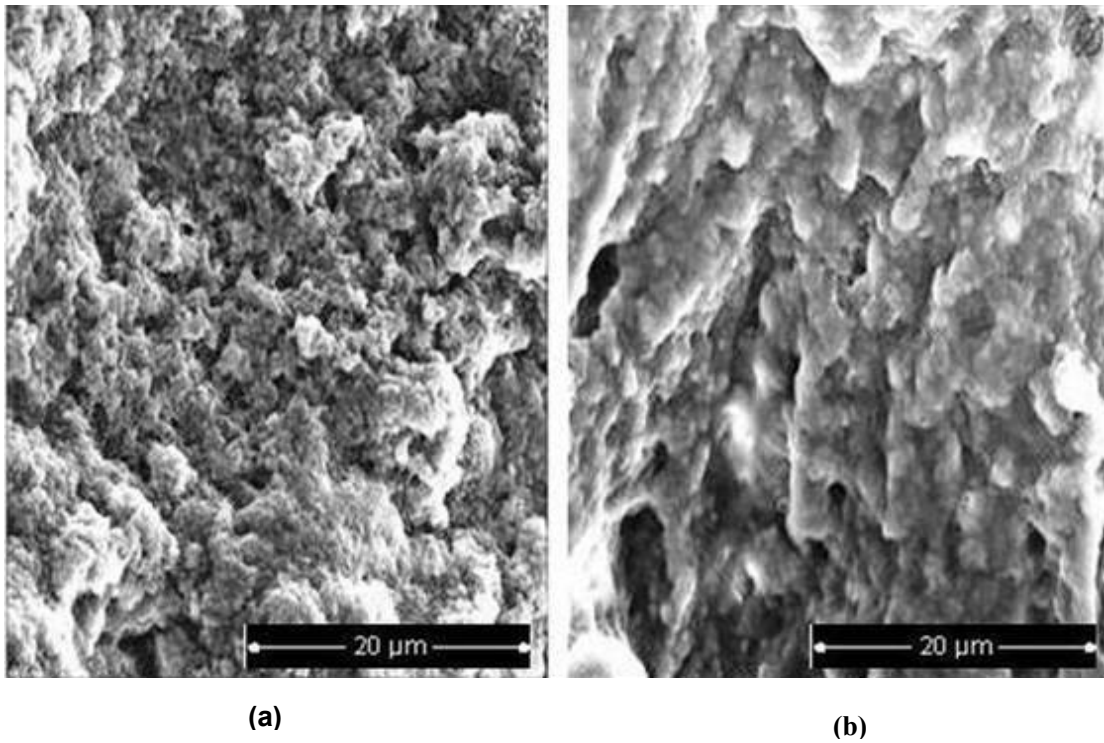
Figure 3. SEM images of the microstructure of the 17 wt % HIWPG formed at (a) 30 min heating time, and (b) 1440 min heating time
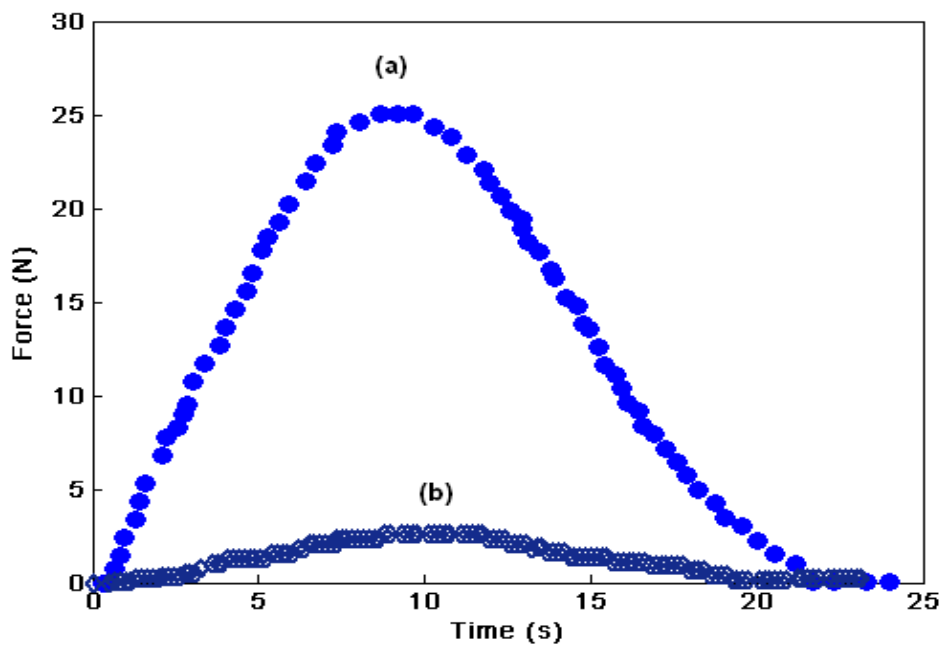

Figure 4. Texture analysis of (a) 17 wt % HIWPG obtained after 1440 min, and (b) 17 wt % HIWPG obtained after 60 min
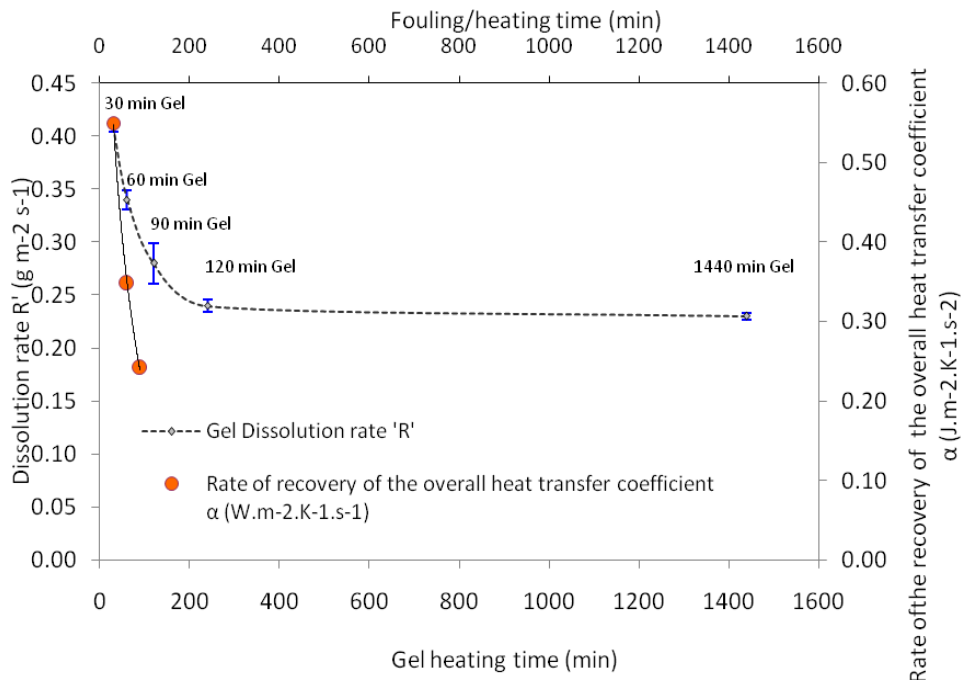
Figure 5. Effect of different heating time (60, 120, 240 and 1440 min) on the dissolution rate 'R' of 