Direct Ohmic Heating and Fouling of Milk

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Abstrakt

Přímý ohmický ohřev je jednou z technologií, kterou lze použít k elektrickému ohřevu potravin. Tepelná energie v potravině vzniká při průchodu elektrického proudu potravinou v důsledku vlastního elektrického odporu ohřívané potraviny. Výhodami tohoto způsobu ohřevu v porovnání s technologiemi nepřímého ohřevu přes teplosměnné plochy, je rychlost a rovnoměrnost ohřevu potraviny. Tyto vlastnosti lze vhodně využít v oblasti termického ošetření potravin, kde je hlavním cílem zajistit mikrobiální bezpečnost potraviny a co nejméně narušit její nutriční hodnoty. Další výhodou je snadná regulace a monitoring procesu ohřevu. Uplatnění technologie přímého ohmického ohřevu lze najít v oblasti potravinářských výrob (např. mlékárenství – UHT ošetření mléka), veřejného stravování atp. Na druhou stranu je technologie přímého ohmického ohřevu spojena s určitými problémy, jako je např. tvorba úsad na elektrodách, které brání širší aplikaci této technologie v průmyslu. Výzkum tvorby úsad na elektrodách během přímého ohmického ohřevu se zaměřuje na zodpovězení základní otázky: Jaký je principiální rozdíl vzniku tvorby úsad při klasickém konvektivním ohřevu a přímém ohmickém ohřevu? Další, doposud nezodpovězené otázky se týkají především vlivu materiálu elektrod, tvaru elektrod, vlastností povrchu elektrod a rovněž také vlivu napájecí frekvence a koroze elektrod. Prezentované výsledky ukazují vliv teploty mléka, průtoku mléka, vliv velikosti hustoty elektrického proudu a materiálu elektrody na proces tvorby úsad na elektrodách. Výsledky experimentů ukazují pravděpodobnou spojitost tvorby úsad s problémem elektrochemické koroze elektrod.

Klíčová slova

Direct ohmic heating; fouling; milk; corrosion; UHT processes

1. Introduction

The general target of heat processes like pasteurization and sterilization is to protect food against microbiological changes (high temperature exposition is needed) and at the same time to lower the quality of the food products as little as possible. High temperature heating with very short exposition is required. These conditions can be realized using the UHT sterilization method. Classic convective methods for heating process fluids by using plate heat exchangers are still the most popular methods in the food industry. Direct ohmic heating (and similar microwave heating or inductive heating) is a modern method for performing the UHT sterilization process. The advantages over conventional indirect heating methods are the speed and uniformity of heating, and also easy control of the heating process. However, direct ohmic heating has problems with fouling and corrosion. The technology of ohmic heating is not new, and has been in use for over 100 years – Jones (1897). During the 20th century, ohmic heating was in regular use, but it was abandoned due to faradaic and corrosion effects. At the end of the 20th century this technology was rediscovered, and has become the subject of many research studies – Stirling (1987); Skudder and Biss (1987); Zhang and Fryer (1993); Zareifard et al. (2003); Icier and Ilicali (2005). Research on this technology covers a wide range of applications, including thermal processing of solid foods, liquid foods, solid-liquid food mixtures, meats, vegetables Samaranayake and Sastry (2005), Amatore et al. (1998), Samaranayake et al. (2005) focused on faradaic processes during ohmic heating. The causes of milk fouling were studied by Ayadi et al. (2004). However, some questions have remained open: What is the effect of the material properties and the shape of the electrodes, the surface properties of the electrodes and the effect of power frequency on the process of heating food; What are the causes of fouling on electrodes, and corrosion of electrodes.

