

MODELLING THE FOULING INDUCTION PERIOD

Mengyan Yang*, Andrew Young, Amir Niyetkaliyev
and Barry Crittenden
Department of Chemical Engineering
University of Bath, Bath, UK, BA2 7AY

*corresponding author: my223@bath.ac.uk

ABSTRACT

A fouling process is often preceded by an induction period in which no significant fouling is observed. In this paper, a simple lumped parameter model based on fractional surface coverage θ has been developed to correlate experimental data in the induction period. The model assumes that active foulant species stick to the surface and gradually cover it, the rate of change of surface coverage $d\theta/dt$ being proportional to the fractional free surface $(1-\theta)$. It is further assumed that the foulant already on the surface acts as a seed, attracting more foulant in a micro-growth manner such that the growth rate is proportional to θ . The two assumptions are combined to obtain the relationship $d\theta/dt = k\theta(1-\theta)$ where k is the lumped rate constant. The fouling layer grows on the covered surface and the fouling rate can be expressed as $\theta R_f'$ where R_f' can be any established fouling rate expression. Experimental data, including the data during induction periods, have been successfully correlated for systems including crude oil, crystallization and whey protein fouling. The physical meanings of the model parameters are discussed. The model supports experimental observations in which shorter induction periods are found with higher surface temperatures.

INTRODUCTION

A fouling process is commonly preceded by an induction period in which no significant fouling is observed. The induction period could be from a few minutes in a laboratory experiment to a month or so in refinery plants. A number of researchers have addressed this phenomenon, including in particular, the factors that can influence the induction period.

For protein fouling, Belmar-Beiny et al. (1993) observed a much shorter induction period for a plate heat exchanger than for a tubular heat exchanger. They considered that this was due to the higher turbulence in the former. Belmar-Beiny and Fryer (1993) analysed the first layer of deposit, which was thought to be formed during the induction period and found that it was made of proteinaceous materials rather than minerals. More recently, Augustin et al. (2007) investigated the effect of surface treatments on protein

fouling and reported the effect of surface temperature on the induction period.

For crystallization fouling, Troup and Richardson (1978) discussed nucleation time which is closely related to the induction period phenomenon. More recently, Geddert et al. (2007) investigated the effects of surface coatings and attempted to correlate the induction with the surface roughness and surface tension but did not reach any quantitative conclusion.

In spite of many observations and investigations of induction phenomena, the induction period has not been studied in a substantially quantitative manner especially for crude oil fouling. The main reason for this can be attributed not only to the complexity of the fouling behaviour in the induction period but also to the fact that no additional experimental information can be gained as the induction period proceeds since it appears from thermal measurements as an apparently steady state operation.

Most fouling models are focused on the fouling rate in the period when the heat transfer resistance is increasing (e.g. Wilson et al., 2005). Few models have addressed the induction period. Vatistas (1987) reported a stochastic model for the induction step in particulate fouling. This model can predict the effect of friction velocity on the induction time but was found to be difficult to use in correlating the experimental data. Malayeri and Müller-Steinhagen (2007) developed a phenomenological model for the prediction of fouling resistance of calcium sulphate solutions during boiling heat transfer, including an empirical estimation of the induction time. However, this model, as they stated, is limited only to the fouling period and it was recommended that one should endeavour to correlate data within the initial period. Neural network techniques can be, and have been, successfully utilized to correlate the data during the entire fouling process (Malayeri and Müller-Steinhagen, 2007; Radhakrishnan et al. 2007). The drawback of this approach is that it is, in essence, a black box technique which does not offer any physical understanding of the process. Fahiminia et al. (2005) derived a relationship between delay time (i.e. induction period) and supersaturation ratio at any given fluid velocity and surface temperature based on classical nucleation theory. They also showed that for any given fluid velocity

and surface temperature, there is an Arrhenius relationship between the reciprocal of the delay time and surface temperature. However, applications of their approach may be limited to crystallization fouling. Moreover, it could be difficult to measure or determine the delay time as there is no sharp boundary between the delay time and the following stage. All of these studies indicate that there is still a significant gap in physical models of the initial phases of fouling processes.