17 wt % HIWPG in 0.5 wt % NaOH at 60 $^{\circ}$C, compared with the slopes '$\alpha 1$, $\alpha 2$ and $\alpha 3$' from the CIP cleaning curves in Fig 11 corresponding to different fouling/heating times

(a)   (b)

Figure 6. (a) Fouled rod heater with, 6 wt % whey protein solution; fouling layer formed in 90 min heating time before cleaning, and (b) after cleaning
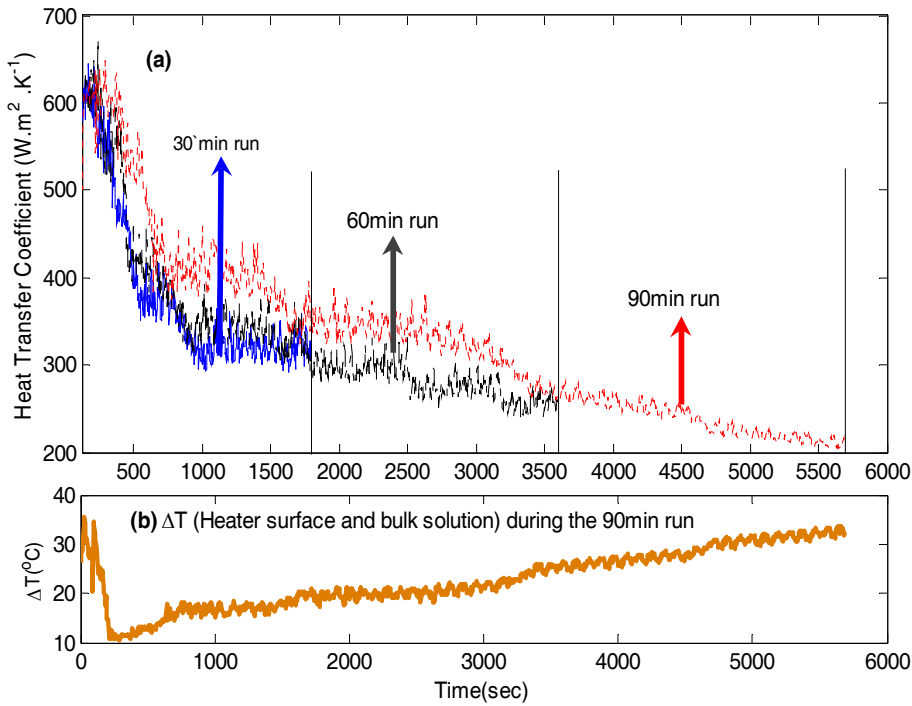