2. Materials and methods

A continuous ohmic heater with a rectangular cross-section was used for the fouling experiments. Two sides of the rectangular channel acted as electrodes. The electrodes were made of stainless steel, stainless steel with a TiN coating and graphite (POCO EDM-1, Poco Graphite Inc., USA). The ohmic heater (and electrodes) was 220 mm in length and 30 mm in width, and the distance between the electrodes was constant at 10 mm. The surface of the electrodes was electrically insulated, except for a small rectangular area in the center of the heater. The active surface area was quite small (0.0012 m^2), so a high current density (about 2500 $A \cdot m^{-2}$) could be achieved using a small laboratory power source. Skimmed milk was used as the process solution. The solution (5 liters of milk) was kept in a tempered tank and was pumped to the ohmic heater. The flow rate of the process fluid could be adjusted with the use of a gate valve. After heating, the milk flowed through a flow-meter back to the tempered tank. Fig.1 shows the scheme of the experimental setup, and Fig.2 shows a photograph of the ohmic heater. A computer controlled data acquisition system was used to record the experimental data. The inlet and outlet temperature, the temperature of the electrodes and the temperature of the solution in the tempered tank were measured using Pt100 sensors. The electric current, voltage and power were measured using an LMG 95 electronic wattmeter (ZES Electronic GmbH, Germany). The evolution of the fouling was monitored by observing the decrease in the electric current passing through the ohmic heater due to the formation of fouling deposits on the electrode surfaces. The experiments were carried out with three types of electrodes (stainless steel electrodes, stainless steel electrodes with a TiN coating, and graphite electrodes). There were three different flow rates of the process fluid and three different current densities. A constant electric current (3 A, 50 Hz) was set at the beginning of all experiments. The duration of the experiment was limited by the decrease in electric current below 0.8 A, when a smell of burnt milk emerged and the surface of the fouling layer turned brown (the maximum duration of the experiments was set to 10800 s).

