Universidade do Estado do Rio de Janeiro
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Analysis of fouling rate of crude oils in the literature: experimental data, models fit and parameter estimation

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Dissertação apresentada, como requisito parcial para obtenção do título de Mestre, ao Programa de Pós-Graduação em Engenharia Química, da Universidade do Estado do Rio de Janeiro. Área de concentração: Processos Químicos, Petróleo e Meio Ambiente

Orientadores: Prof. Dr. André Luis Alberton

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DEDICATÓRIA

Dedico este trabalho a meu marido por todo companheirismo e apoio, principalmente nos momentos mais difíceis!!

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RESUMO

BAPTISTA, Tuanny Rodrigues Coqui. *Analysis of fouling rate of crude oils in the literature:* experimental data, models fit and parameter estimation. 2019. 82 f. Dissertação (Mestrado em Engenharia Química) – Instituto de Química, Universidade do Estado do Rio de Janeiro, Rio de Janeiro, 2019.

Um problema importante em refinarias de petróleo é a resistência à troca térmica, devido à deposição em trocadores de calor para aquecimento de óleo bruto, tipicamente descritos por modelos do tipo Threshold. Esses modelos envolvem parâmetros que devem ser estimados a partir de dados de laboratório ou de refinarias. Com base nisso, algumas questões devem ser feitas: a partir de dados experimentais típicos, é possível determinar todos os parâmetros simultaneamente? Em caso negativo, é necessário manter fixo um conjunto de parâmetros nos quais nenhum erro pode ser atribuído; Se a resposta anterior é positiva, é possível estabelecer quais desses parâmetros ajustam melhor os dados experimentais? Nesse contexto, o objetivo dessa dissertação é: (1) Avaliar a identificabilidade dos parâmetros do modelo do tipo *threshold* utilizando dados experimentais da literatura. Para essa etapa, foi feito um estudo preliminar afim de obter erros experimentais com base em dados fornecidos por alguns autores na literatura; (2) Verificar entre os modelos selecionados se é possível identificar quais modelos são capazes de se ajustar à representação dos dados experimentais. Para a primeira etapa uma análise dos dados da literatura indica que a maioria dos erros de taxa de deposição para um único experimento encontra-se em valores de até 5% e erro relativo. O efeito de compensação aparente entre fator pré-exponencial e energia de ativação aparentemente se confunde com o efeito estatístico na maioria dos casos. A estimação de parâmetros leva a resultados próximos segundo diferentes metodologias de estimação. Na segunda parte, os resultados apresentaram que apesar da maioria dos modelos conseguirem correlacionar a tendência dos valores experimentais e calculados, o erro experimental usado para dados pseudo-experimentais é muito baixo, de forma que poucos modelos conseguiram prever dados dentro dessa precisão.

Palavras-chave: Deposição. Estimação de Parâmetros. Medidas Estatísticas.

ABSTRACT

BAPTISTA, Tuanny Rodrigues Coqui. *Analysis of fouling rate of crude oils in the literature: experimental data, models fit and parameter estimation.* 2019. 82 f. Dissertação (Mestrado em Engenharia Química) – Instituto de Química, Universidade do Estado do Rio de janeiro, Rio de Janeiro, 2019.

An important problem in petroleum refineries is the resistance to thermal exchange, due to the fouling in heat exchangers for heating of crude oil, typically described by threshold models. These models involve parameters that must be estimated from laboratory or refinery data. Based on this, some questions need to be asked: from typical experimental data, is it possible to determinate all the parameters simultaneously? On the contrary is necessary to maintain constant a set of parameters for which no error can be attributed; If the previous answer is positive, is it possible to state which of these parameters best fit the experimental data? In this context, the objective of this dissertation is: (1) To evaluate the identifiability of threshold models parameters making use of experimental data from the literature. For this step, a preliminary study was made in order to obtain experimental errors based on data provided by some authors in the literature; (2) Check among the selected models if it is possible to identify which ones canfit the experimental data. For the first step using the experimental errors obtained in the preliminary study, relative errors of the rate of deposition for a single run are below 5%. The compensation effect between pre-exponential factor and activation energy is confounded with statistical error in the most cases. The parameters estimation methodology lead to similar results. In the second part, the results have shown that, despite most of the models were able to correlate the trend of experimental and calculated values, the experimental error obatined for pseudo-experimental data is quite low. Therefore, few models were able to predict data within this precision.

Keywords: Fouling. Parameter Estimation. Statistic Measures.

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LIST OF ABBREVIATIONS AND ACRONYMS

AIC	Akaike Information Criteria
DOF	Degrees of freedom
EP	Ebert and Panchal model
EPM	Ebert and Panchal modified model
Flu	Fluentes model
GenMod	The model used generated pseudo-experimental data
Ma	Ma model
NEP	Number of experimental points
NG	Nasr and Givi model
Pol	Polley et al. model
Wan	Wang model
Yea	Yeap et al. model

LIST OF SYMBOLS

dRf/dt	Fouling rate, also called r_{net}, m² K/J
Re	Reynolds number, dimensionless
Ea	Activation energy, J/mol
R	Universal gas constant, J/K mol
T	Temperature, K
Rf	Fouling resistance, m² K/W
q	Heat flux, W/m²
v	Velocity, m/s
Pr	Prandtl number, dimensionless
y	Output variable, units vary
p	Parameter vector, dimensionless
Fobj	Objective function, dimensionless
X	Coefficients matrix, dimensionless
DOF	Degrees of freedom, dimensionless
n	Number of experimental data
Vp	Matrix of covariance of parametric uncertainties, dimensionless
A	Entire pre-exponential term of the model, dimensionless
$\bar{T}$	Mean temperature, K
f	Friction factor, dimensionless
Nu	Nusselt number, dimensionless
h	Convective heat transfer coefficient, W/m² K
Vy	Matrix of covariance of experimental uncertainties, dimensionless
B	Sensitivity matrix, dimensionless
χ^2	Statistical measures chi-square
R^2	Coefficient of determination, dimensionless
L	Likelihood function, dimensionless
$\bar{y}$	Vector experimental outputs variables, unit vary
$f(y)$	The true distribution of data, dimensionless
$g(y\|\hat{p})$	Distribution predicted by the mode, dimensionless
$KL_{f,g}$	Kulback-Leibler divergence criterion between such distributions, dimensionless

$E_f[\cdot]$	Expectation regard the distribution function f, dimensionless
$Risk$	Risk function the mean value of $KL_{f,g}$, dimensionless
np	Number of parameters, dimensionless
α	Fouling model parameter, m^2 K/ W h
β	Fouling model parameter, dimensionless
γ	Fouling model parameter, m^2 K/ W h
τ_w	Shear stress, Pa
σ_y^2	The variance of y, dimensionless
σ_y	Standard deviation of variable, units vary
$\sigma_{ln\ A}$	Standard deviation of ln A, dimensionless
σ_p	Standard deviation of parameter, units vary
σ_{Ea}	Standard deviation of Ea, J/mol
ρ	Specific mass, kg/m^3
μ	Dynamic viscosity, Pa s
ϕ	Model probability, dimensionless
ΔAIC^{mod}	The difference between the AIC value of two models, dimensionless
Subscript	
f	Film
s	Probe
c	Crude oil
t	Time interval
t0	time interval zero
w	Fouling surface
Superscripts	
calc	Calculated by model
exp	Experimental data
ref	Reference

SUMMARY

INTRODUCTION

In petroleum refineries, energetic efficiency has immense importance (ASOMANING; PANCHAL; LIAO, 2000), since the energy consumption during the industrial operation is considerable. A considerable fraction of this consumption is associated with the fouling problem in crude preheat trains. Fouling in the crude oil induces an increase on thermal resistance, decreasing the heat exchanger effectiveness, and demanding higher costs with utilities (LEMOS et al., 2015).

In order to predict the behavior of the fouling rate, several semi-empirical models were developed based on the concept of Fouling Threshold, where it is possible to establish an operational region in which fouling is not significant (POLLEY et al., 2002) ("no fouling").

The operation conditions of no fouling can avoid extra costs associated with the reduction of the effectiveness of the heat exchangers. Several works have been dedicated to this subject. The first threshold model was proposed by Ebert-Panchal in 1995 (EBERT; PANCHAL, 1996), presenting the fouling rate in a four parametric model. Based on such proposition, severalmodels have been proposed for describing such phenomena, such as the Polley model (POLLEY et al., 2002).

Nonetheless, one can point out that few works addressed a careful statistical analysis of such models. Most works do not worry about the experimental error and the propagation to model's parameters. Besides, models comparison are scarce in the literature, being the models generally proposed and applied to each set of experimental data carried out by each author.

In this context, the present dissertation has the objective to study the models based on statistical analysis, answering questions as:

- What is the magnitude of the experimental errors in the studies of the literature, and how does it propagate to the estimated parameters of the models?
- Althought the experimentally measured variable is the temperature of the probe, many authors in the literature use fouling resistance or the rate of fouling resistance as experimental data, so three experimental data could be used. Since different experimental data can be used, there may be different ways of estimating from the objective function. Thus, do different forms of parameters estimation regard to objective function lead to similar results of parameter estimation?

- Assuming there are several models proposed in the literature, how is the flexibility of models proposed in the literature for fitting pseudo-experimental data generated with other models?

  In order to answer such questions, the present dissertation is divided as follows:
- Chapter 1 describes the literature review.
- In Chapter 2, it is developed an analysis of experimental data of fouling presented in the literature, evaluating the magnitude of experimental error. A discussion about error propagation is then performed. Besides, taking the Polley model (POLLEY et al., 2002), the estimation considering the temperature, the fouling resistance and the rate of fouling resistance as experimental measurements are compared regard to the estimation of the parameters.
- In Chapter 3, a study about model comparison is performed, generating pseudo-experimental data with each model, and fitting the data to other models. In this study, a total of eight models were considered.
- Finally, it follows the conclusion of the dissertation, together with suggestions and the references.

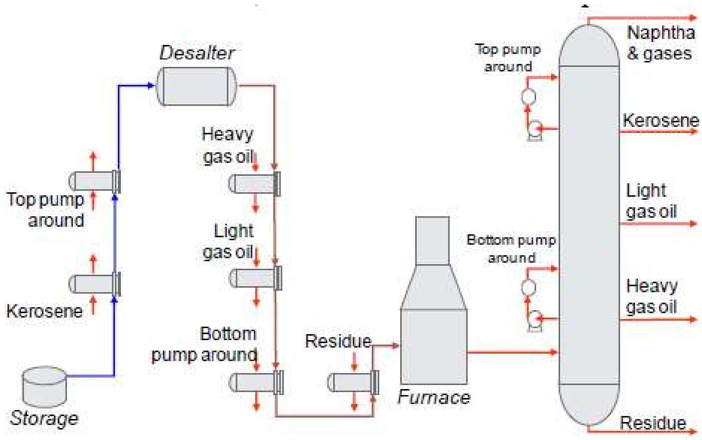
1 LITERATURE REVIEW

This session presents some critical concepts for understanding the objective proposed in this work.

1.1 Fouling in crude oil in pre-heat train heat exchangers

The primary step in the petroleum processing in a refinery is the crude oil distillation. This unit is basically composed of a desalter (which removes inorganic ionic species from the oil) (TAY; KAZARIAN, 2009); a distillation column (which separates the oil into its various fractions) (AKINÇ et al., 2013) a furnace and several heat exchangers that transfer heat to the crude oil stream from the distillation cuts and pumparounds (crude preheat train). Figure 1 schematically shows a typical crude oil distillation unit.

Figure 1 - Typical crude oil distillation unit.



Source: MACCHIETTO et al., 2009.

During the refinery operation, the heat exchangers in the crude preheat train becomes fouled (TAY; KAZARIAN, 2009). The fouling problem may be inorganic , in which the deposits are salts (e.g. FeS); or organic, associated with the presence of asphaltenes (WANG; WATKINSON, 2011).

The most common fouling mechanisms in refineries are: sedimentation, crystallization, chemical reactions and corrosion products (AKINÇ et al., 2013) . The predominance of each fouling mechanism is different depending on the position of the heat exchanger along the crude preheat train.

- Upstream the desalter, the predominant fouling mechanisms are the crystallization of salts and sedimentation due to the presence of impurities (BENNETT et al., 2009);
- Downstream the desalter predominates the fouling by chemical reactions associated with thapresence of asphaltenes that can result in coke (COSTA et al., 2013).

Fouling by a chemical reaction is a complex phenomenon, potentially involving different mechanisms, such as autoxidation, polymerization, cracking and coke formation (AKINÇ et al., 2013). In a furnace, which operates at high temperature, coke formation predominates (RUSSELL; CROZIER; SHARPE, 2010).

Many studies in the literature are performed trying to evaluate the fouling rate in heat exchangers, as discussed below.

1.2 Fouling Threshold Concept

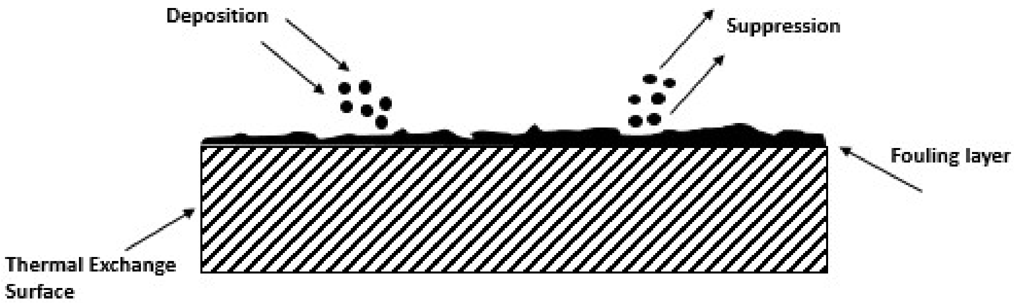
In recent years many studies have been and continue to be made concerning the crude oil fouling in heat exchangers. Significant progress on this subject is that there are models capable of predicting a set of operational conditions that lead to null or almost insignificant values of fouling rate, known as fouling threshold models (YEAP et al., 2005).

The Threshold models assume that the fouling rate is obtained from the difference between a rate of formation and a rate of removal/suppression of deposits.

There is an open debate in the literature involving the two concepts: removal x suppression. The interpretation of the negative term as a removal considers that the deposit once formed can be removed from the heat exchanger surface. The interpretation of the negative term as suppression consider this term as an inhibition of the formation mechanisms, but if the fouling layer is formed, it cannot be removed. The investigation proposed in this dissertation does not include an analysis about each interpretation would be adequate. Aiming at simplifying the text presentation, the term “removal” will be used all along, but it does not mean an affirmation about what it really occurs in the system (DIAZ-BEJARANO; COLETTI; MACCHIETTO, 2017).

Figure 2 shows schematically the deposition process.

Figure 2 - Schematic fouling mechanisms.