The importance of the induction period should not be underestimated. If a good knowledge existed of why induction periods occurred, then it might be possible to exploit this knowledge and extend the induction period indefinitely. A first step in understanding is to create a simple model which can account for the influence of process parameters on the induction period. In this paper, a relatively simple model is developed to correlate the data in the induction period, considering the related factors in a lumped form. The focus is initially on the effect of surface temperature.

MODEL DEVELOPMENT

Model assumptions and description

The fouling on heat exchanger surfaces may be described in the following manner. Firstly, in the induction, or pre-conditioning period, the active fouling species adhere to the heat transfer surface and gradually cover the surface from a fractional coverage of $\theta = 0$ to the total coverage at $\theta = 1$. Here the surface, to which the fractional coverage θ refers, is defined as an idealized area where the surface temperature is considered to be uniform and is measured by a specified thermocouple. This pre-conditioning layer is very thin, though not necessarily a single molecular layer and so the increase in fouling resistance R_f is negligible.

Secondly, in the fouling period the fouling layer may start to grow immediately on the covered/pre-conditioned surface when it may be assumed that the growth rate is proportional to θ . However, the overall fouling growth is very slow until the surface is considerably covered/pre-conditioned. The overall rate of fouling resistance growth can be expressed as:

$$\frac{dR_f}{dt} = \theta R_f' \tag{1}$$

Here, R_f' can be any form of established fouling rate expression, such as Crittenden et al. (1987), Epstein (1994), Ebert and Panchal (1997), etc. These models describe the fouling rate in a near linear form. Table 1 lists the functions of R_f' used for the fouling rates in all figures presented in this article.

In the early stage of the surface pre-conditioning, active species can be captured and adhere to the surface. Here, it is assumed that the rate of growth of surface coverage is proportional to the available free surface:

$$\frac{d\theta}{dt} \propto (1 - \theta) \tag{2}$$

Meanwhile, the particles that stick to the surface act as seeds, attracting more foulant around them, such that fouling proceeds in a micro-growth manner. The growth rate is assumed to be proportional to the fractional surface coverage θ as in Eq. (3).

Table 1. Functions of R_f' used for the fouling rates in the figures in this paper

R_f' ($\text{Km}^2/(\text{kWhour})$)	Applications and conditions	Figure number
0.011	Crude B, 376°C	Fig. 2
0.019	Crude B, 385°C	Fig. 3
0.024	Crude B, 411°C	Fig. 3
0.0156	Crude A 378°C	Fig. 5
0.0028	Crude A, 369.5°C	Fig. 6
$(2E-5) - (7E-8)*t$	Crystallization	Fig. 7
7.3E-5	Whey protein	Fig. 8
7.4E-5	Whey protein	Fig. 8
8.9E-5	Whey protein	Fig. 8

t : time (hour)

$$\frac{d\theta}{dt} \propto \theta \tag{3}$$

Combining these two proportionalities gives a second order growth rate in fractional surface coverage:

$$\frac{d\theta}{dt} = k\theta(1 - \theta) \tag{4}$$

The constant k is the lumped rate constant which may be assumed to depend on the surface temperature according to the Arrhenius equation:

$$k = k_a e^{-E_i/RT} \tag{5}$$

E_i is the activation energy for the induction/surface preconditioning phenomenon and k_a is the pre-exponential factor. Rearranging Eq. (4) gives:

$$\frac{d\theta}{\theta(1 - \theta)} = k dt \tag{6}$$

This equation can be rearranged for integration:

$$\left(\frac{1}{\theta} + \frac{1}{1 - \theta}\right)d\theta = k dt \tag{7}$$

Integration gives

$$-\ln\left(\frac{1 - \theta}{\theta}\right) = k t - \ln c \tag{8}$$

where $\ln c$ is the constant of integration. Hence,

$$\frac{1 - \theta}{\theta} = c e^{-kt} \quad (9)$$

And,

$$\theta = \frac{1}{1 + c e^{-kt}} \quad (10)$$

The initial surface condition determines θ at $t = 0$. In most cases, θ is finite or very small at $t=0$, which requires c to be large. This can be explained by assuming that a few very active spots are formed instantaneously once the heat transfer surface is contacted with the fluid. However, θ could be large or even equal to unity. In some extreme cases, for instance where the surface is a pre-conditioned 'non-wetting' one, this would result in an infinity value for c . On the other hand, in any cases when t is sufficiently large, θ can approach the value of unity.