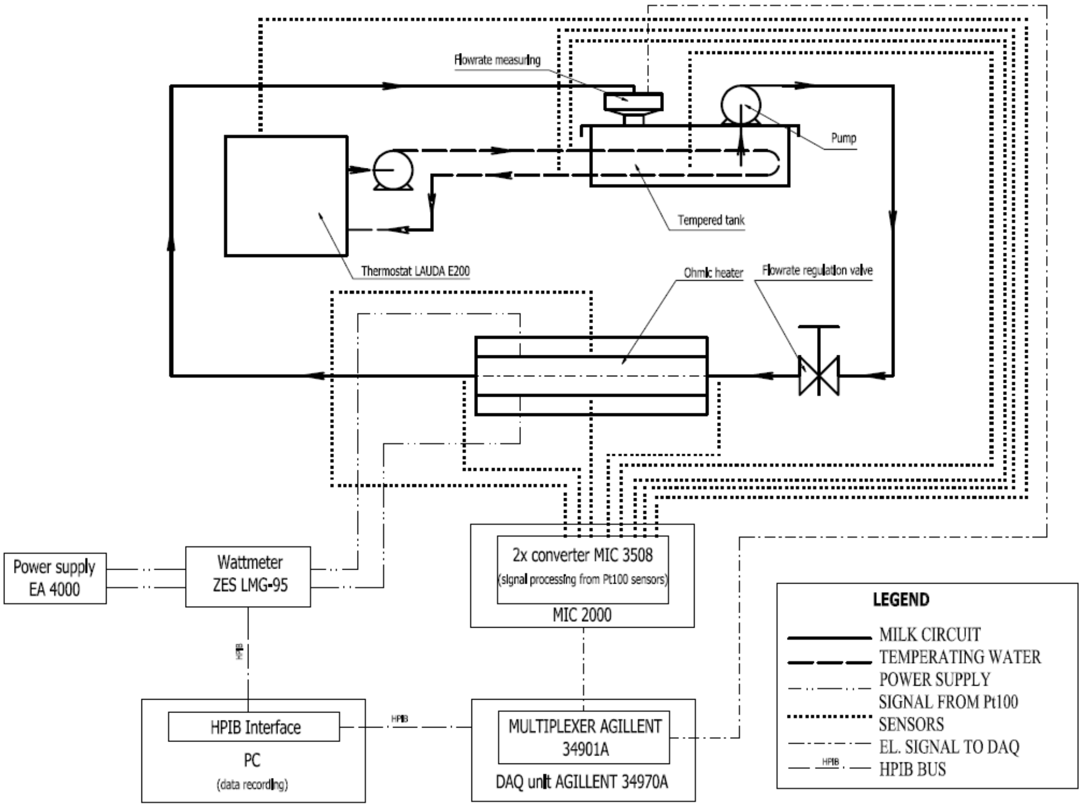

Fig. 1 *Experimental setup*

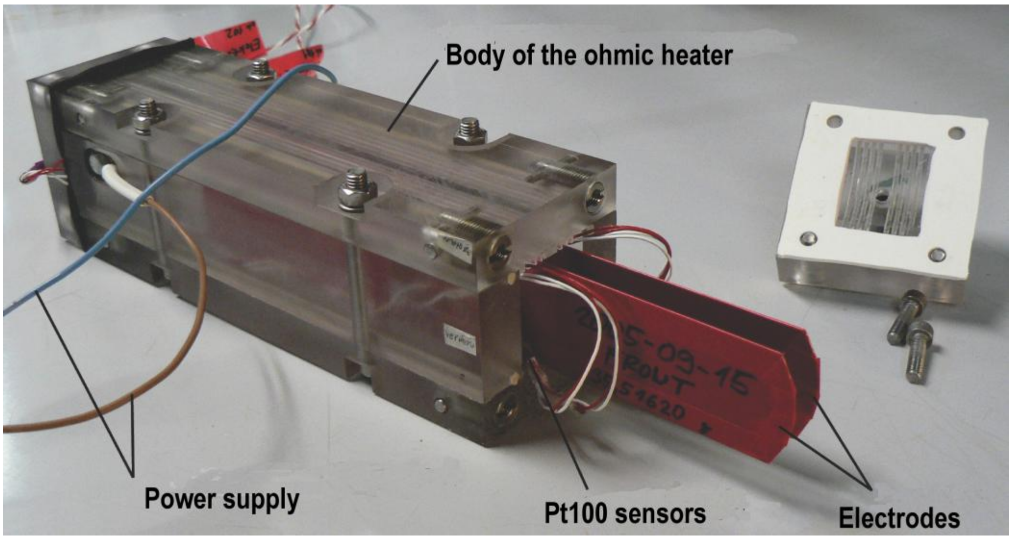

Fig. 2 *Ohmic heater photography*

Each fouling experiment was repeated three times, and thus the accuracy of the results can be estimated: the standard deviation of the recorded lag-time was 10 % (repeated experiments with stainless steel electrodes), 18 % (for TiN coated electrodes) and 7 % (graphite electrodes). The duration of each experiment was about 3 hours, and all 27 experiments with stainless steel electrodes, 27 experiments with TiN coated electrodes and 10 experiments with graphite electrodes were carried out with new pairs of electrodes. Only one tracked parameter (flow rate, inlet temperature or current density) was changed, and the two other parameters were kept constant during the experiment.

3. Results and discussion

A typical time course of the power during ohmic heating is shown in Fig.3, and the corresponding lag-times are summarized in Tab.1.