Source: Adapted from Awad (AWAD, 2011).

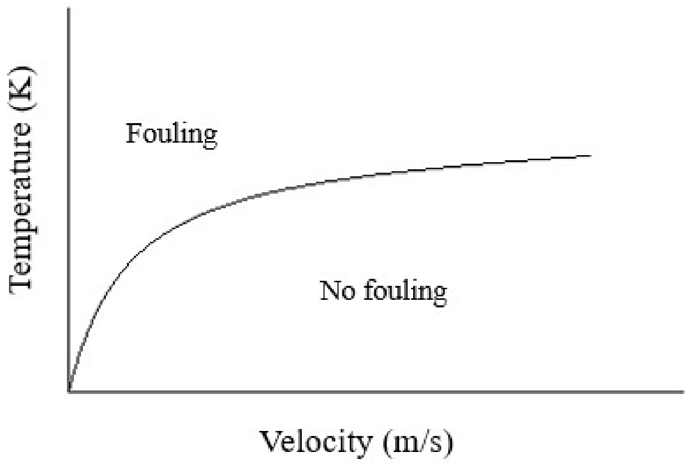
Ebert and Panchal were the first to develop one of the models capable of predicting threshold conditions that would minimize or eliminate the crude oil fouling on tube side of heat exchangers (WILSON; POLLEY; PUGH, 2005). In this model, as shown in Equation (1), the formation term is related to chemical reaction and is dependent on the temperature of the fluid and the Reynolds number, while the removal term is dependent on the shear stress at the surface of the tube (JAFARI NASR; MAJIDI GIVI, 2006b).

$$\frac{dR_f}{dt} = \underbrace{\alpha\, Re^{\beta}\, e^{\left(\frac{-E_a}{R\,T_f}\right)}}_{Deposition} - \underbrace{\gamma\, \tau_w}_{Removal} \tag{1}$$

This formulation allows predicting that if the first term is dominant deposition will occur on the thermal exchange surface; if the terms are in equilibrium there will be no deposition as well as when the removal term is dominant, resulting in what some call the negative fouling rate (YANG; O'MEARA; CRITTENDEN, 2011).

Figure 3 schematically represents the threshold condirions that delimits the existence of a fouling region and no fouling region in a *fouling envelop*.

Figure 3 – Fouling envelop.



Source: Costa et al., 2011.

After the emergence of the model proposed by Ebert and Panchal several other models appeared always involving the two competing mechanisms. Some of the other models found in the literature will be mentioned later.

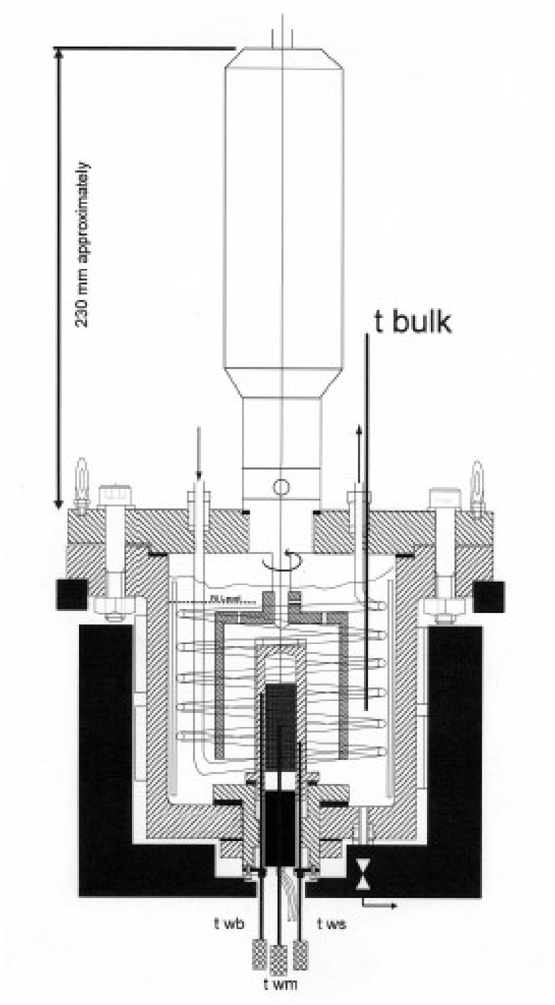
1.3 Experimental apparatus – variable measurement and calculations

Crude oil fouling is a widespread problem in petroleum refineries, which promotes a great interest in studying this phenomenon. However, the investigation of fouling using industrial data involves several obstacles that make the process difficult. With this in mind, several researchers have developed procedures capable of experimentally representing how fouling occurs.

Many authors mention the procedure used by Young et al.(YOUNG et al., 2011), due to the flexibility of modifications throughout the process, such as oil change, gas spraying and the ability to alter and analyze the thermal exchange surface (YANG; O'MEARA; CRITTENDEN, 2011; YOUNG et al., 2011; YANG et al., 2012, 2013; WANG et al., 2015).

The equipment developed by Young et al., as shown in Figure 4, consists of the recirculation of crude oil by a stirred cell that operates under conditions of temperature and pressure close to those found in the crude preheat trains. This cell is pressurized with nitrogen to keep the system pressure controlled.

Figure 4 – Experimental apparatus developed by Young et al.



Source: YOUNG et al., 2011

At the base of the cell there is an electrically heated internal probe that controls the flow. Around this probe there are thermocouples capable of measuring the thermocouple temperature. The heat flow, the crude oil velocity and the oil temperature are controlled variables. This information and more details about the experiment are described by Young et al. (2011).

Other experimental apparatus employs a cylindrical probe and the crude oil flow is parallel to the heated surface. The investigation explored in this dissertation focus on data obtained using this kind of system. A more detailed description is provided in Chapter 2.

The presence of thermocouples in the experimental system allows measuring the temperature of the probe. However there is no way to measure the thermal resistance and fouling rate directly in an experiment. Therefore thermal resistance is calculated from the temperature measured over time, according to the equation:

$$R_f = \left(\frac{T_s - T_c}{q}\right)_t - \left(\frac{T_s - T_c}{q}\right)_0 \qquad (2)$$

As the heat flux and crude oil temperature are kept constant, the equation can be simplified to:

$$R_f = \left(\frac{T_{st} - T_{s0}}{q}\right) \qquad (3)$$

where T_{st} and T_{s0} represent the temperature of the probe in time interval t and zero respectively.

Most authors in the literature describe a procedure similar to Young et al. (2011), which basically consists of recirculating crude oil in a tank through an electrically heated probe (ASOMANING; PANCHAL; LIAO, 2000; SALEH; SHEIKHOLESLAMI; WATKINSON, 2005a, 2005b; SRINIVASAN; WATKINSON, 2005; WATKINSON, 2007; BENNETT et al., 2009; HONG; WATKINSON, 2009). Coupled to the probe are thermocouples capable of controlling a manipulator as system temperatures. A variable measured during the experiment is the temperature of the probe, with which it is possible to obtain the thermal resistance of the deposition, as shown in Equation (3).

1.4 Parameter estimation

Studies on the parameters estimation, more precisely in models of crude oil fouling rate, are rarely found in the literature. The most used method of statistical analysis among researchers is based on the fit of the experimental data, through a linear model.

However, Diaz-Bejarano and Coletti (2015) in their studies regarding the modeling of a shell-and-tube heat exchanger, uses as a method to analyze the parameter estimation for the fouling model, the principle of maximum likelihood. This method consists of finding the values of the parametric and experimental uncertainties that can minimize the likelihood function, thus increasing the probability that this model will be able to predict the experimental data (COLETTI; MACCHIETTO, 2011).

In general, all authors who approach the statistical analysis of predicted and calculated data in some way use temperature as an experimentally measured variable.

1.5 The present work concerning the literature

It is worth remembering that the fouling in heat exchangers causes them to lose effectiveness, which implies in a substantial economic impact, thus making it enjoyable to evaluate the fouling rate in this equipment to support investigations related to cleaning optimization, for example. For this, fouling thresholds models are used.

According to this context, this work seeks to contribute to the literature with a parametric analysis of crude oil fouling rate models. The papers in the literature that obtained experimental data of crude oil fouling and employed parameter estimation to fit these data to fouling rate models did not include any statistical analysis, resulting in a gap in the literature.

The following chapters show that there are several threshold models capable of predicting the fouling rate in heat exchangers. Thus, another contribution to the literature through this work is the analysis, based on some statistical measures, of which of the presented models can fit the experimental data.

2 EXPERIMENTAL ERROR AND PARAMETRIC ANALYSIS OF POLLEY MODEL

Abstract

In this chapter, it is presented how data are collected a typical experimental apparatus. Data reported in papers were obtained and evaluated regard to their uncertainties. A discussion about different approaches for parameter estimation is presented, and the results are illustrated for the Polley model.

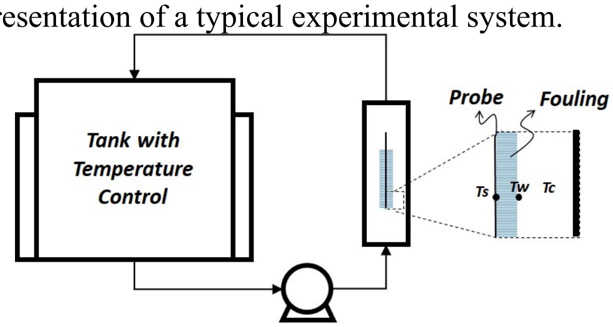
2.1 Experimental data and parameter estimation fouling rate models

Briefly, it is presented how data are collected, besides the model of Polley et al. for describing the fouling rate with operational variables. The Polley model was arbitrarily chosen because it is one of the most used models in the literature.

2.1.1 <u>Typical experimental apparatus and data</u>

A schematic representation of a typical experimental system for collecting fouling data, based on an experimental procedure used by most authors in the literature, as seen in the previous section, is presented in Figure 5. This procedure describe basically the recirculation of crude oil around an annular probe.

Figure 5 – Schematic representation of a typical experimental system.



Source: The author, 2019.

The system illustrated in Figure 5 can control the velocity of crude oil (v), the heat flux (q) and the inlet temperature of crude oil (T_c). Therefore, the temperatures of the probe surface and the interface between the fouling layer and the crude oil stream cannot be controlled, being classified as outlet variables. The system has thermocouples able to measure crude oil temperature (T_c) and the probe surface temperature (T_s), the former considered constant throughout the process (BENNETT et al., 2009).

The experiment is dynamic and the fouling resistance increases with time, however, the hypothesis of the pseudo-stationary approach for heat transfer can be adopted Therefore, one can write:

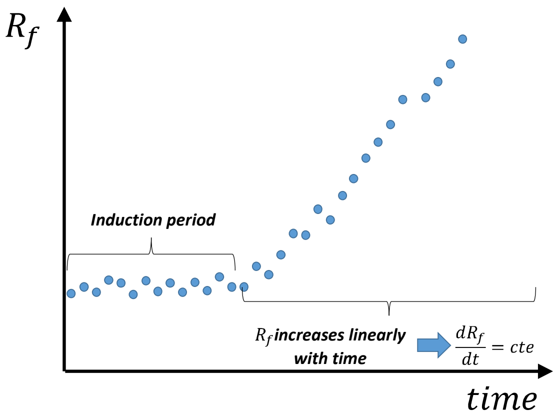
$$q = \frac{1}{R_f + R_0}(T_s - T_c) \qquad (4)$$

where R_0 is the thermal resistance at the beginning of the experiment and R_f is the fouling resistance over the probe. Knowing R_0 from data at the beginning of the experiment, and measuring q and $(T_s - T_c)$, one can obtain “experimental values” of R_f as:

$$R_f = \frac{q}{(T_s - T_c)} + R_0 \qquad (5)$$

The experimental conditions are controlled to keep fixed values of v, q and T_c in order to allow a fixed fouling rate during the experimental run (excluding the induction period). Thus, typically, authors obtain a graph of R_f versus time as illustrated in Figure 6.

Figure 6 – Typical behavior of experimental data reported in the literature



Source: The author, 2019.

The experiment illustrated in Figure 6 allows obtaining one single value of dR_f/dt. The experiment can be carried out at different conditions of fluid velocity, the temperature of crude oil and heat flux.

2.1.2 Polley model for fouling rate

The concept of the fouling threshold started with the studies developed by Ebert and Panchal in 1995. Several models were developed after this and, in 2002, Polley et al. (POLLEY et al., 2002) proposed a model that correlates the formation term with the temperature of fouling surface (instead of to the film temperature) and the removal term with Reynolds number (instead of the shear stress).

$$\frac{dR_f}{dt} = \alpha\ Re^{-0.8}\ Pr^{-0.33}\ e^{\left(\frac{-E_a}{R\,T_w}\right)} - \gamma\ \ Re^{0.8} \qquad (6)$$

The Polley model was employed by several authors (YEAP et al., 2004; WILSON; POLLEY; PUGH, 2005; JAFARI NASR; MAJIDI GIVI, 2006a, 2006b; COSTA et al., 2013; RATEL et al., 2013), in which most of them do not provide a present statistical analysis of data and of estimated parameters in detail. Some authors present only a regression or calculation of deviation concerning the predicted and calculated variable (YEAP et al., 2004; JAFARI NASR; MAJIDI GIVI, 2006a, 2006b), while Costa et al. (COSTA et al., 2013) performed an analysis of parameter estimation. Thus, for such systems, the following questions remain:

- What is the order of magnitude of experimental uncertainties?
- How much experimental uncertainties propagate to uncertainties in the parameters estimated of the model?
- Do the different objective functions, for their respective variables, lead to similar results for estimation?

In order to answer these questions, this dissertation presents a study about experimental data reported in the literature, obtaining typical experimental errors and propagated errors to the model parameters. Besides, considering Polley model, it is performed a discussion about the different forms to perform the parameter estimation, showing that they lead to similar results for typical experimental errors reported in the literature.