Figure 7. (a) Effect of run/heating time and aging on the fouling behaviour of whey protein solution, (b) Heater surface and bulk solution temperature difference during the 90 min fouling
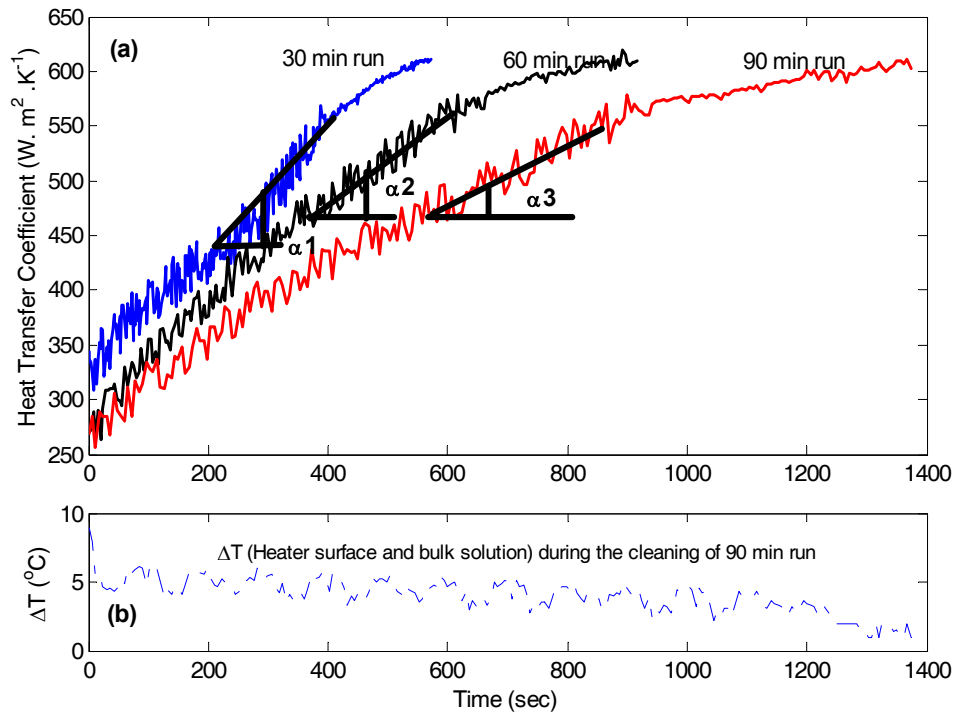

Figure 8. (a) Cleaning-in-place (CIP) behaviour of whey protein concentrates (WPC) solution fouled at various run times; α1, α2 & α3 show the significant slopes towards the finish of the cleaning run. (b) Heater surface and bulk solution temperature difference profile during the cleaning of the 90 min fouling