The constant c , introduced as the integration constant, may be related to the surface characteristics such as the material, its roughness, charge, wettability etc, as well its history of use and the properties of the fouling fluid. In general, it needs to be large. Therefore, at the beginning of a fouling process, when t is close to zero, θ is approximately equal to $1/c$. This gives the physical meaning to the constant c in that $1/c$ is the initial fractional surface coverage.

It is assumed that the adhesion/attachment step is much faster than any removal step in the induction/pre-conditioning period. Hence, the induction/attachment step is almost irreversible in this period. Consider the following argument. Covering the surface may significantly reduce the system free energy G . Hence, if fouling does occur the irreversibility assumption is understandable. This model of the induction period is universal. In this paper it is applied to crude oil, crystallization, and protein fouling. However, it could be applicable to all other types of fouling as well.

Typical θ profiles and length of the induction period

Figure 1 shows some typical simulated profiles of the surface coverage θ against time t . Typically, at the beginning the rate of increase in θ is small. However, this rate increases with time. At a certain point, when $\theta = 0.5$, the rate of change of θ with time reaches its maximum as does the increase of fouling rate with the time according Eq. (1) when R_f' is taken to be constant (in most cases, this is true, especially in the early stage of fouling). This means that, at $t_{0.5}$, R_f is at a turning point. At this point, the fouling is expected to become noticeable. Accordingly, it is useful to introduce a term $t_{0.5}$ to define the time when θ reaches a value of 0.5. Hence $t_{0.5}$ can be used as a measure of the duration of the induction period. Given now that we are interested in $\theta = 0.5$, we have according to Eq. (10):

$$t_{0.5} = \frac{\ln c}{k} \quad (11)$$

In fact, the surface coverage is very low in the early stages of the induction period as shown in Fig. 1, so that the

impact of deposit growing on top of already existing deposit at a possibly different rate would be negligible.

For illustrative purposes in Fig. 1, c has been taken arbitrarily to be equal to 10,000 as a sufficiently large number. For the solid line when k is equal to 7.0 hour^{-1} , $t_{0.5}$ is equal to 1.32 hours. For the dotted line when k is equal to 4.0 hour^{-1} , $t_{0.5}$ is equal to 2.30 hours. Hence, a lower value of k results in a longer induction period. Given the relationship between k and T shown in Eq. (5), k would increase with increasing surface temperature, thereby making the induction period shorter. This prediction is in agreement with reported data (Augustin et al., 2007; Crittenden et al., 1987; Saleh et al., 2005).

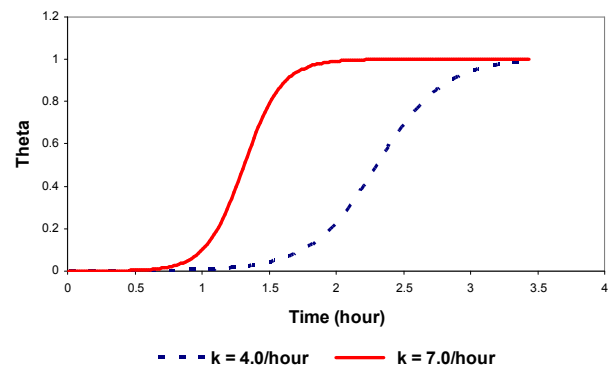


Fig. 1 Typical θ profiles

Saleh et al. (2005) reported the effect of surface temperature as well as the bulk temperature on the length of the induction period. In the present paper, only the effect of surface temperature is discussed. Indeed, the model parameters included to date might suggest a belief that there is only a weak influence of the velocity on the fouling behaviour during the induction period. Published experimental data (Saleh et al. 2005; Tijging et al., 2007) support this suggestion. Their results show only a weak influence of velocity on fouling resistance. However more experimental data may be needed to draw a firmer conclusion.