2.2 Extracting information of experimental uncertainties from literature data

First, several papers were analyzed and classified about experimental data that could be achieved. The results are presented in Table 20 (Appendix A). It was observed that several works did not present detailed information on experimental data (EBERT; PANCHAL, 1996; POLLEY et al., 2002; BORIES; PATUREAUX, 2003; JOSHI; BRONS, 2003; YEAP et al., 2004, 2005; WILSON; POLLEY; PUGH, 2005; ISHIYAMA; PATERSON; WILSON, 2007; JOSHI; HOEVE; ZIJDEN, 2007; PANCHAL; LJUBICIC, 2007; JOSHI; SHILPI; AGARWAL, 2009; MACCHIETTO et al., 2009; TAY; KAZARIAN, 2009; VENDITTI et al., 2009; WATERS; AKINRADEWO; LAMB, 2009; RUSSELL; CROZIER; SHARPE, 2010; COSTA et al., 2011; FAN et al., 2011; JEGLA; KOHOUTEK; STEHLIK, 2011; SAHIN et al., 2011; WANG; WATKINSON, 2011, 2013; WATKINSON; FAN; PETKOVIC, 2011; RATEL et al., 2013; PETKOVIC; WATKINSON, 2014; CHAMBON et al., 2015; STEPHENSON et al., 2015; WILSON; ISHIYAMA; POLLEY, 2015; CHUNANGAD; CHANG; CASEBOLT, 2017; SMITH et al., 2017). On the other hand, some authors present more detailed information, showing thermal resistance profiles and curves of fouling rate versus temperature (EBERT; PANCHAL, 1996; WATKINSON, 2003, 2007; JAFARI NASR; MAJIDI GIVI, 2006a; POLLEY et al., 2007; WATKINSON; LI, 2009; FAN et al., 2011; YANG; O'MEARA; CRITTENDEN, 2011; YOUNG et al., 2011; YANG et al., 2012). Data reported in papers in graphical form were collected using the software *PegaPonto* © (OLIVEIRA; GAMBETTA; PINTO, 2006). In these cases, linear regression was performed.

It is noteworthy that the data capture of articles from the software used can generate errors, since it is a manual collection. In curver reported with few points or with a clear dispersion of data, this error is minimized because the point to be collected becomes more evident. However, in situations with many points, the error in the collection tends to increase, as the data appears as a point cloud, making it more difficult to know exactly which data to collect.

2.2.1 Linear regression

This topic briefly discusses the linear regression that will be applied to the data provided by the authors to obtain the values of the linear model parameters, as well as the parametric uncertainties.

For a simple linear regression of y versus x, given data $x = [x_1, x_2, .., x_L]$ also, $y = [y_1, y_2, .., y_L]$, where one seeks to fit the following model to obtain the values of the model parameters (p_0 and p_1):

$$y[t,p]^{cal} = p_0 + p_1 \cdot x \tag{7}$$

Admitting the following hypothesis:

h 1. The variance of y is constant and represented by σ_y^2;

h 2. The experimental points are independent.

h 3. The y_i^{exp} is distributed according to a normal distribution function $y_i^{exp} \sim N\left[y[t_i,p]^{cal}, \sigma_y^2\right]$

Then, considering the maximum likelihood approach, the estimated parameter values $p = [p_0 \quad p_1]^T$ can be obtained minimizing the objective function:

$$p = arg\,min\,F_{obj} \tag{8}$$

where:

$$F_{obj} = \frac{\sum_{i=1}^{L}\left(y_i^{exp} - y[t_i,p]^{cal}\right)^2}{\sigma_y^2} \tag{9}$$

The minimization of the objective function leads to:

$$p = (X^T \cdot X)^{-1} \cdot X^T \cdot y^{exp} \tag{10}$$

where:

$$X = \begin{bmatrix} 1 & x_1 \\ 1 & x_2 \\ \dots & \dots \\ 1 & x_L \end{bmatrix} \tag{11}$$

Under hypothesis h1 to h3, the objective function follows a chi-square distribution. The expected value of chi-square distribution is the number of degrees of freedom (DOF), which can be calculated as the number of experimental data less the number of parameters. In this case:

$$DOF = n - 2 \tag{12}$$

since there are two parameters. Then, the experimental error can be estimated taking the expectation of objective function, as follows:

$$\sigma_y^2 = \frac{\sum_{i=1}^{L} \left(y_i^{exp} - y[t_i, p]^{cal}\right)^2}{n - 2} \tag{13}$$

Finally, the covariance matrix of parameters uncertainties can be estimated as:

$$V_p = \sigma_y^2 \cdot (X^T \cdot X)^{-1} \tag{14}$$

The diagonal of V_p correspond to the variance of parameters p_0 and p_1.

2.2.2 Linear regression applied to R_f versus time

In this case, the data set was composed of time $t = [x_1, x_2, .., x_L]$ moreover, experimental fouling resistance (obtained according to Equation (5)) $R_f^{exp} = [R_{f,1}^{exp}, R_{f,2}^{exp}, .., R_{f,L}^{exp}]$, equivalent to Figure 6, after despising the induction period and considering $t = 0$ s when the linear growth of R_f started.

Applying the methodology described in Equations (8) to (14), it was possible to obtain as parameters of the curve R_0 and $\frac{dR_f}{dt}$. Typically, one is only interested in $\frac{dR_f}{dt}$, then, the R_0 was not considered in our future estimation (actually, most authors present only the growth part of the curve in a relative y-axis, not allowing to obtain R_0, i.e., their curves are dislocated to make $R_f = 0$ in the beginning of the curve). Moreover, under the hypothesis that the objective function follows the chi-square distribution, it was possible to estimate the experimental error to Rf (σ_{Rf}), as well as the parametric error of rate deposition ($\sigma_{\frac{dR_f}{dt}}$).Table 1 presents the results for the errors of σ_{Rf}, the value of $\frac{dR_f}{dt}$ obtained in a single experiment from data of R_f versus time (since authors, when present data of R_f versus time, show typically only one graph), and data deviation of $\sigma_{\frac{dR_f}{dt}}$, as well as the relative error of the rate, the approximate number of experimental points (NEP) considered in the curve *Rf* x *t* and the oil used. This analysis becomes interesting because few authors in the literature mention experimental errors over the fouling rate (JAFARI NASR; MAJIDI GIVI, 2006a; BENNETT et al., 2009) (SALEH; SHEIKHOLESLAMI; WATKINSON, 2005a; POLLEY et al., 2007; BENNETT et al., 2009) while only one shows error over the fouling resistance (YANG et al., 2012). It is important to emphasize that all units in Table 1 are in SI.

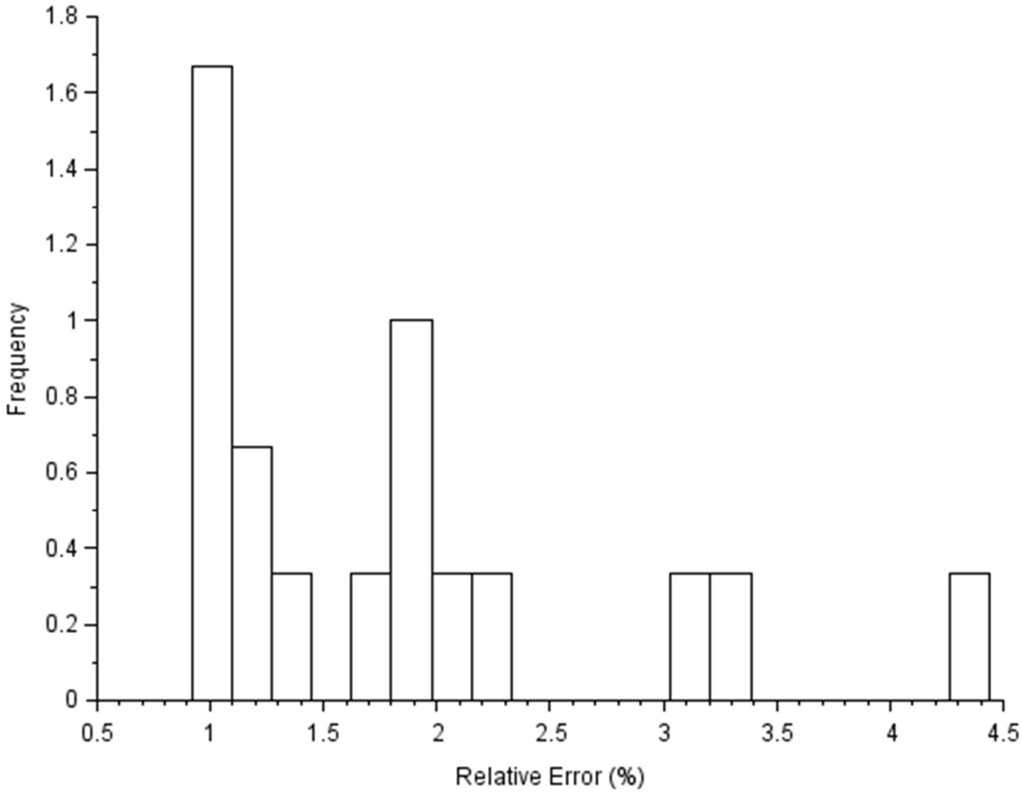
The Table 1 also shows the relative error for the fouling rate found by the authors. This analysis shows that the values are less than 5%, except for the error provided by (WATKINSON, 2003), which presents a high value when compared to the others. This high relative error is because, in this specific case, the *Rf* x *t* curve has few experimental points (only 3), which adjusts with a high error. Figure 7 represents, in the form of a histogram, the frequency of the errors in the data set of Table 1. It is worth noting that the error obtained by (WATKINSON, 2003) was neglected in this case, due to its value dissenting from the others.

Table 1 – Values of d*Rf*/d*t*, standard deviations of d*Rf*/d*t and Rf*.

Author	$\sigma_{R_f} \cdot 10^6$	$\frac{dR_f}{dt} \cdot 10^9$	$\sigma_{\frac{dR_f}{dt}} \cdot 10^{10}$	Relative rate error (%)	NEP	Oil used
(YANG et al., 2012)	2.03	2.5	0.23	0.92	210	uninformed
(HONG; WATKINSON, 2009)	2.67	0.6	0.06	1.00	119	Blend oil (10% cold lake vacuum residue e 90% paraflex)
(BENNETT et al., 2009)	4.09	11	1.15	1.05	55	uninformed
(SRINIVASAN; WATKINSON, 2005)	1.23	0.38	0.04	1.05	53	LSB
(YANG; O'MEARA; CRITTENDEN, 2011)	1.13	1.5	0.16	1.07	183	uninformed
(SALEH; SHEIKHOLESLAMI; WATKINSON, 2005a)	3.57	0.54	0.06	1.11	89	Gippsland (light oil)
(WATKINSON, 2007)	1	0.08	0.01	1.25	68	SSB
(YANG et al., 2013)	1.5	2.9	0.39	1.34	138	Uninformed
(ASOMANING; PANCHAL; LIAO, 2000)	0.87	0.03	0.005	1.67	193	Uninformed
(YOUNG et al., 2011)	3.6	4.6	0.88	1.91	196	Uninformed
(SALEH; SHEIKHOLESLAMI; WATKINSON, 2005a)	3.27	0.31	0.06	1.94	58	Gippsland (light oil)
(WANG et al., 2015)	3.47	4.8	0.94	1.96	83	Uninformed
(WATKINSON, 2007)	1.67	0.1	0.02	2.00	63	HBO
(SALEH; SHEIKHOLESLAMI; WATKINSON, 2005b)	4.6	0.49	0.11	2.24	43	100% Gippsland oil
(SMITH, 2013)	164.67	2.7	0.86	3.19	187	Arab medium 610-1
(POLLEY et al., 2007)	137.67	0.03	0.01	3.33	196	uninformed
(LANE; HARRIS, 2015)	1.1	0.81	0.36	4.44	78	uninformed
(WATKINSON, 2003)	580	84	610	72.62	3	uninformed

[*] All units are in the SI.
Source: The author ,2019.

Figure 7 – Histogram on the frequency of the errors reported in Table 1.



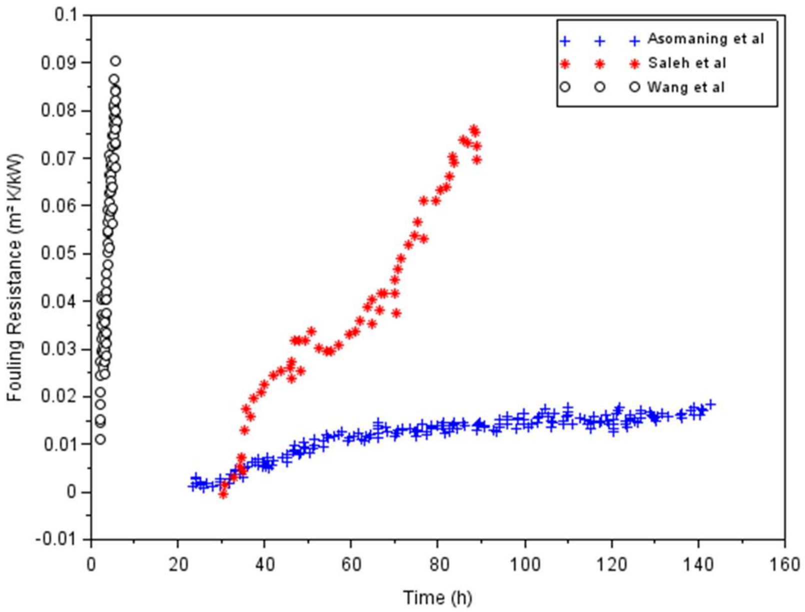
Source: The author, 2019.

The analysis of data from the literature shows that there are very distinct fouling rates of varying magnitude, ranging from $0.03{\cdot}10^{-9}$ to $84{\cdot}10^{-9}$ m² K/J, which means that the rate at which the deposit is formed varies according to the type of oil used, so that some oils foul faster than others (however part of this difference can also be attributed to differences related to flow velocity).

Figure 8 compares curves of *Rf* x *t* for three authors that have the fouling rate in different order of magnitude, showing that the rate at which fouling occurs varies greatly. Possibly this difference in the value of the rates occurs due to the different oils used during the experiment.

As it can be seen in Table 1, typical values of σ_{R_f} are around 10^{-6} m² K/W, however there is some values much higher due to experimental error or few data in the curve. The $\frac{dR_f}{dt}$ obtained from such experiments are in the order of 10^{-9} m² K/J, while its deviation is much lower (around 10^{-11} m² K/J) in the most cases. One can argue that such result is expected since several points of R_f versus time are obtained; thus, the precision of parameters estimated from that curve is very high.

Figure 8 –Comparing papers with different values of Rf.



Source: The author, 2019.

2.2.3 Linear regression applied to dR_f/dt versus 1/T

Some authors do not present curves of the R_f versus time, but presented curves of ln dR_f/dt versus $1/T$ (JAFARI NASR; MAJIDI GIVI, 2006a, 2006b; CRITTENDEN et al., 2009; FAN et al., 2010; BARRIE et al., 2013; MOZDIANFARD; BEHRANVAND, 2015; SMITH; JOSHI, 2015). In these cases, it is not possible to determine the value of the standard deviation of the parameter R_f, however, it can be obtained the propagations of uncertainties for a linear and an angular coefficients of the corresponding model. The input data was considered as the inverse of temperature $1/T = [1/T_1, 1/T_2, .., 1/T_L]$ and the output data $ln\, r_{net}^{exp} = \left[ln\left[dR_{f,1}^{exp}/dt\right], ln\left[dR_{f,2}^{exp}/dt\right], .., ln\left[dR_{f,L}^{exp}/dt\right]\right]$.