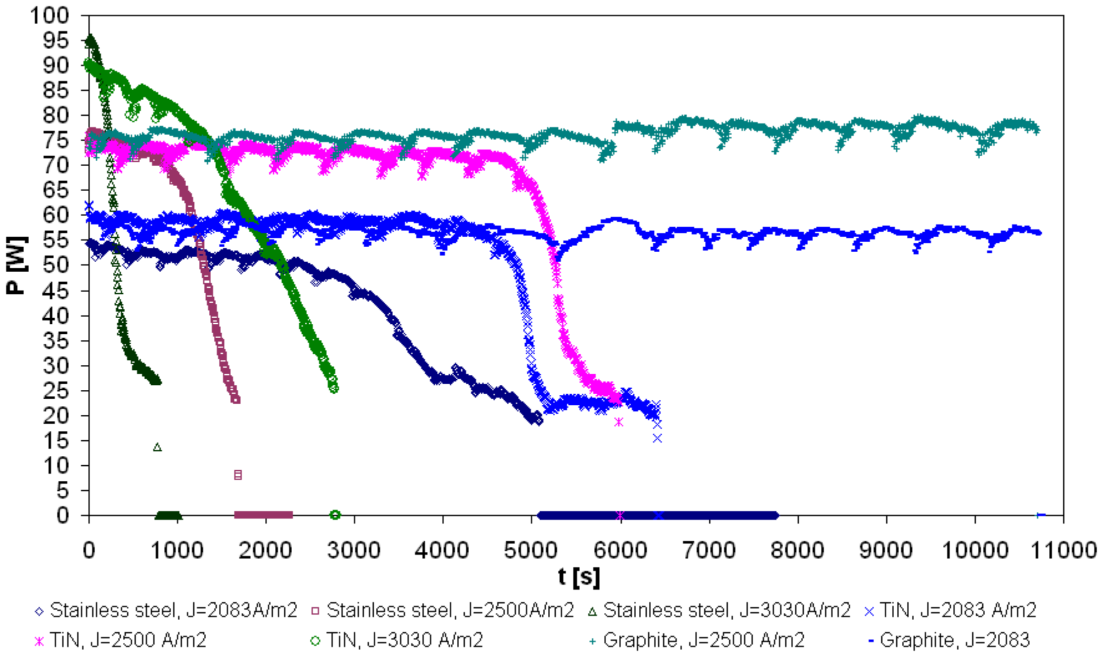

Fig. 3 *Dependence of current density and the material of the electrodes on milk fouling during direct ohmic heating (arithmetic mean from 3 measurements)*

Tab. 1 *Lag-phase duration (lag time) for 3 types of the electrodes in dependence on current density*

	t_{lg} [s] stainless steel electrode	t_{lg} [s] TiN coated electrode	t_{lg} [s] graphite electrode
J=2000 A·m^{-2}	2860	4650	10800
J=2500 A·m^{-2}	1340	4770	10800
J=3030 A·m^{-2}	165	820	

The first part of the course shows approximately constant power (latent phase or lag-phase), while in the second part the power was falling rapidly (falling phase, i.e., a phase of increasing fouling). The latent phase and also the falling phase are affected by operational parameters like flow rate or inlet temperature, by the material of the electrodes, and by current density. Experiments confirmed a relatively strong effect of the electrode material and a significant effect of current density (Fig. 3). The most rapid drop in power was recorded with electrodes made of stainless steel material, especially when high current density values were used. While stainless steel was the worst case, the best results were obtained using graphite electrodes. These results show that during ohmic heating of milk, electrochemical effects and corrosion of the electrodes are of the same importance as denaturation of the whey proteins. The effect of the flowrate is shown in Fig. 4.