MODEL APPLICATION AND DISCUSSION

The model is now applied to three systems: crude oil, crystallization, and protein fouling.

Crude oil fouling

The induction period model was applied to the experimental results obtained in the laboratory at Bath using a batch stirred cell system based on Eaton's patent (Eaton, 1983). The system's design and operation are described in other presentations (Young et al., 2009; Yang et al., 2009). All fouling runs were conducted under constant heat flux, constant bulk temperature and constant interfacial temperature. It was found that there was always an induction period prior to measurable fouling if the experiment was started with a fresh heated probe. After the induction period, the fouling resistance was almost always seen to be in a

near-linear form with time. In this case, the overall fouling rate can be simply expressed as θ times the constant rate. The model fit of the fouling resistance was, hence, the integration of the overall fouling rate against time, which was carried out numerically.

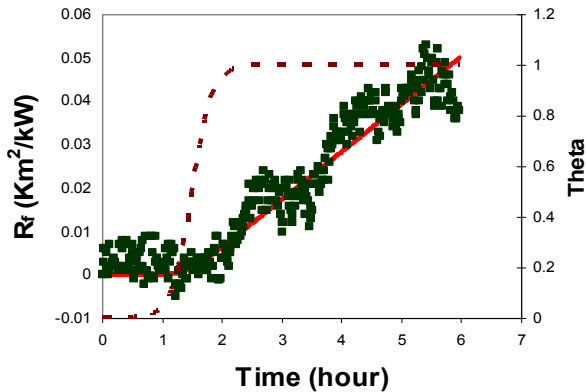


Fig 2. Model application to Crude B with a heated probe with surface temperature of 376°C, and average heat flux of 106 kW/m²

Figure 2 shows the model fit to the data of the experiment of a fouling run of Crude B in the stirred cell system. The surface temperature of the heated probe was 376°C and the batch cell was operated with a stirrer speed of 200 rpm (3.33 Hz) giving a surface shear stress predicted by CFD simulation of 0.52 Pa (Yang et al., 2009). The data points indicate the experimental fouling data whilst the dotted and thick lines represent the θ profile and model fit, respectively. The model constants k and c were determined by curve fitting the data and were found to be 6.03 hour⁻¹ and 8800, respectively. The value of $t_{0.5}$ was found to be equal to 1.51 hours.

Figure 3 shows the model fitted to the experimental data for the same crude oil but at the higher surface temperatures of 385°C and 411°C. The stirrer speed was unchanged and so the surface shear stress was little changed. A comparison of Figs. 2 and 3 reveals, as expected, that the fouling rate increases with increasing surface temperature. For the data fits in Fig. 3, c values obtained by curve fitting are almost the same as for the data shown in Fig. 2. For simplicity therefore, the same value of c (8800) is kept whilst k is allowed to vary. For 385°C, $k = 6.41$ hour⁻¹ and $t_{0.5} = 1.42$ hours. For 411°C, $k = 11.9$ hour⁻¹ and $t_{0.5} = 0.76$ hours.

The error in the R_f measurement could be ± 0.005 Km²/kW, which is mainly caused not only by the heating power and stirring speed fluctuations, but also by the randomly partial removal of the deposit under fluid shear.

Using the k values at the three surface temperatures, an Arrhenius plot based on Eq. 5 is obtained (Fig. 4). The fine linearity of the plot supports the model assumptions. From Fig. 4, the activation energy for the induction period is calculated to be 78.2 kJ/mol. This value is needed in order to predict the effect of surface temperature on k , and thereby to predict the length of the induction period.