Since the graphs are presented as $ln\, r_{net}^{exp}$, from the linear regression one can estimate the experimental error, in this case $\sigma_{ln\, r_{net}}$. Thus, to obtain the error in r_{net} a simple error propagation can be applied as (DROSG, 2007, p. 28):

$$\sigma_{ln\, r_{net}} = \frac{\partial\, \sigma_{ln\, r_{net}}}{\partial\, r_{net}} . \sigma_{r_{net}} \quad \rightarrow \quad \sigma_{ln\, r_{net}} = \frac{1}{r_{net}} . \sigma_{r_{net}} \tag{15}$$

Admitting a typical deviation of $\sigma_{r_{net}}$ obtained to according with the procedure in the previous topic, it is possible to determine the deviation of linear and angular coefficients of the curve of $ln\, r_{net}^{exp}$ versus $1/T$. Since the experimental error is admitted being known, it would be possible to evaluate the model fit; however, since only some experiments were presented in the papers, it was preferred not to perform the model evaluation.

Instead, applying Equations (8) to (14) (except Equation (13), since the experimental error was admitted known from the previous section), it was possible to estimate the uncertainties in the linear and angular coefficients of the curve r_{net}^{exp} versus $1/T$. As the net rate of fouling follows typically an Arrhenius model regard to the temperature for several models, despising $r_{removal}$, one have:

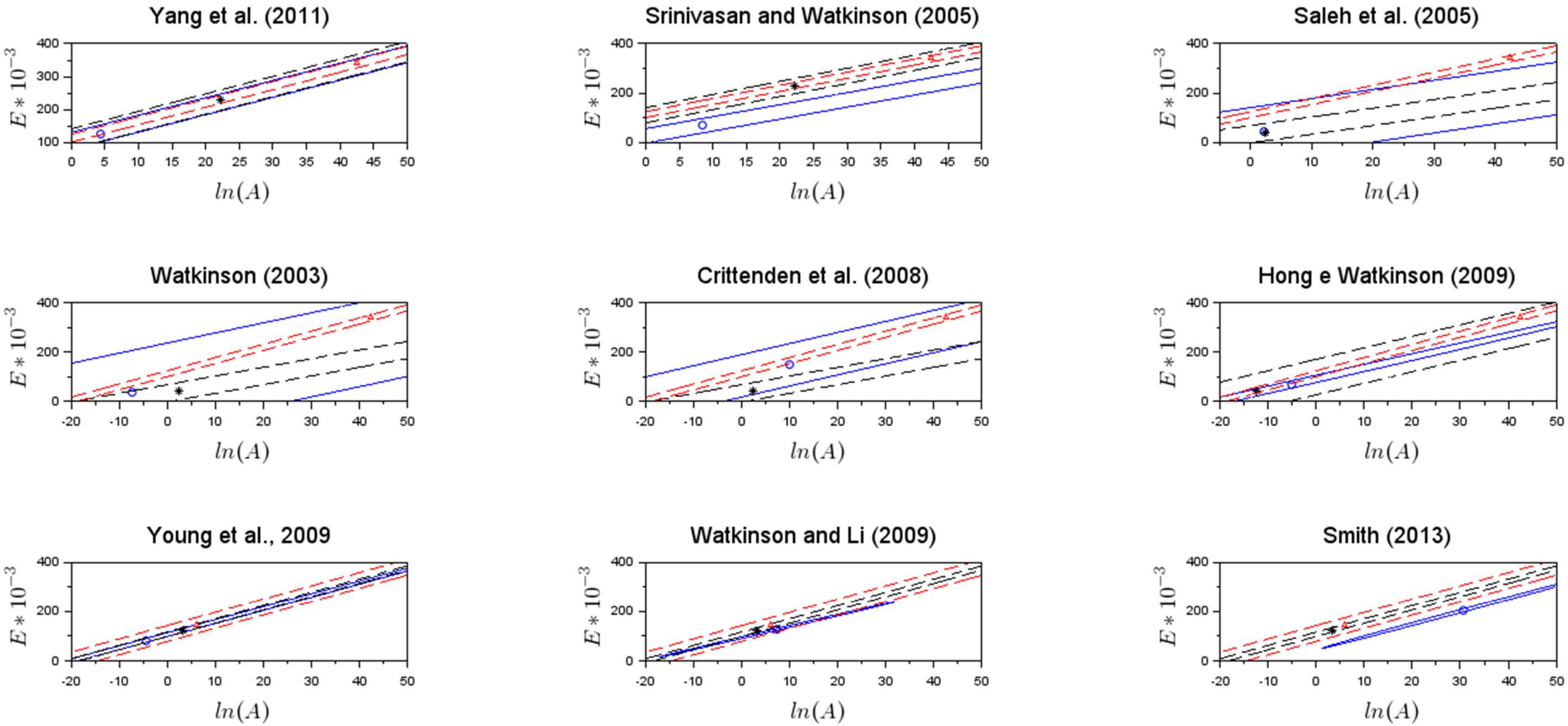
$$r_{net} \approx A \cdot e^{-\frac{Ea}{R \cdot T}} \tag{16}$$

where A is the pre-exponential factor and Ea is the activation energy, the linear coefficient is associated with $ln\, A$ also, the angular coefficient is associated with -Ea/R. Then, after obtaining the parameters p_0 and p_1 of the curve (from to Equation (7)), and their standard deviations σ_{p_0} and σ_{p_1} from the diagonal of the matrix of Equation (14), the standard deviation of $ln\, A$ and Ea can be obtained as:

$$\begin{aligned} \sigma_{ln\, A} &= \sigma_{p_0} \\ \sigma_{Ea} &= R \cdot \sigma_{p_1} \end{aligned} \tag{17}$$

Figure 9 shows values of $ln\, A$ and Ea obtained from works presented in the literature and they respective references.

Figure 9 – Values of Ea and A obtained from linear fit for each author*



Source: The author, 2019.

It must be pointed out that an apparent trend of activation energy and pre-exponential factor cannot be distinguished from statistical error. This effect is known as *compensation effect*, where both $ln\ A$ moreover, E increases, and the effects of rate increase due to $ln\ A$ is partially compensated by the effects of E increase, leading to a small change in the net rate. In fact, such topic has been discussed previously in the literature. Bennett et al. (BENNETT et al., 2009) suggested that as there is a correlation between the pre-exponential factor and the activation energy, the compensation effect allows that the pre-exponential factor can be obtained from the activation energy. However Barrie et al. (FAN et al., 2010) suggested that this effect is statistical, which is caused by random errors measured in a series of experimental for different fluids. This work present additional support evidence that the compensation effect is due only from statistical error, as also discussed by Barrie et al. (FAN et al., 2010).

2.3 Different forms of parameter estimation

This topic is dedicated to the following question: since there are different types of "experimental data", do they lead to similar results in parameter estimation? In other words, which objective function one should use?

$$F_{obj} = \frac{\sum_i \left(R_{f,i}^{exp} - R_{f,i}^{cal}\right)^2}{\sigma_{R_f}^2} \text{ or } \frac{\sum_i \left(T_{s,i}^{exp} - T_{s,i}^{cal}\right)^2}{\sigma_{T_s}^2} \text{ or } \frac{\sum_i \left(r_{net,i}^{exp} - r_{net,i}^{cal}\right)^2}{\sigma_{r_{net}}^2} \qquad (18)$$

Since the original measure is T_s^{exp}, one can argue that this the most proper outlet variable, since the hypothesis that leads to least square objective function is generally associated to measured outlet variable (distributed according to normal distribution function, constant error, and independent variables).

Among the papers that present results of parameter estimation of threshold models, there are authors who use the fouling resistance for parameter estimation (COSTA et al., 2013; MIRSADRAEE; MALAYERI, 2015) or even the fouling rate (CHAMBON et al., 2015; YEAP et al., 2004). Besides these, there are two authors who probably make use of temperature as a form of estimation in their studies (COLETTI; MACCHIETTO, 2011; DIAZ-BEJARANO; COLETTI, 2015).

To answer this question about objective function, first it is presented a discussion based on the models and on the theory of parameter estimation. After an example is presented for different estimation procedures.

2.3.1 Are they expected to be different?

First, one can suppose that outlet variable $T_{s,i}$ follows a normal distribution function with mean $\bar{T}_{s,i}$ also, standard deviation σ_{T_i}, as follows:

$$T_{s,i} \sim N\left(\bar{T}_{s,i}, \sigma_{T_i}\right) \tag{19}$$

From Equation (4), R_f can be isolated as (in $[T_s]$ is the representation of variables that are a function of T_s):

$$R_f[T_s] = R_0[T_s] + \frac{(T_s - T_c)}{q} \tag{20}$$

In the above equation, $R_0[T_s]$ is the resistance of the crude oil in the absence of deposits. Its values have associated with temperature of the probe and the temperature of the crude oil. Nonetheless, such dependence is quite small for these systems if T_s is not expected to vary significantly. Consequently, the relation of $R_f[T_s]$ is practically linearly dependent on T_s. Since a linear transformation of a normally distributed function leads to a new normally distributed function with similar properties; then, the estimation of first and the second objective functions of Equation (5) are expected to lead to similar results.

The other strategy is associated to the r_{net} as “experimental variable” instead of T_s or R_f. In this case, $r_{net}[T_s]$ is a function of T_s. Despite algebraic manipulations, applying Taylor series expansion of $r_{net}[T_s]$ around the mean value of T_s, it is possible to obtain:

$$r_{net}[T_s] \approx r_{net}[\overline{T}_s] + \left.\frac{dfc[T_s]}{dT_s}\right|_{\overline{T}_s} \cdot \sigma_{T_s} + \underbrace{\cdots}_{\substack{disconsidered\ if \\ error\ in\ T_s\ is\ low}} \quad (21)$$

As shown in Equation (21) and as a known statistical result, if the original variable has a small error, then the error propagation to other variables associated to uncertainties in the original variables will also lead to an almost linear dependence between uncertainties (SCHWAAB; PINTO, 2007). Thus, if the error in σ_{T_s} is small enough, then, $r_{net}[T_s]$ can be admitted to also follow a normal distribution. The least-squares would remain valid. The question is: how small is the error in T_s of typical experimental studies and estimation?

2.3.2 Different approaches for estimation of Polley model

2.3.2.1 Procedure for parameter estimation

First, to make a consistent comparison, the errors are considered to be comparable between all forms. Taking the errors reported in the literature for σ_{Rf}, it was possible to obtain errors for $\sigma_{dRf/dt}$ considering some authors that presented graphs of Rf versus time and also propagated such uncertainties to recover the original disturbance in T_s. From Equation (20), an error propagation despising the influence of $R_f[T_s]$ leads to:

$$\sigma_{T_s} = q \cdot \sigma_{R_f} \quad (22)$$

Then, it was considered as experimental deviations eight different possible values of errors, obtained from literature data, as shown in Table 2.

Table 2 – Estimated values of standard deviation from some authors.

Author	σ_{Ts} (K)	$\sigma_{R_f} \cdot 10^6$ (m² K/W)	$\sigma_{\frac{dR_f}{dt}} \cdot 10^{10}$ (m² K/J)
Yang et al. (2011)	0.12	1.13	0.16
Srinivasan and Watkinson (2005)	0.46	1.23	0.04
Saleh et al. (2005a)	1.31	3.27	0.06
Young et al. (2011)	0.37	3.60	0.88
Yang et al. (2012)	0.21	2.03	0.23
Yang et al. (2013)	0.16	1.50	0.39
Wang et al. (2015)	0.37	3.47	0.94
Asomaning et al. (2000)	0.09	0.87	0.005

Source: The author, 2019.

For each experimental error set from the previous table, it was performed parameter estimation where pseudo-experimental data were generated using Polley model. The experiments were generated according to the values presented in Table 3.

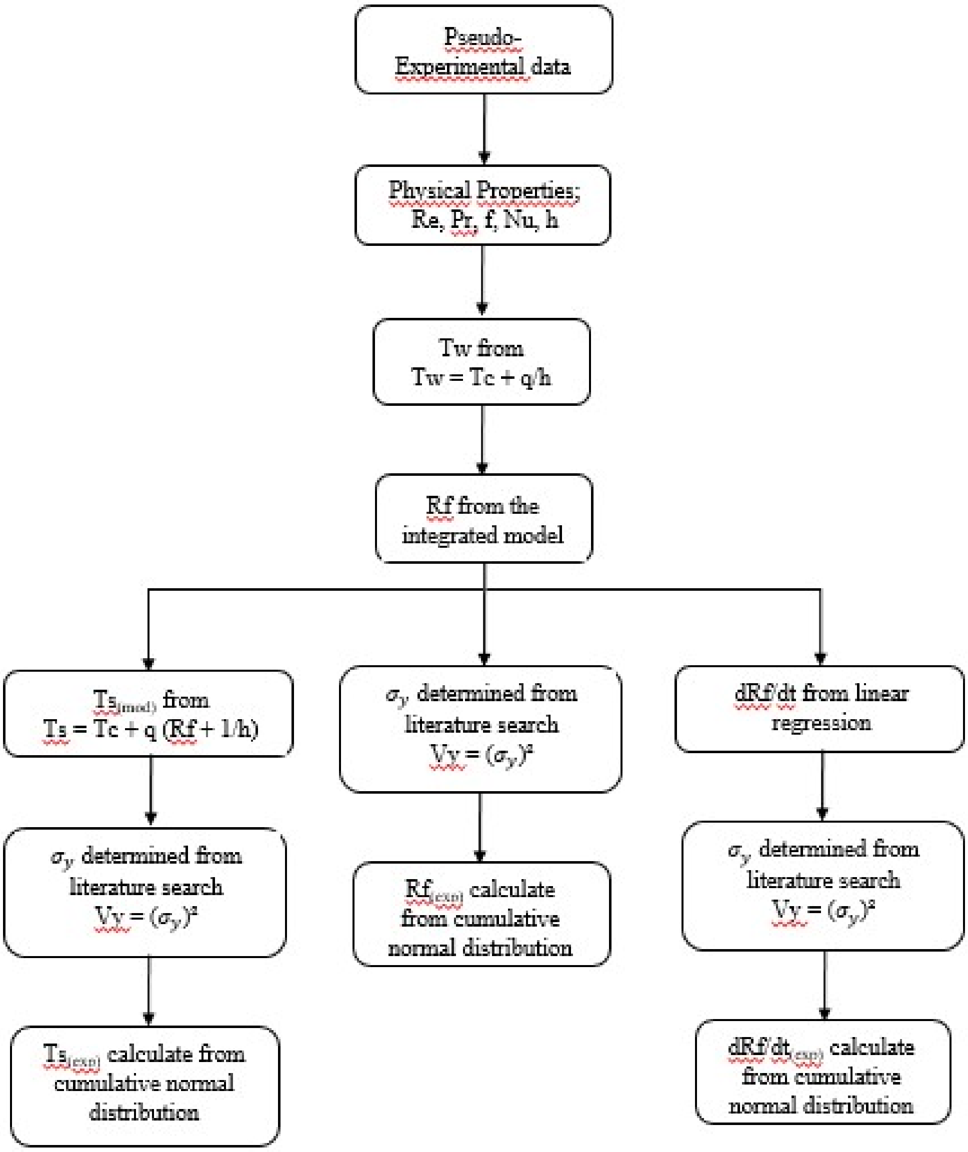
Table 3 – Values used in experimental design.

t (s)	q (W/m²)	v (m/s)	Tc (K)
360	29100	0.5	570
360	64550	0.5	570
360	100000	0.5	570
360	29100	2.75	570
360	64550	2.75	570
360	100000	2.75	570
360	29100	5	570
360	64550	5	570
360	100000	5	570
360	29100	0.5	636.67
360	64550	0.5	636.67
360	100000	0.5	636.67
360	29100	2.75	636.67
360	64550	2.75	636.67
360	100000	2.75	636.67
360	29100	5	636.67
360	64550	5	636.67
360	100000	5	636.67

Source: The author, 2019.