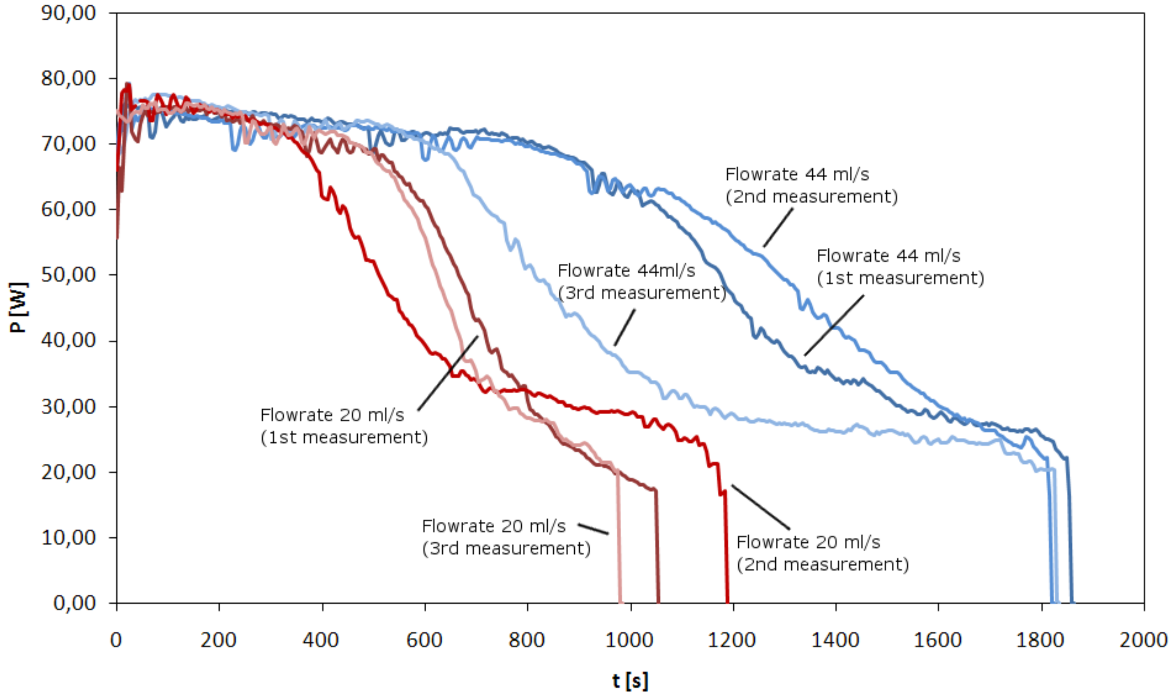

Fig. 4 *Dependence of flow rate on milk fouling during direct ohmic heating*

A higher flow rate extends the latent phase. The effect of the inlet temperature show by using higher inlet temperatures (higher than 75 ˚C) greatly extends the latent phase. This effect may be explained by the denaturation process of the whey proteins in a tempered storage tank. The periodic oscillation of power in the recorded data from the experiments was due to temperature control when the electric heater in the thermostat switched on and off.

4. Mechanism of the origin of fouling during ohmic heating of milk

Although the mechanism and the reactions between all milk components are not yet fully understood, a relationship has been described between the denaturation of native β-lactoglobuline (β-Lg) and the fouling of heat exchangers Jong (1996), Toyoda and Fryer (1994). The rate of the denaturation and agglomeration of proteins depends on the temperature and composition of the milk, and can be described by the following equations – Toyoda and Fryer (1994):

$$\mathrm{N} \rightarrow \mathrm{D} \tag{1}$$

$$2\mathrm{D} \rightarrow \mathrm{A} \tag{2}$$

where N represents native β-lg proteins, D denaturated β-lg proteins and A formation of agglomerates.

$$-r_N = \frac{dC_N}{dt} = k_D C_N \tag{3}$$

$$-r_D = \frac{dC_D}{dt} = k_D C_N - k_A C_D^2 \tag{4}$$

$$-r_A = \frac{dC_A}{dt} = k_A C_D^2 \tag{5}$$

These equations may also be valid for direct ohmic heating, but they alone are not able to explain the observed fouling rates and the relationship between lag phase, electric current density, frequency, and the properties of the electrode surface. Several hypotheses specific for direct ohmic heating can be discussed:

4.1 Singularities of the electric field

Roughness of the surface of the electrode - sharp edges form singularities in the electric field, with the possibility of local overheating. Thus a micro layer of deposits can be formed by thermal denaturation of proteins according to Eqs. (1-5). As soon as this layer exceeds a critical thickness (and a critical electrical resistance), the lag phase ends and the much faster growing phase begins, promoted by the additional heat source from this serially connected additional resistor. However, it seems that this hypothesis is not correct, because our experiments performed with polished and rough stainless steel electrodes do not exhibit very different results.

4.2 Corrosion and mass transport from electrodes to the process solution

Electrochemical reactions in the electrode and electrolyte system (redox reactions and electrochemical corrosion processes) start immediately after the power supply is connected. A primary micro layer can be formed on the electrodes due to oxidation or reduction reactions at relatively high current density values. This layer forms a part of the electric circuit as an additional electric resistance. Because the relative electric resistance value of the primary micro layer is quite high, more electric energy is dissipated into heat in this layer, thus promoting biochemical changes in whey proteins. The reactions of stainless steel material corrosion in an ohmic heater were summarized by Assiry et al. (2006):

1. Electrolytic decomposition of water:

$$\text{Cathode (C):} \quad 2\mathrm{H^+(aq)} + 2\mathrm{e^-} \rightarrow \mathrm{H_2\ (g)} \tag{6}$$

$$\text{Anode (A):} \quad 2\mathrm{H_2O\ (l)} \rightarrow \mathrm{O_2\ (g)} + 4\mathrm{H^+(aq)} + 4\mathrm{e^-} \tag{7}$$

$$\text{Globally:} \quad 2\mathrm{H_2O\ (l)} \rightarrow 2\mathrm{H_2\ (g)} + \mathrm{O_2\ (g)} \tag{8}$$

2. Corrosion of stainless steel electrodes:

$$\text{Cathode (C):}\quad 2H^{+}(aq)+2e^{-} \rightarrow H_2\ (g) \tag{9}$$

$$\text{Anode (A):}\quad Fe(s) \rightarrow Fe^{2+}(aq) + 2e^{-} \tag{10}$$

$$Cr(s) \rightarrow Cr^{3+}(aq) + 3e^{-} \tag{11}$$

$$Ni(s) \rightarrow Ni^{2+}(aq) + 2e^{-} \tag{12}$$

$$Mo(s) \rightarrow Mo^{3+}(aq) + 3e^{-} \tag{13}$$

Metal that is migrating into the medium can be oxidized and can start new secondary reactions. For example, Fe^{2+} or Fe^{3+} can be responsible as catalyzers. This hypothesis corresponds quite well with experiments. It explains the relatively strong effect of the material that the electrodes are made of, and the significant effect of the current density. Fig.5 shows photographs of electrodes made of stainless steel (A), coated stainless steel (B) and graphite (C).