In summary, $t_{0.5}$ has been calculated to be 1.51, 1.42, and 0.76 hours at 376, 385, and 411°C, respectively. At these time points, inspection of Figs. 2 and 3 confirms that fouling has become noticeable, thereby demonstrating the value of $t_{0.5}$ as the parameter which provides a practical measure of the length of the induction period.

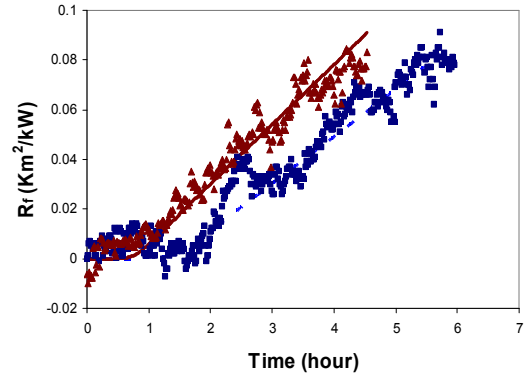


Fig 3. Model application to Crude B crude (■: surface temperature 385°C, average heat flux 110 kW/m²; ▲: Surface temperature 411°C, average heat flux: 116 kW/m²); Lines: model fits.

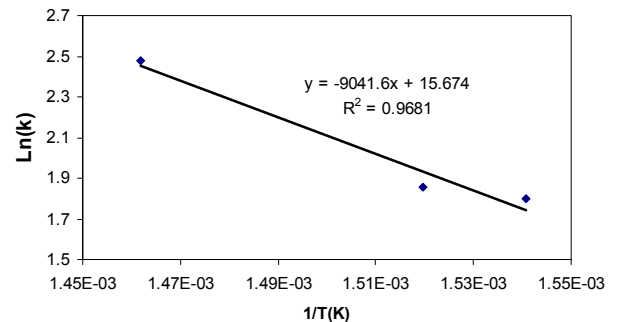


Fig. 4 Temperature dependence of the model constant k

Figure 5 shows the model fit to the experimental fouling data using Crude A. Here, the surface temperature was 378°C and the stirrer speed was 100 rpm (1.67 Hz). With the same value of c as for Crude B (8800), the model provides an excellent fit of the fouling data including the induction period with $k = 5.68$ hours⁻¹ and $t_{0.5} = 1.6$ hours. The value of k is slightly lower than that for the Crude B at a similar surface temperature.

All the fouling data presented in Figs. 2, 3, and 5 were obtained with an initially well-cleaned heated test probe surface. In all these cases, the induction period was clearly observed. In other experimental runs, the probe was not removed from the batch stirred cell for thorough cleaning but was cleaned in-situ. In this case, the authors are doubtful whether the probe surface would have been restored to its perfectly clean condition. In one such case, Fig. 6 shows that an induction period is not observable. The θ profile is shown as the thin line and the model fit to the experimental data is shown as before. The surface temperature was 369.5°C and the stirrer speed was 300 rpm (5 Hz). With a considerable departure in the value of c down to 1.6, the other model

parameters are $k = 5.68 \text{ hour}^{-1}$ and $t_{0.5} = 0.1 \text{ hours}$. The initial surface coverage is calculated to be 0.38.

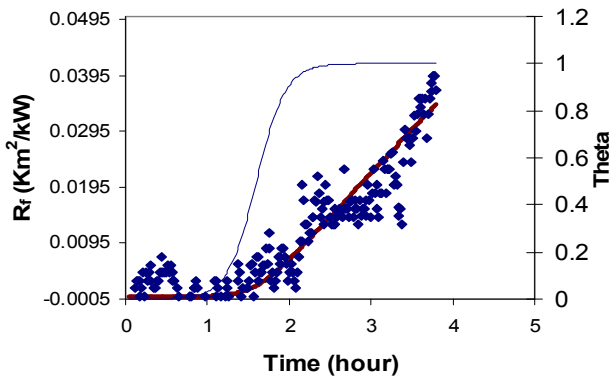


Fig. 5 Model application to Crude A
Surface temperature 378°C , average heat flux 92 kW/m^2
Thin line: θ profile; Thick line: model fit