The procedure for generating pseudo-experimental data and parameter estimation is illustrated in Figure 10.

Figure 10 – Schematic representation of the algorithm used to determine the calculated and experimental variables for each case.



Source: The author, 2019.

The model used in this step, as already mentioned, was the Polley model. The three parameters of this model have very different order of magnitude. Therefore, the model admitted as the "true model" (for obtaining pseudo-experimental data) was repaired where its parameters were considered as reference parameters α^{ref} = 1500 m² K/W h, Ea^{ref} = 48000 J/mol, γ^{ref} = 1.5x10^{-9} m² K/W h (POLLEY et al., 2007). The model was re-written in terms of reference parameters, as follows:

$$\alpha = p_0 \cdot \alpha^{ref}, E = p_1 \cdot E^{ref}, \gamma = p_2 \cdot \gamma^{ref}$$

$$\frac{dR_f}{dt} = p_0 \cdot \alpha^{ref}\, Re^{-0.8}\, Pr^{-0,33}\, e^{\left(\frac{-p_1 \cdot E^{ref}}{R\,T_w}\right)} - p_2 \cdot \gamma^{ref}\, Re^{0.8} \tag{23}$$

Then, the parameters $p = [p_0 \quad p_1 \quad p_2]^T$ becomes $p = [1 \quad 1 \quad 1]^T$ for generating the experimental data. Also the estimation was performed in terms of normalized parameters.

Based on the typical experimental conditions in the literature, pseudo-experimental data were generated as shown the Table 3. From these values the physical properties are obtained according to routines presented in the Appendix B, which allows us to determine the calculated Rf from the integrated model equation (Equation (23)). Thus, the calculated deposition rate was obtained from a linear regression, while the probe temperature was obtained from Equation (5).

The variables were then perturbed with a random error with standard deviation mentioned by the authors in the Table 2. Finally, the estimation of parameters was performed, applying each respective objective functions in Equation (18). The routines were written in Scilab © code. The optimization was performed with the package *optmin* of Scilab (which uses the Broyden-Fletcher-Goldfarb-Shanno - BFGS - method), and the calculated values obtained, as illustrated in Figure 10.

The parameters uncertainties were obtained after estimation from the covariance matrix of parameters uncertainties, written for the nonlinear problem as follows:

$$V_p = \sigma_y^2 \cdot (B^T \cdot B)^{-1} \tag{24}$$

where y is the considered variable $\{T_s, R_f, r_{net}\}$ also, B is the sensitivity matrix, where each element i, j is defined as:

$$B_{i,j} = \frac{\partial y_i}{\partial p_j} \tag{25}$$

2.4 Results and Discussion

The results of estimation are presented in Figure 11, Figure 12 and Figure 13. Table 4, Table 5 and Table 6 shown the value of the estimated parameter together with their standard deviation.

Figure 11 presents data of the predicted temperature x calculated temperature for the temperature of the probe obtained from different authors presented in Table 4. It is interesting to observe that the estimation of all parameters simultaneously for some cases presents considerably relevant results, in which the values of the normalized parameters are close to 1, besides presenting small deviations. However, in some situations the value of the parameter α is out of the expected, showing that in these cases the parameter is not as estimated as the other parameters

Figure 12 presents data of predicted thermal resistance x calculated thermal resistance for different deviations, as shown in Table 5. For this case again the parameter α presents a small deviation beyond the expected, meaning that the estimation is not so good compared to the other parameters. .

Figure 11 - Comparison of the calculated and experimental data for the temperature of the probe.

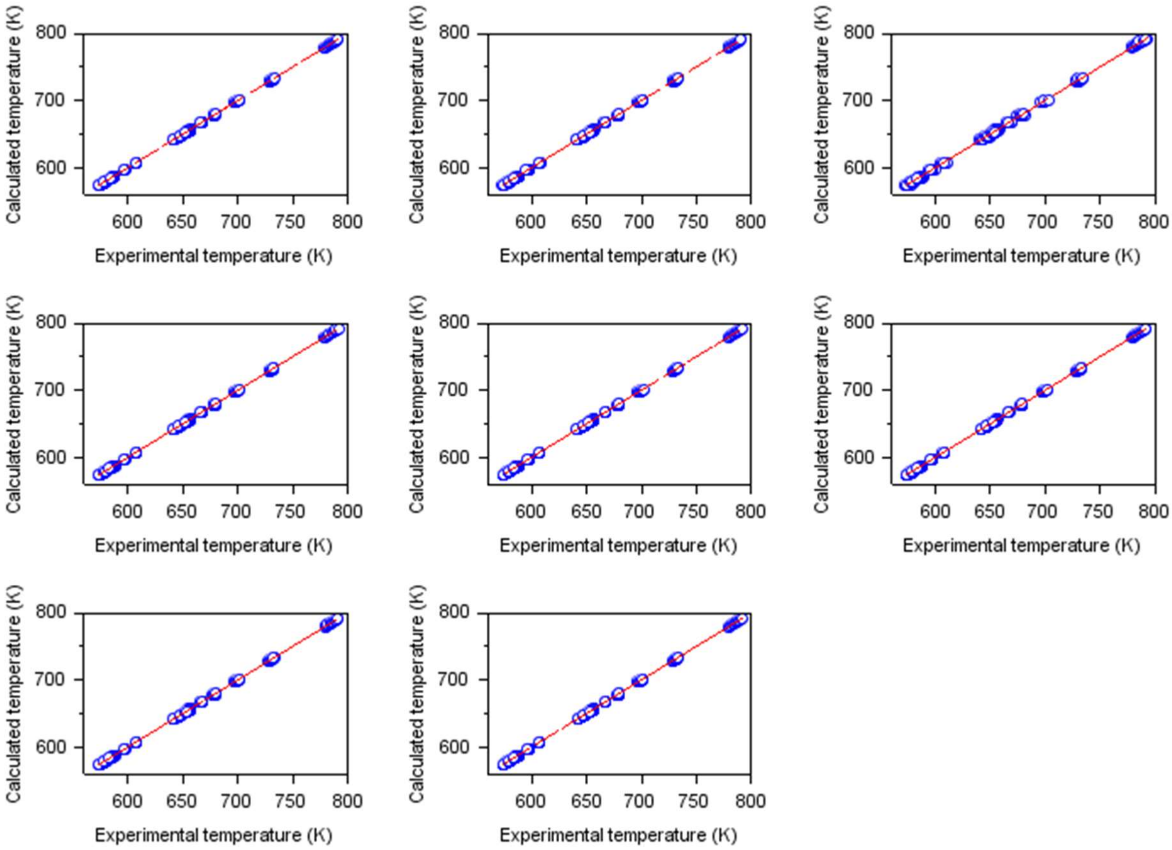


Fig a) Yang et al. Fig b) Srinivasan and Watkinson. Fig c) Saleh et al. Fig d) Young et al. Fig e) Yang et al. Fig f) Yang et al. Fig g) Wang et al. Fig h) Asomaning et al.
Source: The author, 2019.

Table 4 - Values of the estimated parameter and their standard deviation obtained for each author for the temperature of the probe.

	Yang et al. . (2011)		Srinivasan and Watkinson (2005)		Saleh et al. (2005a)	
	parameter	standard deviation	parameter	standard deviation	parameter	standard deviation
p1	1.122	0.074	1.159	0.294	1.017	0.707
p2	1.015	0.009	1.019	0.033	0.999	0.091
p3	0.982	0.009	1.015	0.034	0.953	0.096
	Young et al. (2011)		Yang et al. (2012)		Yang et al. . (2013)	
p1	1.168	0.236	1.032	0.117	1.087	0.095
p2	1.02	0.027	1.003	0.015	1.01	0.011
p3	0.998	0.027	0.995	0.015	1.012	0.012
	Wang et al. . (2015)		Asomaning et al. (2000)			
p1	1.068	0.214	0.953	0.046		
p2	1.008	0.026	0.994	0.006		
p3	0.97	0.027	1.001	0.007		

Source: The author, 2019.

Figure 12 - Comparison of the calculated and experimental data for the thermal fouling resistance.

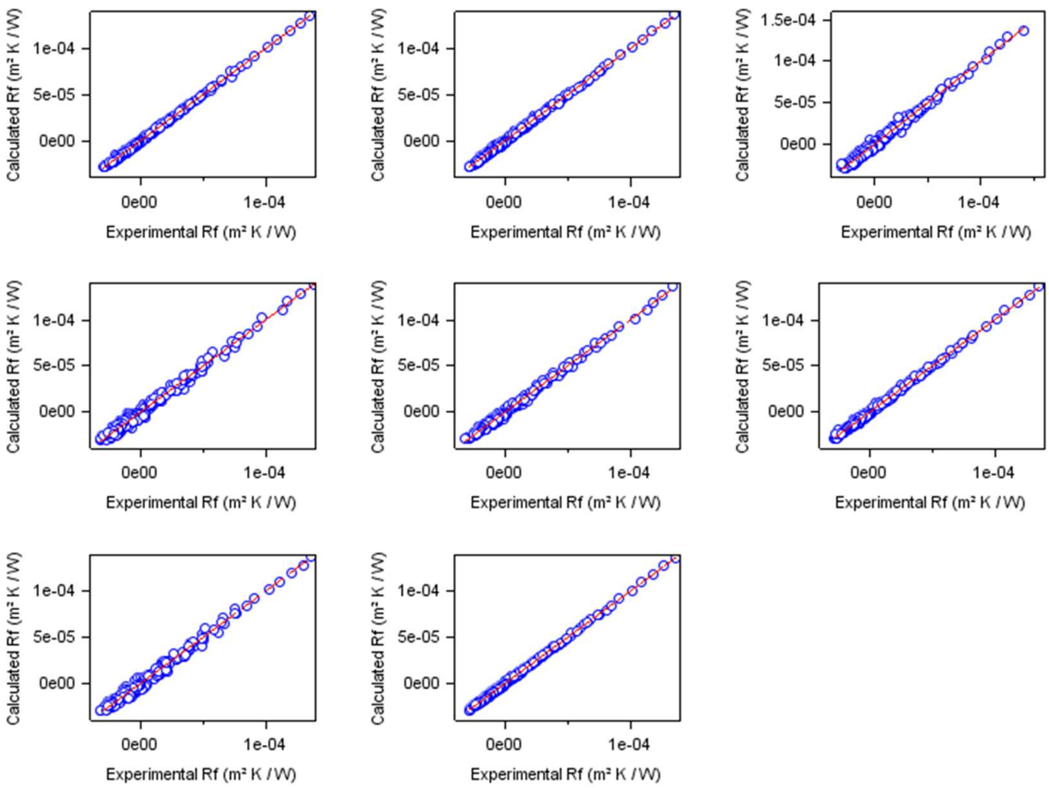


Fig a) Yang et al. Fig b) Srinivasan and Watkinson. Fig c) Saleh et al. Fig d) Young et al. Fig e) Yang et al. Fig f) Yang et al. Fig g) Wang et al. Fig h) Asomaning et al.

Source: The author, 2019.

Table 5 - Values of the estimated parameter and their standard deviation obtained for each author for the thermal fouling resistance.

	Yang et al. . (2011)		Srinivasan and Watkinson (2005)		Saleh et al. (2005a)	
	parameter	standard deviation	parameter	standard deviation	parameter	standard deviation
p1	0.98	0.043	0.983	0.047	1.224	0.162
p2	0.998	0.006	0.998	0.006	1.026	0.017
p3	1.001	0.006	0.995	0.006	0.99	0.017
	Young et al. (2011)		Yang et al. (2012)		Yang et al. . (2013)	
p1	0.938	0.132	1.024	0.081	1.058	0.063
p2	0.99	0.018	1.003	0.01	1.008	0.008
p3	1.033	0.019	0.999	0.01	0.99	0.008
	Wang et al. . (2015)		Asomaning et al. (2000)			
p1	0.925	0.126	0.987	0.034		
p2	0.989	0.018	0.999	0.004		
p3	1.015	0.018	0.996	0.004		

Source: The author (2019).

Figure 13 shows data of the predicted fouling rate x calculated fouling rate for different authors. The deviation for d*Rf*/d*t* used was obtained through *Rf* error propagation, present in Table 2, from a linear fit similar to that used to obtain errors for *Ts*. This error propagation was done because the errors for d*Rf*/d*t* presented in Table 2 were obtained from graphs provided by the author with a high number of experimental points, thus presenting minimal values. In the present work, the number of experimental points used in the estimation was smaller Therefore, it was necessary to use an error for d*Rf*/d*t* compatible with the number of points. In this case again the parameter α presents a small deviation beyond the expected.

Figure 13 - Comparison of the calculated and experimental data for the fouling rate.