A
Stainless steel electrode

B
TiN coated stainless steel el.

C
Graphite electrode

Fig. 5 *Electrodes after fouling test*

The corrosion effects can be suppressed either by using a noble material like gold or platinum for the electrodes, or by using an increased frequency above 50 Hz. The positive effect of higher frequencies has been known for a long time Samaranayake and Sastry (2005), but it has only recently been studied in detail, above all the effect of the shape of the pulses, see Samaranayake et al. (2005). For this purpose, a new power supply has been developed, based on a full bridge of power MOSFETs driven by a full bridge power MOSFETs controller. This design enables us not only to adjust the feeding frequency, but also to have the optimal delay between pulses that is necessary for the discharge of an electric double-layer at the electrode surface.

4.3 A model of an ohmic heater to describe the processes in the fouling layer

While the fouling layer plays only a passive role as a thermal resistance in classic convective heating, it emerges as an additional volumetric heat source in direct ohmic heating. This means that heat can be generated by current passing through the fouling layer. The fouling layer has an electrical conductivity whose value is lower than the liquid (about 0.007°-°0.03°S·m^{-1} - an experimentally deduced value), and therefore overheating can be expected. The purpose of the following model is to quantify the overheating effect, affected above all by the thickness of the fouling layer, but also by the convective heat transfer in a liquid flowing near the layer. Our model is quite simple, and relies on partial differential equations for steady state electric potential U, temperature T and velocity of liquid u. The velocity field can be described with the use of Navier-Stokes equations (14), the temperature field with the use of the Fourier-Kirchhoff equation (15-liquid, 16-fouling layer), and the electric field can be described by the Laplace equation (17-liquid,18-fouling layer):

$$\rho \frac{\mathrm{D}\vec{u}}{\mathrm{D}t} = -\nabla p + \mu \nabla^2 \vec{u} + \rho \vec{f} \tag{14}$$

$$\rho c_p \vec{u} \cdot \nabla T = \lambda \nabla^2 T + \kappa (\nabla U)^2 \tag{15}$$

$$0 = \lambda_f \nabla^2 T + \kappa_f (\nabla U)^2 \tag{16}$$

$$\nabla \cdot (\kappa \nabla U) = 0 \tag{17}$$

$$\nabla \cdot \left(\kappa_f \nabla U\right) = 0 \tag{18}$$

The system of equations was solved numerically in 2D by the small FEM solver FEMINA, for the following parameters of a wide rectangular heating channel, as shown in Fig.2. The width of the channel (distance of the planar electrodes) = 10 mm, channel length (distance from inlet, where a uniform temperature $T = 70^\circ$C is assumed) = 40 mm, thickness of the electrode = 1.5 mm (material stainless steel, λ =15 W·m^{-1}·K^{-1}), flow rate of the liquid (milk) = 36 ml·s^{-1}, specific electric conductivity of milk κ=1.05 S·m^{-1}, specific electric conductivity of the fouling layer κ_f =0.015 S·m^{-1}, thermal conductivity of milk λ = 0.547 W·m^{-1}·K^{-1}, thermal conductivity of the fouling layer: same as milk. Density of milk ρ = 1036 kg·m^{-3} and specific heat capacity of milk c_p = 3.96 kJ·kg^{-1}·K^{-1}. The thickness of the fouling layer was adjusted to 0 mm, 0.1 mm, 0.5 mm, 0.7 mm and 1 mm. For simulation, constant values of the inlet temperature and fixed voltage between electrodes were set (in reality, there was a voltage increase of about 12% during the experiments). The temperatures profiles for thickness of the fouling layer 0 mm and 0.5 mm are shown in Fig.6 and Fig.7.