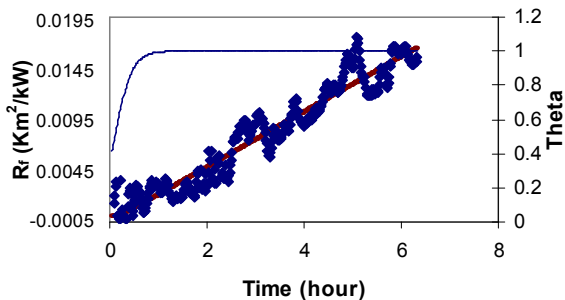


Fig. 6 Model application to Crude A with an imperfectly cleaned probe surface; average heat flux 88 kW/m^2
Thin line: θ profile; Thick line: model fit

Figure 6 demonstrates that the model is capable of fitting fouling data that shows no induction period, or that shows a very short induction period. Interestingly, the best fitting was obtained with the same k value as for the fouling run with a well-cleaned probe but with a significantly reduced value of c . This is as expected since in the same experimental system of probe, crude oil and surface temperature, the value of k should remain constant. However, because the probe surface was not thoroughly cleaned, the initial surface coverage is expected to be much more significant and this is manifested in the much lower value of c .

Crystallization fouling

Geddert et al. (2007) were interested in extending the induction period of crystallization fouling by the application of various surface coatings onto stainless steel. Coatings extended the induction period in calcium sulphate scaling for every flow velocity studied. Indeed, the induction period could be extended by a factor of two for low fluid velocities and by more than twenty for higher velocities. Figure 7 shows the fitting of the present model for Geddert et al's

fouling curve shown as their Figure 2. The model parameters were $c = 14,290$, $k = 0.114 \text{ hour}^{-1}$, and $t_{0.5} = 83.9 \text{ hours}$. The fouling rate was set to be linear with stepwise reduction according to the shape of the fouling curve.

The model has also been fitted successfully to other published inorganic fouling data. As an example, the model has been fitted to the fouling due to deposit formation in the heat exchangers of an industrial sulphuric acid evaporation plant reported by Müller-Steinhagen and Lancefield (2007).

Induction periods in inorganic fouling are, in much the same way as in hydrocarbon fouling, dependent upon many parameters which can be sub-divided into the process conditions and the interface conditions (Geddert et al., 2007). In the present paper, effort has been concentrated initially on temperature.

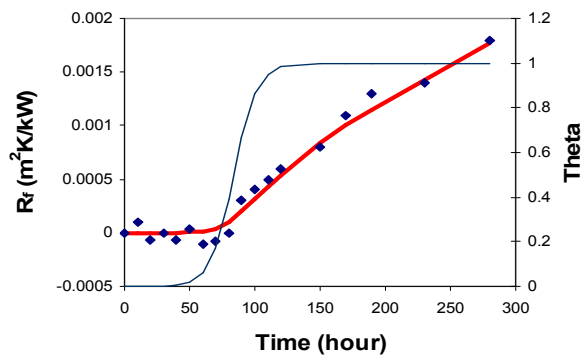


Fig. 7 Model application to the fouling data reported by Geddert et al. (2007)
Thin line: θ profile; Thick line: model fit

Protein fouling

Fouling curves as a function of surface temperature for whey protein fouling as obtained by Augustin et al. (2007) are shown in Fig. 8. The experimental data shows how the surface temperature affects the length of the induction period. The solid lines show, in addition, how well the new model fits the experimental data. The data were obtained directly from the figure in the original paper but were truncated, showing only the data in the early stages, including the induction period and the early linear parts of the fouling curves.