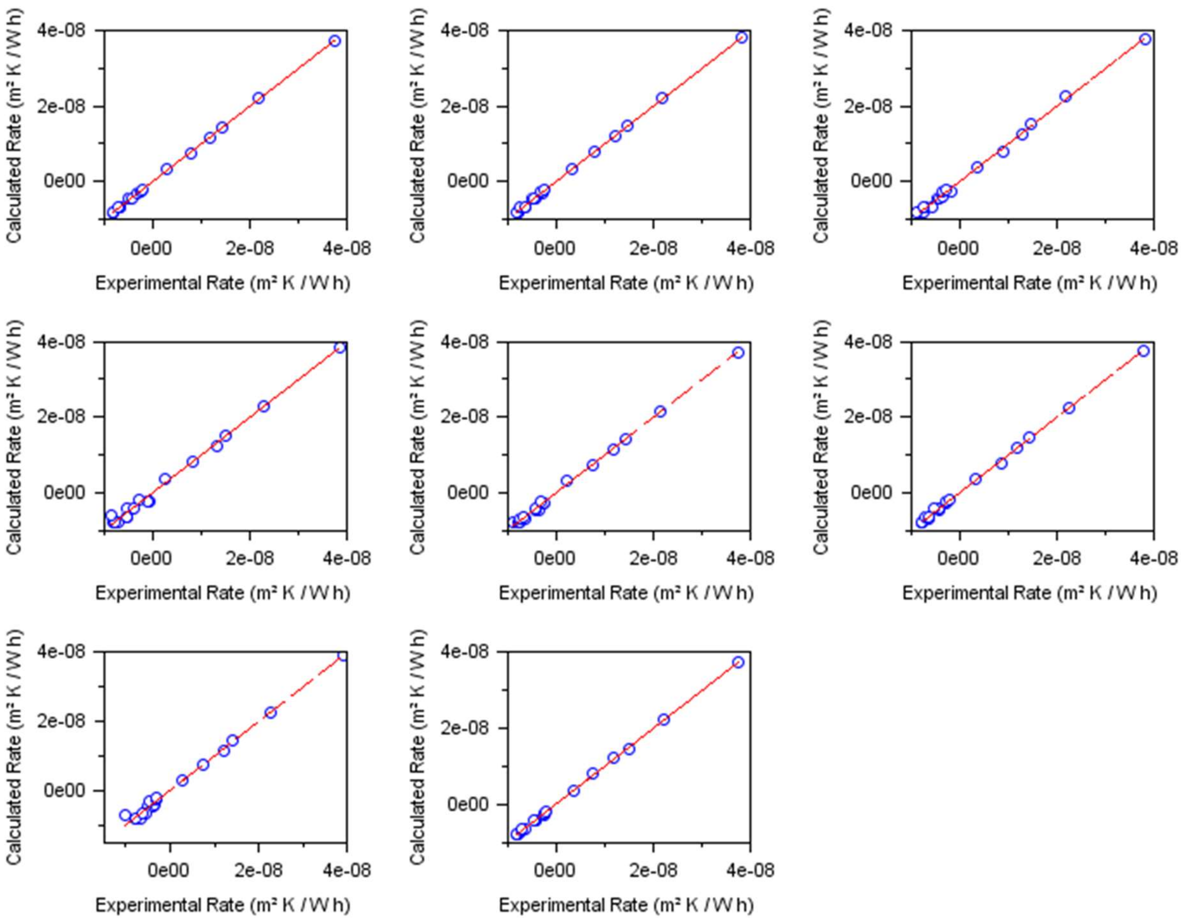


Fig a) Yang et al. Fig b) Srinivasan and Watkinson. Fig c) Saleh et al. Fig d) Young et al. Fig e) Yang et al. Fig f) Yang et al. Fig g) Wang et al. Fig h) Asomaning et al.
Source: The author, 2019.

Table 6 - Values of the estimated parameter and their standard deviation obtained for each author for the fouling rate.

	Yang et al. (2011)		Srinivasan and Watkinson (2005)		Saleh et al. (2005a)	
	Parameter	standard deviation	parameter	standard deviation	parameter	standard deviation
p1	0.982	0.096	0.989	0.104	0.784	0.221
p2	0.999	0.013	0.998	0.014	0.967	0.036
p3	1.007	0.013	1.002	0.014	1.002	0.038
	Young et al. (2011)		Yang et al. (2012)		Yang et al. . (2013)	
p1	0.856	0.263	1.097	0.199	0.876	0.115
p2	0.978	0.04	1.015	0.023	0.983	0.017
p3	0.955	0.041	0.989	0.023	0.994	0.017
	Wang et al. . (2015)		Asomaning et al. (2000)			
p1	1.491	0.462	0.827	0.063		
p2	1.049	0.04	0.976	0.01		
p3	1.008	0.04	1.001	0.01		

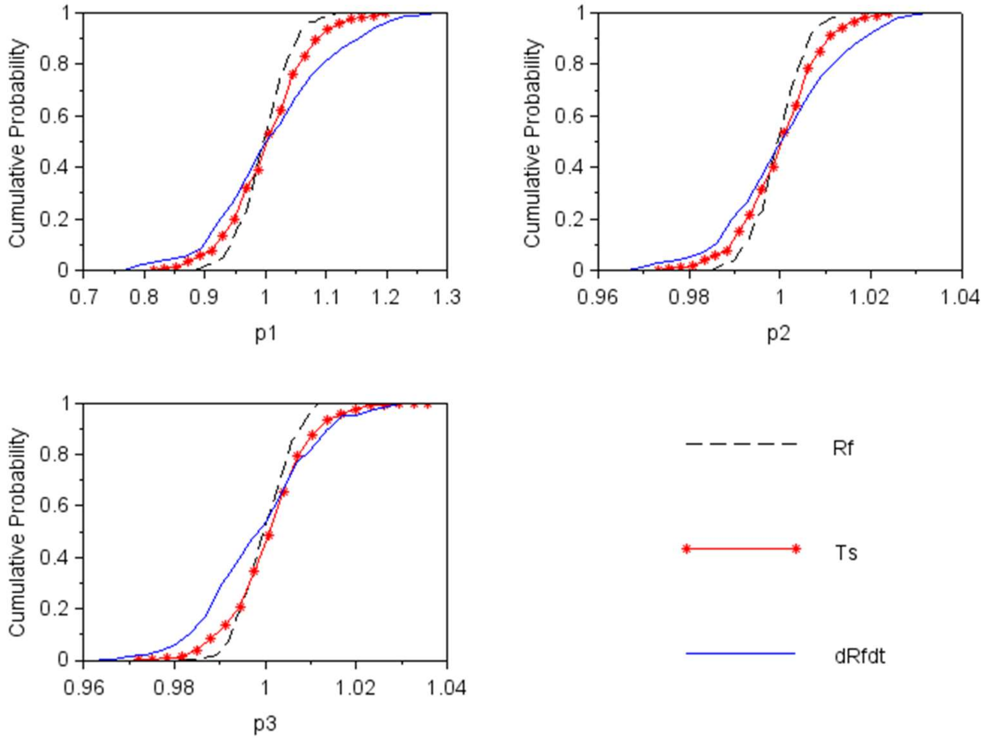
Source: The author, 2019.

In all cases, it can be observed that shallow errors were obtained from almost all estimations. Besides, there is no difference in the parameter estimation regardless of the form used, because the considered errors were propagated between them.

It is noteworthy that the parameter estimation used involves a probabilistic component, which means that with each execution of the different routine values for the parameters are obtained. Thus, in order to evaluate whether the previously presented estimation has the behavior as expected, we also analyzed the dispersion of the parameter values. For this, a loop was created in the parameter estimation routines capable of performing the estimation a high number of times (250 times more precisely) in which the values of the estimated parameters were stored. This analysis was done for the three different forms of estimation, but considering only data provided by one author (for simplicity only), but it can be performed for all others.

Figure 14 represents the cumulative frequency data of the parameters for the three forms of estimation using the errors for the variables obtained by the author. Cumulative probability results show that the distribution of the estimated parameters is very similar for the three estimation forms, which confirms that there is no difference between them, since the errors of the variables are propagated to each other.

Figure 14 - Normalized cumulative distribution of the estimated parameters.



Source: The author, 2019.

3 MODELS EVALUATION

Abstract

In this chapter, some models are presented to predict the fouling rate in heat exchangers. Based on these models, a discussion is conducted on which can fit the experimental data considering the experimental error obtained in the study performed in the previous chapter. This analysis is done through statistical measures such as AIC, χ^2 and R^2.

3.1 Crude oil Fouling Models

To evaluate and to overcome problems caused by crude oil fouling, several semi-empirical models were developed capable of predicting the fouling rate based on the concept of fouling threshold. Only the most common models will be studied here.

The concept of fouling threshold started with the studies developed by Ebert and Panchal in 1995 (EP) and consists in a model that correlated the fouling rate with film temperature and the shear stress (EBERT; PANCHAL, 1996), in two terms, deposition and removal (WILSON; POLLEY; PUGH, 2005).

$$\frac{dR_f}{dt} = \alpha \, Re^{\beta} \, e^{\left(\frac{-E_a}{R\,T_f}\right)} - \gamma \, \tau_w \tag{26}$$

where the film temperature and the shear stress are represented by the Equations (27) and (28), respectively (WANG et al., 2015):

$$Tf = Tc + 0.55\,(Tw - Tc) \tag{27}$$

$$\tau_w = \frac{f\,\rho\,v^2}{2} \tag{28}$$

Afterward, other researchers proposed some modifications in this model. At the first modification issue in 1999, Ebert and Panchal (EPM) inserted the Prandtl number in the deposition term (COSTA et al., 2013) in order to better the specific thermal conductivity content (WANG et al., 2015).

$$\frac{dR_f}{dt} = \alpha\, Re^{\beta}\, Pr^{-0.33}\, e^{\left(\frac{-E_a}{R\,T_f}\right)} - \gamma\, \tau_w \tag{29}$$

Latter, Polley et al. (Pol) concluded in their studies that the temperature threshold rises rapidly with flow velocity. Otherwise, select replace the film temperature with surface temperature in the Arrhenius equation, a term containing Re to replace the shear stress. (WANG et al., 2015)

$$\frac{dR_f}{dt} = \alpha\, Re^{-0.8}\ Pr^{-0.33}\, e^{\left(\frac{-E_a}{R\,T_w}\right)} - \gamma\ Re^{0.8} \tag{30}$$

In 2004, Yeap et al. (Yea) proposed a model based on Epstein's model that considered cases of chemical reaction fouling (YEAP et al., 2004). In this model, it was employed the term $v^{0.8}$ to represent the removal term caused by turbulent mass transfer (WANG et al., 2015).

$$\frac{dR_f}{dt} = \frac{\alpha\, f\, v\, T_W^{2/3} \rho^{2/3} \mu^{-4/3}}{1+\beta\, v^3\, f^2 \rho^{-1/3} \mu^{-\frac{1}{3}}\, T_W^{2/3} e^{\left(\frac{Ea}{R\,T_W}\right)}} - \gamma\, v^{0.8} \tag{31}$$

Nasr and Givi, in 2006 (NG), proposed the model below, free of Prandtl number and modifying the removal term (COSTA et al., 2013):

$$\frac{dR_f}{dt} = \alpha\, Re^{\beta} e^{\left(\frac{-E_a}{R\,T_f}\right)} - \gamma\ Re^{0.4} \tag{32}$$

Ma (Ma) and Wang (Wan) suggested the following models for the fouling rate (WANG et al., 2015):

$$\frac{dR_f}{dt} = \alpha \; Re^{-0.35} \; Pr^{-0.33} \; e^{\left(\frac{-E_a}{R\,T_f}\right)} - \gamma \;\; Re^{0.35} \tag{33}$$

$$\frac{dR_f}{dt} = \alpha \; Re^{-0.35} \; Pr^{-0.33} \; e^{\left(\frac{-E_a}{R\,T_f}\right)} - \gamma \;\; Re^{0.8} \tag{34}$$

Fluentes (Flu) introduced the internal heat transfer coefficient in the fouling term for predicting the reaction fouling rate (WANG et al., 2015):

$$\frac{dR_f}{dt} = \frac{\alpha}{h_i} e^{\left(\frac{-E_a}{R\,T_f}\right)} - \gamma \, \tau_w \tag{35}$$

Some new expressions are needed to evaluate of the models mentioned before, for example: to determine the physical properties of the crude oil, Reynolds and Prandtl numbers, friction factor, Nusselt number and the heat transfer coefficient. These expressions are found in the Appendix B.

When candidate models are investigated, statistical measures and tests can be used to evaluate the suitability of such models to data, besides model comparison. In the next section, an overview of methods used for such purpose is presented.

To facilitate understanding and simplifying the candidate models to be evaluates, Table 7 summarizes which parameters are to be estimated in each model used in this part of this work.

Table 7 – Parameters that are estimated in each Threshold type model used.

Model	Estimated parameters
EP	α, β, E_a and γ
EPM	α, β, E_a and γ
Pol	α, E_a and γ
Yea	α, β, E_a and γ
NG	α, β, E_a and γ
Ma	α, E_a and γ
Wan	α, E_a and γ
Flu	α, E_a and γ

Source: The author, 2019.

3.2 Statistic measures for models evaluation

The following hypothesis is typically admitted in parameter estimation:

- Errors of inlet variables are negligible.
- The outlet variables y^{exp} follow a normal distribution with the mean given by the model y^{calc} $[p]$ (the model is admitted to be true) and a known variance σ_y^2 (admitted to be constant in this case).
- The experimental points are independent one each other.
- The probability of experimental points obtained is maximal.

From the above hypothesis, the likelihood function becomes (BARD, 1974):

$$L = \frac{1}{\sqrt{\left(2\,\pi\,\sigma_y^2\right)^n}}\,\exp\left(-\frac{1}{2}\sum_{i=1}^{n}\frac{\left(y_i^{exp}-y_i^{calc}\right)^2}{\sigma_y^2}\right) \tag{36}$$

The logarithm of the likelihood function becomes:

$$\ln L = -\frac{n}{2}\ln\left(2\,\pi\sigma_y^2\right) - \frac{1}{2\cdot\sigma_y^2}\sum_{i=1}^{n}\left(y_i^{exp}-y_i^{calc}\right)^2 \tag{37}$$

Defining F_{obj} as:

$$F_{obj} = \frac{1}{\sigma_y^2}\sum_{i=1}^{n}\left(y_i^{exp}-y_i^{calc}\right)^2 \tag{38}$$

Then:

$$\ln L = -\frac{n}{2}\ln\left(2\,\pi\sigma_y^2\right) - \frac{F_{obj}}{2} \tag{39}$$

For achieving parameter estimation, maximizing L is the same as maximizing $\ln L$ alternatively, minimizing F_{obj}.

$$\hat{p} = \arg\min_p F_{obj} \tag{40}$$

After estimation, some statistical tests can be used to evaluate models adherence to experimental data, and models comparison one each other. In the following, the criteria used in this work are described.

3.2.1 Chi-square test

Under the hypothesis stated previously, the F_{obj} for a given model follows a chi-square distribution function. Then, one can evaluate the model adequacy evaluating the following relation:

$$\phi = 1 - \int_0^{F_{obj}} \mathcal{P}[\chi^2] \cdot d\chi^2 \tag{41}$$

The values of ϕ have been called as *the model probability* for its simplicity (SCHWAAB et al., 2006). When ϕ becomes lower than a defined limit (for example, 5%), the model is considered inadequate to explain experimental data given the considered experimental uncertainty; otherwise, the model is admitted to explain the experimental data.