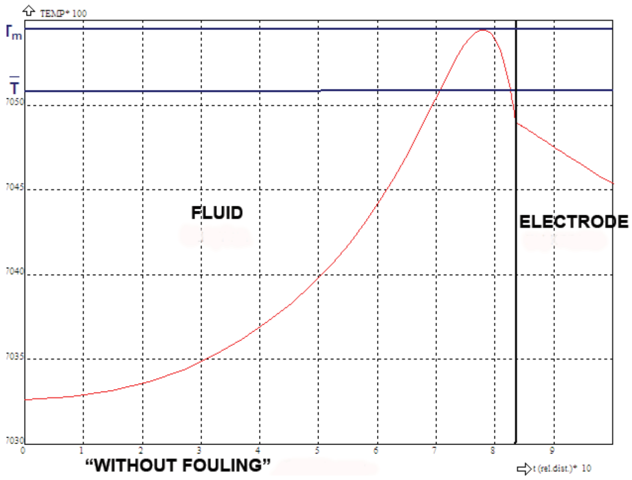

Fig. 6 Temperature profile for a non-fouling situation

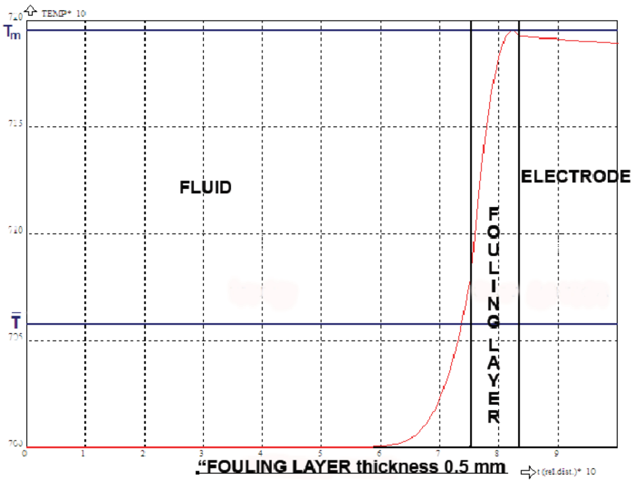

Fig. 7 *Temperature profile for thickness of the fouling layer 0.5~mm*

The horizontal lines T represent the mean calorific temperature of milk. The results and the relative importance of overheating can be expressed in terms of dimensionless parameters, see Fig.8. The ratio between the thickness of the fouling layer and the height of the channel (distance between the electrodes) is given on the x axis and a criterion establishing the ratio of the temperatures when heating with and without a fouling layer (20, 21) is given on the y axis. A graphic visualization is shown in Fig.8.

$$\eta = \frac{h}{H} \tag{19}$$

$$\varepsilon = \frac{T_{max} - \bar{T}}{\bar{T} - T_0} \tag{20}$$

$$\bar{T} = \frac{\kappa \Delta U^2 S}{\bar{u} \rho c_p H} + T_0 \tag{21}$$

It can be seen that the fouling layer can really be overheated during ohmic heating. This overheating is most significant for lower values of the thickness of the fouling layer. The results of the simulation show that overheating in the fouling layer can really constitute the starting mechanism for active fouling.