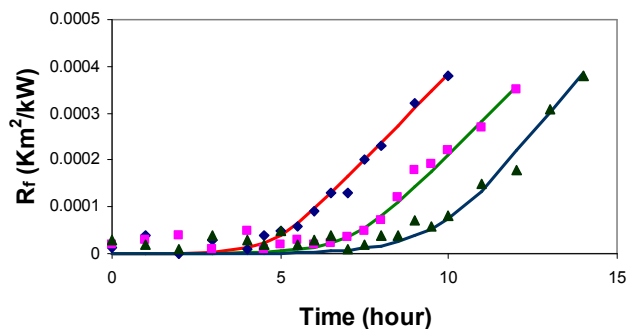


Fig. 8 Model application to whey protein fouling data reported by Augustin et al. (2007)

In Fig. 8, ▲ represents data for a surface temperature of 69.8°C for which the model fitting parameters were $k = 0.86 \text{ hour}^{-1}$, $c = 6466$ and $t_{0.5} = 10.2$ hours. The symbol ■ represents data for a surface temperature 75.7°C, the model parameters being $k = 1.18 \text{ hour}^{-1}$, $c = 6466$, $t_{0.5} = 7.4$ hours whilst the symbol ♦ represents data for a surface temperature of 81.4°C, the model parameters being $k = 1.738 \text{ hour}^{-1}$, $c = 6466$ and $t_{0.5} = 5.05$ hours. The results show that, as for crude oil fouling, the length of the induction period reduces as the surface temperature is increased. In turn, it is found that as the surface temperature is increased, the value of k is increased and the value of $t_{0.5}$ is decreased whilst the value of c is held constant at a high value. Since c is related to the initial surface coverage θ , it would be expected to be independent of temperature, though further investigation is needed to confirm this.

Figure 9 shows the Arrhenius plot for the model constant k as a function of the surface temperature. The apparent activation energy for the induction period of this whey protein fouling system was calculated to be 61.2 kJ/mol. Although there are only three data points, the fine linearity of the Arrhenius plot for the constant k supports the new induction period model.

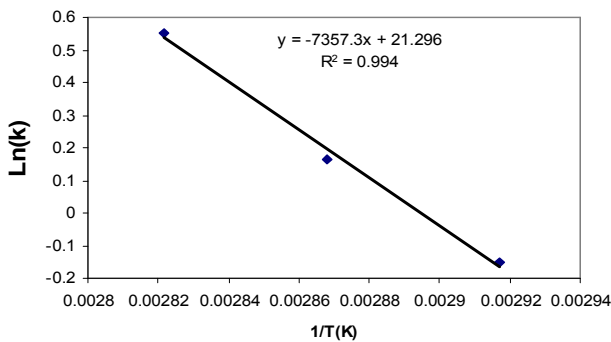


Fig. 9 Temperature dependence of the model constant k for the whey protein fouling reported by Augustin et al. (2007)

CONCLUSIONS

1. The new model makes it possible to describe the fouling process from the start of the induction period up to the constant fouling rate stage using a single and simple mathematical expression. The model has been tested on experimental data from crude oil fouling, calcium sulphate fouling and whey protein fouling.
2. The proposed term $t_{0.5}$ which is the time to reach 50% of initial surface coverage θ provides a practical measure of the length of the induction period.
3. This model quantitatively describes the influence of the surface temperature on the length of the induction period for the crude oil and whey protein fouling systems. Further work is needed to define the physical meaning of the constant k on parameters other than surface temperature.
4. Further work is also needed to define the physical meaning of the constant c on experimental parameters such as surface characteristics, flow velocity, etc. In

addition, the model only addresses fouling as determined from thermal measurements. Hence, there will be a sensitivity limit below which the deposit cannot be detected. Since this complicates the interpretation of velocity effects, further studies on this sensitivity are needed.

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NOMENCLATURE

c	constant of integration
E_i	activation energy, kJ/mol
G	free energy, kJ/mol
k	lumped rate constant, 1/hours
k_a	pre-exponential factor, 1/hours
R	universal gas constant, 8.314 J/mol K
R_f	fouling resistance, $\text{m}^2 \text{ K/kW}$
R_f'	fouling rate, $\text{m}^2 \text{ K/kJ}$
T	absolute surface temperature, K
t	time, hours
$t_{0.5}$	time when $d\theta/dt$ is a maximum
θ	fractional surface coverage

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