3.2.2 Coefficient of determination R²

After the parameter estimation for a given model, one gets all the pairs $\left(y_i^{exp}, y_i^{calc}\right)$. Then, the coefficient of determination R^2 can be computed as:

$$R^2 = 1 - \frac{\sum_i \left(y_i^{exp} - y_i^{calc}\right)^2}{\sum_i \left(y_i^{exp} - \overline{y^{exp}}\right)^2} \quad (42)$$

where $\overline{y^{exp}}$ is the mean of the experimental output variable vector. In R^2 expression, $\sum_i \left(y_i^{exp} - y_i^{calc}\right)^2$ is proportional to the variance of the model residuals; while $\sum_i \left(y_i^{exp} - \overline{y^{exp}}\right)^2$ is proportional to the variance of experimental data. So, the ratio $\frac{\sum_i \left(y_i^{exp} - y_i^{calc}\right)^2}{\sum_i \left(y_i^{exp} - \overline{y^{exp}}\right)^2}$ is proportional to the variance of residuals (variance explained by model) divided by the variance of data. Thus, if the model is good, $\sum_i \left(y_i^{exp} - y_i^{calc}\right)^2$ is low enough and R^2 reaches near the unit value. It can be noted that R^2 can assume negative values if the model explains data worse than a simple mean of experimental data.

3.2.3 Akaike Information Criteria (AIC)

AIC can be understood as an index, helping to suggest the quality of the model. Typically, it is used in model comparisons (this is why it is so useful), and seeks to lead to the best model, not necessarily the perfect one. AIC is derived from Kulback-Leibler divergence criterion, considering the true distribution of data $f(y)$ moreover, a model that depends on parameters $\hat{p}$, representing the distribution predicted by the model as $g(y|\hat{p})$. The Kulback-Leibler divergence criterion between such distributions $KL_{f,g}$ can be written as:

$$KL_{f,g} = \int f[y] \cdot \ln \frac{f[y]}{g[y|\hat{p}]} \, dy \quad (43)$$

It must be emphasized that the true distribution $f[y]$ is generally not known; otherwise, estimation would be not necessary. For simplicity, let us represent $f[y]$ as f and $g[y|\hat{p}]$ as $\hat{g}$. The above expression can be written as:

$$KL_{f,\hat{g}} = \int f \cdot \ln f \, dy - \int f \cdot \ln \hat{g} \, dy \quad (44)$$

$$KL_{f,\hat{g}} = E_f[\ln f] - E_f[\ln \hat{g}] \tag{45}$$

where $E_f[\cdot]$ is the expectation regarding the distribution function f. In the above expression, $E_f[\ln f]$ does not depend on the model, while $E_f[\ln \hat{g}]$ is dependent on the model. Assuming as a risk function the mean value of $KL_{f,\hat{g}}$ (BOZDOGAN, 1987), i.e.:

$$Risk = E_f\left[KL_{f,\hat{g}}\right] \tag{46}$$

Substituting the above expression in the previous one:

$$Risk = E_f\left[E_f[\ln f] - E_f[\ln \hat{g}]\right] = E_f\left[E_f[\ln f]\right] - E_f\left[E_f[\ln \hat{g}]\right] \tag{47}$$

Since $E_f\left[E_f[\ln f]\right]$ does not depend on the model; the risk can be evaluated according to the expression:

$$-E_f\left[E_f[\ln \hat{g}]\right] \tag{48}$$

Akaike showed that, under specific hypothesis, the evaluation of the term above is similar to evaluate the logarithmic of likelihood function of estimated model ($\log L[\hat{p}]$) plus the number of parameters of the model n_p, i.e. (BURNHAM; ANDERSON, 2010):

$$\text{Evaluate} - E_f\left[E_f[\ln \hat{g}]\right] \rightarrow \text{Evaluate}\left(-\log L[\hat{p}] + n_p\right) \tag{49}$$

Then, theAIC criterion was defined as:

$$AIC = -2 \cdot \log L[\hat{p}] + 2 \cdot n_p \tag{50}$$

Since the parameters are estimated, $L[\hat{p}]$ should be maximized, then as higher $L[\hat{p}]$, lower $-2 \cdot \log L[\hat{p}]$ (using Equation (39)) and consequently lower is AIC. Thus, lower values of AIC indicate better adherence of data to the model.

AIC for models comparison

Usually, when comparing different models the AIC is ranked from the best model to the worst model (from a lower value of AIC to a higher value of AIC). Then, ΔAIC^{mod} for a model mod can be computed as:

$$\Delta AIC^{mod} = AIC^{mod} - \min_m[AIC^m] \tag{51}$$

Unfortunately, ΔAIC^{mod} is not a statistical test, but an index for the quality of the models' comparison. Despite a cut-off value cannot be rigorously defined, Burnham and Anderson (BURNHAM; ANDERSON, 2010) suggested the values for evaluation as shown in Table 8, despite a cut-off value cannot be rigorously defined, because it is a heuristic used by some authors in the literature.

Table 8 – Rough level of support of the model according to ΔAIC^{mod}.

ΔAIC^{mod}	**Level of support of model mod**
0-2	Substantial
4-7	Considerably less
>10	Essentially none

Source: BURNHAM; ANDERSON, 2010.

3.3 This work

Among a large number of models, some questions arise, such as:

- How is the adherence of models to fit data generated using other models?
- Can the models be discriminated for typical experimental conditions and typical error of output variables, when one of the models is considerd "true model" in a set of candidate models?

- Is there a '*best model*' that can be used independently of the model used to generate the pseudo-experimental data?

To answer these questions, in the present work, it was simulated pseudo-experimental data for each one of the eight models previously described, in typical experimental conditions. The experimental error was taken from the typical values found in the literature, discussed in the previous section of this work. For each model considered to generate pseudo-experimental data, estimation of parameters of all models were obtained, as well as the criterions of ΔAIC, χ^2 e R^2. The results for each model used to generate pseudo experimental data were evaluated and discussed based on these three criterion. Concerns about the weight of the experimental error on the models' evaluation are also presented.

3.4 Methodology

3.4.1 Experimental error from literature typical values

As already mentioned, the comparison of different models was performed only considering d*Rf*/d*t* as output variable (i.e., *Rf* and T_s were not considered since the results are expected to be similar). The experimental error presents a pivotal role in the models evaluation; thus, efforts in characterizing such error seem to be necessary. In the previous section, typical experimental errors were obtained from the literature results, and summarized in Table 2. For the sake of clarity, the data regard to $\sigma_{dRf/dt}$ are reproduced bellow, together with the mean value (Table 9).

Table 9 – Range of values used in the simulation of experimental data (m² K/J).

Author	$\boldsymbol{\sigma_{dRf/dt}}$***10¹⁰**
Yang et al (2011)	0.16
Srinivasan and Watkinson (2005)	0.04
Saleh et al (2005a)	0.06
Young et al (2011)	0.88
Yang et al (2012)	0.23
Yang et al (2013)	0.39
Wang et al (2015)	0.94
Asomaning et al (2000)	0.005
Mean value	**0.34**

Source: The author, 2019.

From Table 9, the mean value of the standard deviation of $\overline{\sigma_{dRf/dt}}$ is $3.38 \cdot 10^{-11}$ m² K/J . It is important to emphasize that the order of magnitude of dR_f/dt is around 10^{-9} to 10^{-10} m² K/J for several studies in the literature; thus, the standard error adopted for dR_f/dt is quite low. Such standard error comes from curves of R_f versus t; consequently, a large number of points used in experimental curves of R_f versus t implies in very precise values of inclination of the curve of R_f versus t, leading to such small errors in dR_f/dt. Other sources of error were not considered, besides they could change the magnitude of the fouling rate.

3.4.2 Pseudo-experimental data

In this section, the pseudo-experimental data were generated from a factorial planning according to the values presented in Table 10 based on the experimental analysis used for some authors discussed in the previous sections.

For simplicity, GenMod is defined as the model used to generate pseudo-experimental data.

Table 10 – Values used in a factorial planning for generated the pseudo-experimental data.

q (W/m³)	v (m/s)	Tc (K)
29100	0.5	570
100000	0.5	570
29100	5	570
100000	5	570
29100	0.5	636.67
100000	0.5	636.67
29100	5	636.67
100000	5	636.67

Source: The author, 2019.

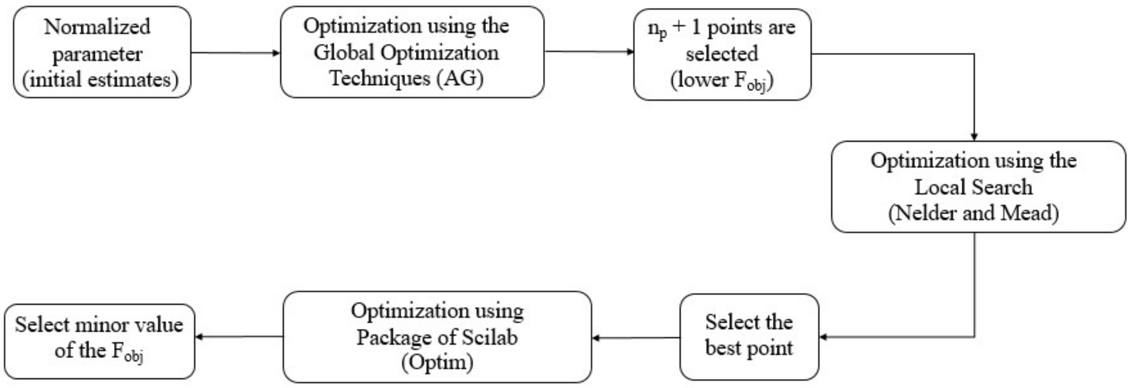
3.4.3 Parameter estimation

The objective function considered in the optimization was:

$$F_{obj} = \frac{\sum_i \left(r_{net,i}^{exp} - r_{net,i}^{cal}\right)^2}{\sigma_{r_{net}}^2} \tag{52}$$

A hybrid algorithm was used for parameter estimation. First, genetic algorithm was employed to get the best $n_p + 1$ points; after, such best points were informed to a Nelder and Mead algorithm for achieving the second layer of optimization. After that, the optimization package of Scilab *optim* was used (BFGS method), receiving the best value of the parameters in previous sections. Figure 15 illustrates these procedures.

Figure 15 – Procedure of the hybrid algorithm used.



Source: The author, 2019.

The inputs of the algorithms are presented in Table 11Table 11 – Inputs of different algorithms*. Previous studies on the estimation using genetic algorithm showed that a larger number of generation did not modify the estimation results. Besides, after the GA, a local search algorithm is used, which improves the quality of the result.

Table 11 – Inputs of different algorithms*.

Genetic Algorithm	
Population size	20
Maximum number of generation	50
Crossover probability	0.90
Mutation probability	0.07
Nelder and Mead	
Tolerance*	10^{-4}
Coefficient of reflection	1
Coefficient of expansion	2
Contraction coefficient	0.5

Source: The author, 2019.

3.4.4 Evaluation of models

The models were evaluated according to ΔAIC, χ^2 e R^2, described in previous sections.

3.5 Results and Discussion

The results obtained in the analysis of each model for ΔAIC, χ^2 e R^2 separated by the model used to generate data are presents below. The ordering of the models is done in all cases considering from the best to the worst model and is presented in parenthesis. Comments for each generated model will be presented.

It must be pointed out that ΔAIC and χ^2 can lead to different results,although both criteria depend on sum of squared residual and on the number of model parameters, this last term penalizes each criteria differently. In χ^2 the degrees of freedom is reduced with higher parameters values, while ΔAIC contains the number of parameters explicitly inside the criterion. Thus different results between ΔAIC and χ^2 are possible.

For Polley as GenMod, ΔAIC, χ^2 and R^2 criteria present the same order of the models (best to worst) as shown in the Table 12. The results obtained for the ΔAIC show that all models, except the own generator, present values of the ΔAIC much higher than the limit of 10 (suggested in Table 8), suggesting that only the Polley model can fit the experimental data generated by itself. The χ^2 criterion confirms this analysis from the low values obtained for the probability of adjustment, that is, values lower than 5%. Nonetheless, the R^2 criterion shows a strong correlation between predicted and calculated fouling rate values for all models (except Yea), suggesting a good adherence of experimental data to each model. The results can be easily explained: the experimental error of $\overline{\sigma_{dRf/dt}} = 3.38 \cdot 10^{-11}$ m² K/J used for pseudo-experimental data is quite low; thus, few models were able to predict data within this precision, despite the most of them were able to correlate the trend of experimental and calculated values.

Table 12 - Evaluation the suitability of each model fit the data generated with the Polley model from statistical measures and tests.

Model	ΔAIC		χ^2		R^2	
Pol	0.00	(1)	0.796	(1)	1.000	(1)
NG	33716.99	(4)	0.000	(4)	0.979	(4)
EPM	27824.46	(2)	0.000	(2)	0.983	(2)
Ma	44656.75	(7)	0.000	(7)	0.972	(7)
Wan	36917.08	(6)	0.000	(6)	0.977	(6)
EP	29254.22	(3)	0.000	(3)	0.982	(3)
Flu	33832.44	(5)	0.000	(5)	0.979	(5)
Yea	1.57E+06	(8)	0.000	(8)	0.026	(8)

Source: The author, 2019.

For Nasr and Givi as GenMod, the order of the models is different according to the three criteria: ΔAIC, χ^2 and R^2; despite the results are similar for χ^2 and R^2 as shown the Table 13. Values obtained for the ΔAIC and χ^2 obey the ranges of values characterizing that all models can fit pseudo-experimental data within their precision. This case deserves particular attention, because although all models can provide a good fit according ΔAIC and χ^2, they also present a low correlation between the predicted and calculated rate values. The poor results for the R^2 criterion indicate that the models do not follow the trend of the experimental data. Again, the results can be explained based on the relative error of dRf/dt: the pseudo-experimental data generated with parameters of Nasr and Givi model reported in work of Wang et al (WANG et al., 2015) has the same order of experimental uncertainties. In other words, the error is too high for the experimental data; thus, the fit within the data uncertainties seems to lead to a good result, besides models could not represent the tendency of experimental data. Since the pseudo-experimental data are corrupted with a random error (Gaussian with mean given by the model and $\overline{\sigma_{dRf/dt}} = 3.38 \cdot 10^{-11}$ m² K/J), where the order of magnitude of the measure is the same of the error, experimental data have no tendency; thus, even the NG model was not able to lead to a good R^2 value.

Table 13 - Evaluation the suitability of each model fit the data generated with the Nasr and Givi model from statistical measures and tests.

Model	ΔAIC		χ^2		R²	
Pol	0.00	(1)	0.280	(1)	0.258	(1)
NG	2.07	(5)	0.175	(4)	0.257	(2)
EPM	2.08	(6)	0.174	(5)	0.248	(5)
Ma	2.14	(7)	0.135	(7)	0.185	(7)
Wan	0.09	(3)	0.271	(6)	0.247	(6)
EP	2.06	(4)	0.175	(2)	0.251	(3)
Flu	0.07	(2)	0.274	(3)	0.250	(4)
Yea	4.28	(8)	0.073	(8)	0.009	(8)

Source: The author, 2019.