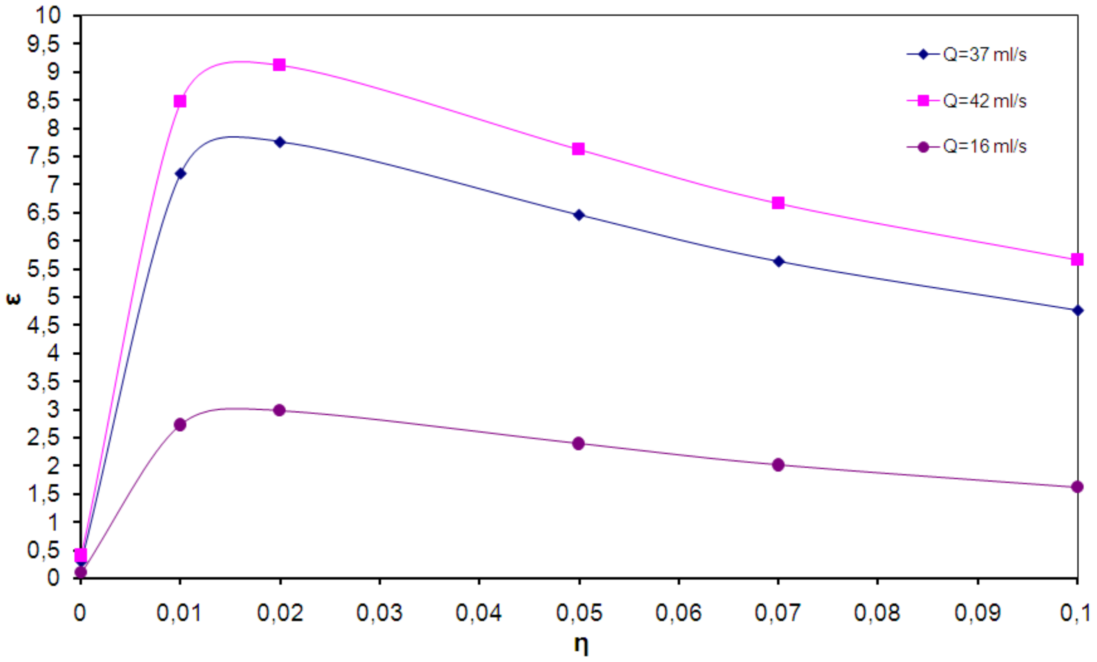

Fig. 8 *Generalized graphic visualization of the mathematical simulation*

5. Conclusions

The results of the experiments show a strong effect of the material of the electrode and a significant effect of current density. The fastest drop in power was recorded with electrodes made of stainless steel material, especially when high current density values were used. Numerical simulation showed that even a relatively thin layer of deposits can be overheated, and can accelerate changes in whey proteins, leading to the final growing phase of fouling. The suggested working hypothesis can be summarized as follows: During the lag-phase, corrosion of the electrodes takes place due to faradaic processes, resulting in an initial microlayer of deposits. This layer presents an inner volumetric source of heat. The microlayer is overheated due to the electric current passing through. The overheating initiates denaturation of the whey proteins in milk and the formation of aggregates, which adhere to the electrodes. This causes the power to drop. To check this hypothesis of the origination of fouling, a new power supply with variable frequencies will be used. Future work will be devoted to a study of the effect of power frequency on faradaic processes and the formation of fouling during direct ohmic heating of foods.

Acknowledgements

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Nomenclature

c_p	Specific heat capacity of milk	$[J.kg^{-1}.K^{-1}]$
C_A	Concentration - aggregated proteins	$[Kg.m^{-3}]$
C_D	Concentration - denaturated proteins	$[Kg.m^{-3}]$
C_N	Concentration - native proteins	$[Kg.m^{-3}]$
h	Thickness of the fouling layer	$[m]$
H	Distance between electrodes (height of the channel)	$[m]$
J	Current density	$[A.m^{-2}]$
k_A	Rate constant for protein reactions - aggregated proteins	$[s^{-1}]$
k_D	Rate constant for protein reactions - denaturated proteins	$[s^{-1}]$
P	Pressure	$[Pa]$
P	Electric power	$[W]$
r_A	Reaction rate - aggregated proteins	$[kg.m^{-3}.s^{-1}]$
r_D	Reaction rate - denaturated proteins	$[kg.m^{-3}.s^{-1}]$
r_N	Reaction rate - native proteins	$[kg.m^{-3}.s^{-1}]$
S	Area	$[m^2]$
t	Process time	$[s]$
t_{lg}	lag-time (lag phase duration)	$[s]$
T	Temperature	$[^{\circ}C]$
T_0	Temperature on inlet	$[^{\circ}C]$
T, □	Mean calorific temperature	$[^{\circ}C]$
u	Velocity of the fluid	$[m.s^{-1}]$
U	Voltage	$[V]$
ε	Criterion establishing the ratio of temperatures at rating	$[-]$
η	Ratio between thickness of the fouling layer and height of the channel	$[-]$
κ	Specific electric conductivity of milk	$[S.m^{-1}]$
κ_f	Specific electric conductivity of the fouling layer	$[S.m^{-1}]$
λ	Thermal conductivity of milk	$[W.m^{-1}.K^{-1}]$
λ_f	Thermal conductivity of the fouling layer	$[W.m^{-1}.K^{-1}]$
μ	Dynamic viscosity	$[Pa.s]$
ρ	Density of milk	$[kg.m^{-3}]$

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