For modified Ebert and Panchal as GenMod, the order of the models is the same for ΔAIC and χ^2, and a slightly different for R^2, as shown the Table 14. Despite the excellent correlation obtained for most of the models (except Yea), the results obtained shown that EPM and Flu fit suitably the experimental data according to ΔAIC and χ^2, while Wan and EP presents a considerably lower probability according to χ^2 also, level of support according to ΔAIC. The inadequacy of models according to χ^2 also, ΔAIC, despite to a high value of R^2 for almost all models, suggests a low value for experimental error.

Table 14 - Evaluation the suitability of each model fit the data generated with the modified Ebert and Panchal model from statistical measures and tests.

Model	ΔAIC		χ^2		R²	
Pol	258.73	(6)	0.000	(6)	0.994	(7)
NG	266.70	(7)	0.000	(7)	0.995	(6)
EPM	0.00	(1)	0.473	(1)	1.000	(2)
Ma	45.80	(5)	0.000	(5)	0.999	(5)
Wan	4.09	(3)	0.087	(3)	1.000	(4)
EP	6.87	(4)	0.034	(4)	1.000	(1)
Flu	0.20	(2)	0.333	(2)	1.000	(3)
Yea	13840.15	(8)	0.000	(8)	0.694	(8)

Source: The author, 2019.

For Ma and Yeap as GenMod, the order of the models is almost totally similar for all criteria as shown the Table 15 and Table 16, respectively. Results obtained for ΔAIC and χ^2 show that only own generated data fit suitably the experimental data according to the ranges of values for these criteria. In all other cases, adjustment is not possible according to the χ^2 also, ΔAIC criteria, despite the excellent correlation that exists in some of the cases, which suggests a low value for experimental error.

Table 15 - Evaluation the suitability of each model fit the data generated with the Ma model from statistical measures and tests.

Model	ΔAIC		χ^2		R^2	
Pol	2.07E+09	(4)	0.000	(4)	0.497	(4)
NG	70943.52	(2)	0.000	(2)	1.000	(2)
EPM	4.85E+09	(7)	0.000	(7)	-0.181	(7)
Ma	0.001	(1)	0.082	(1)	1.000	(1)
Wan	2.07E+09	(5)	0.000	(5)	0.497	(5)
EP	4.85E+09	(8)	0.000	(8)	-0.181	(6)
Flu	4.85E+09	(6)	0.000	(6)	-0.181	(8)
Yea	1.72E+08	(3)	0.000	(3)	0.977	(3)

Source: The author, 2019.

Table 16 - Evaluation the suitability of each model fit the data generated with the Yeap model from statistical measures and tests.

Model	ΔAIC		χ^2		R^2	
Pol	3.515E+15	(8)	0.000	(8)	-0.785	(8)
NG	2.765E+14	(6)	0.000	(6)	0.860	(6)
EPM	1.347E+14	(4)	0.000	(3)	0.932	(3)
Ma	3.858E+14	(7)	0.000	(7)	0.804	(7)
Wan	3.16E+12	(2)	0.000	(2)	0.998	(2)
EP	1.347E+14	(5)	0.000	(4)	0.932	(4)
Flu	1.347E+14	(3)	0.000	(5)	0.932	(5)
Yea	0.001	(1)	0.366	(1)	1.000	(1)

Source: The author, 2019.

For Wang as GenMod, the order of the models is the same for ΔAIC and χ^2, and a slightly different for R^2, as shown the Table 17. Despite the excellent correlation obtained for all models, the results obtained show that the own generated data fit suitably the experimental data both according for ΔAIC and χ^2, while Pol presents a considerably inferior probability according to χ^2 and level of support according to ΔAIC. The inadequacy of models according to χ^2 also, ΔAIC, despite to the high value of R^2 for almost all models, suggests a low value for experimental error.

Table 17 - Evaluation the suitability of each model fit the data generated with the Wang model from statistical measures and tests.

Model	ΔAIC		χ^2		R^2	
Pol	3.55	(2)	0.057	(2)	1.000	(2)
NG	3.27E+05	(7)	0.000	(7)	0.859	(8)
EPM	8.14E+05	(8)	0.000	(8)	0.931	(7)
Ma	9.45E+02	(4)	0.000	(4)	1.000	(4)
Wan	0.001	(1)	0.209	(1)	1.000	(1)
EP	1.42E+05	(6)	0.000	(6)	0.939	(6)
Flu	1.42E+05	(5)	0.000	(5)	0.939	(5)
Yea	2.57E+02	(3)	0.000	(3)	1.000	(3)

Source: The author, 2019.

For Ebert and Panchal as GenMod, the order of the models is the same for χ^2 also, ΔAIC, and a slightly different for R^2, as shown the Table 18. The results obtained showed that only EPM and the own generated data fit suitably the experimental data according to ΔAIC and χ^2, despite the excellent correlation obtained for most of the models (except Yea), which suggests a low value for experimental error.

Table 18 - Evaluation the suitability of each model fit the data generated with the Ebert and Panchal model from statistical measures and tests.

Model	ΔAIC		χ^2		R^2	
Pol	3365.30	(7)	0.000	(7)	0.984	(7)
NG	460.44	(4)	0.000	(4)	0.998	(4)
EPM	0.00	(1)	0.463	(1)	1.000	(2)
Ma	1565.81	(6)	0.000	(6)	0.992	(6)
Wan	624.38	(5)	0.000	(5)	0.997	(5)
EP	0.99	(2)	0.332	(2)	1.000	(1)
Flu	15.31	(3)	0.001	(3)	1.000	(3)
Yea	178024.50	(8)	0.000	(8)	0.150	(8)

Source: The author, 2019.

For Fluentes as GenMod, the order of the models is the same for $\Delta AIC, \chi^2$ and R^2 criteria, as shown the Table 19. Despite the excellent correlation obtained for most of the models (except Yea), the results obtained showed that only own generated data fit suitably the experimental data both according to for ΔAIC and χ^2, while NG presents a considerably inferior probability according χ^2 and level of support according to ΔAIC. The inadequacy of models according to χ^2 also, ΔAIC, despite to the high value of R^2 for almost all models, suggests a low value for experimental error.

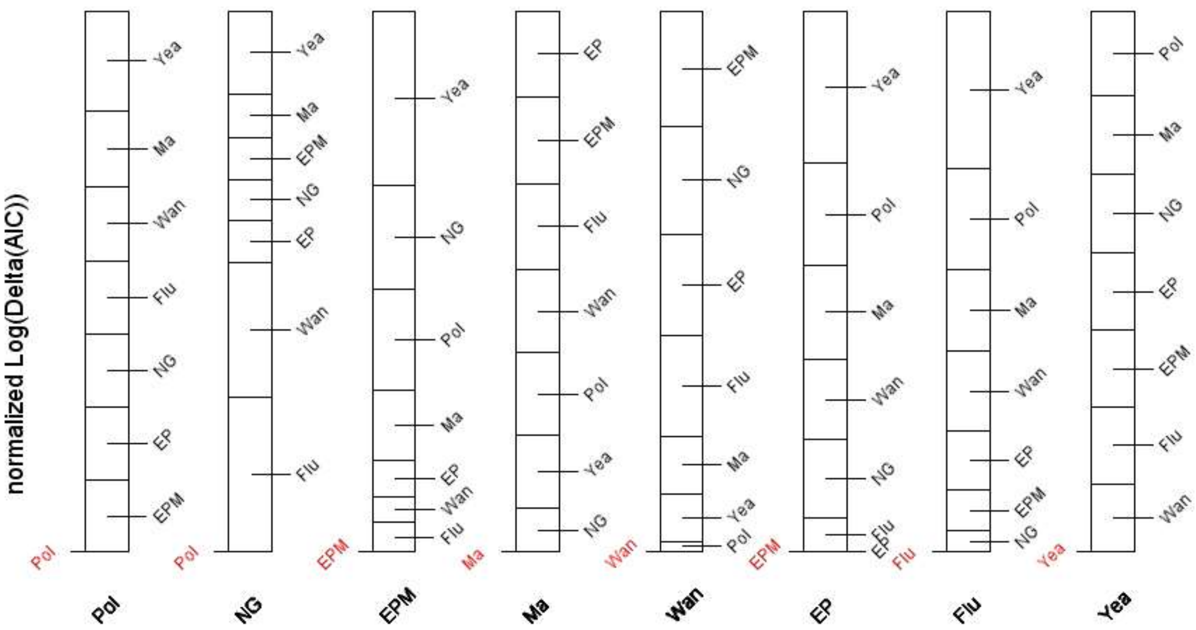
Table 19 - Evaluation the suitability of each model fit the data generated with the Fluentes model from statistical measures and tests.

Model	ΔAIC		χ^2		R²	
Pol	1011.23	(7)	0.000	(7)	0.970	(7)
NG	4.26	(2)	0.004	(2)	1.000	(2)
EPM	16.46	(3)	0.000	(3)	0.999	(3)
Ma	270.95	(6)	0.000	(6)	0.992	(6)
Wan	251.90	(5)	0.000	(5)	0.992	(5)
EP	56.69	(4)	0.000	(4)	0.999	(4)
Flu	0.00	(1)	0.024	(1)	1.000	(1)
Yea	52078.28	(8)	0.000	(8)	-0.540	(8)

Source: The author, 2019.

In most cases, except when the Nasr and Givi model is considered as GenMod, the determination coefficient shows good results compared to the other criteria. This results leads to the conclusion that the considered experimental error, in these cases, is minimal. Figure 16 represents in an orderly way, from the best to the worst model, the sequence of models for all situations according to the AIC criterion.

Figure 16 - Normalized Log (ΔAIC) for all generator models, ordered from the best to the worst model.



Source: The author, 2019.

A comparison between the models used in this work is found in the in Appendix C, where all modifications made in one model for the others are presented. It can be seen that some models are similar to each other, while some have a significant difference between them, which makes difficult to find a way to group them. Thus, together with the results obtained previously, it is difficult to determine the model that best fits the data.

4 CONCLUSIONS AND SUGGESTIONS

This chapter presented the conclusions and suggestions for future work.

Conclusions

The information of experimental uncertainties from the linear regression based on fouling data from the literature allowed the propagated error for the model parameters. It was observed that relative errors of rate of deposition for a single run are bellow 5% in the most cases presented in the literature. Besides, it is possible to observe that relation of pre-exponential factor and the activation energy, known as *the compensation effect*, is confounded with statistical effect int the most cases.

Based on the error of the variable *Rf* obtained through the literature, the parameter estimation for the three experimental data types does not present very different results among them, in which the values of the parameters remained close to the reference values, characterizing a good parametric estimation. This result was already expected since the original errors are propagated to other variables linearly. Thus, regardless of which experimental data are used in the estimation of parameters, the results will be very similar.

Results of the statistical analysis among fouling rate models showed that the experimental error used for pseudo-experimental data is quite low for the number of experimental points used in this work, when compared to the authors. Thus, few models were able to predict data within this precision, although most of them were able to correlate the trend of experimental and calculated values. For this reason, it is possible that none of the models can adjust to the experimental data with precision, regardless of the generator models considered.

Suggestions

- Consider and evaluate the adjustment of the experimental data using other models to predict the fouling rate in heat exchangers, even if they are not very common.
- Use other statistical measures to evaluate the possibility of adjusting the models.
- Use tests based on literature data that may not follow a normal distribution,

REFERENCES

AKINÇ, H. E. et al. Fouling monitoring in crude oil preheat trains. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING X, 2013, Budapest, Hungary. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning X*. Budapest, Hungary: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2013.

ANDERSSON, E.; QUAH, J.; POLLEY, G. T. Experience in the application of COMPABLOC^{TM} in refinery pre-heat trains and first analysis of data from an operational unit. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING VIII, 2009, Schladming, Austria. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning VIII*. Schladming, Austria: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2009.

ASOMANING, S.; PANCHAL, C. B.; LIAO, C. F. Correlating Field and Laboratory Data for Crude Oil Fouling. *Heat Transfer Engineering*, v. 21, n. 3, p. 17–23, jul. 2000.

AWAD, M. M. Fouling of Heat Transfer Surfaces. *In*: BELMILOUDI, A. (Ed.). *Heat Transfer - Theoretical Analysis, Experimental Investigations and Industrial Systems*. InTech, 2011.

BARD, Y. *Nonlinear parameter estimation*. New York: Academic Press, 1974.

BARRIE, P. J. et al. A Critical Analysis of the Compensation Effect and Its Application to Heat Exchanger Fouling Studies. *Heat Transfer Engineering*, v. 34, n. 8–9, p. 744–752, 2013.

BENNETT, C. A. et al. Observation of an Isokinetic Temperature and Compensation Effect for High-Temperature Crude Oil Fouling. *Heat Transfer Engineering*, v. 30, n. 10–11, p. 794–804, 2009.

BORIES, M.; PATUREAUX, T. Preheat Train Crude Distillation Fouling Propensity Evaluation by the Ebert and Panchal Model. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING V, 2003, Santa Fe, New Mexico, USA. *Proceedings*...: *Heat Exchanger Fouling And Cleaning: Fundamentals And Applications*. Santa Fe, New Mexico, USA: 2003.

BOZDOGAN, H. Model Selection and Akaike's Information Criterion (AIC): The General Theory and Its Analytical Extensions. *Psychometrika*, v. 52, n. 3, p. 345–370, set. 1987.

BRIGNONE, M. et al. Embaffle © in refinery service. On-field study and data validation through Smartpm©. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING XI, 2015, Enfield (Dublin), Ireland. *Proceedings*...: *International Conference On Heat Exchanger Fouling And Cleaning XI*. Enfield (Dublin), Ireland: M.R. Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2015.

BURNHAM, K. P.; ANDERSON, D. R. *Model selection and multimodel inference:* a practical iformation-theoric approach. 2. Ed. [S.l.]: Springer, 2002.

CHAMBON, A. et al. Fouling in the crude oil distillation preheat train: comparison of experimental data with model results. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING XI, 2015, Enfield (Dublin), Ireland. *Proceedings*...: *International Conference On Heat Exchanger Fouling And Cleaning XI*. Enfield (Dublin), Ireland: M.R. Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2015.

CHUNANGAD, K. S.; CHANG, R. Y.; CASEBOLT, R. P. Evaluation and prediction of the thermal and hydraulic impact of crude oil fouling on exchanger performance using pressure measurements. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING XII, Aranjuez (Madrid), Spain. *Proceedings... International Conference On Heat Exchanger Fouling And Cleaning XII* Aranjuez (Madrid), Spain: H. U. Zettler, 2017.

COLETTI, F. et al. Heat exchanger design with high shear stress: reducing fouling or throughput? *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING XI, 2015, Enfield (Dublin), Ireland. *Proceedings*...: *International Conference On Heat Exchanger Fouling And Cleaning XI*. Enfield (Dublin), Ireland: M.R. Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2015.

COLETTI, F.; MACCHIETTO, S. A Dynamic, Distributed Model of Shell-and-Tube Heat Exchangers Undergoing Crude Oil Fouling. *Industrial & Engineering Chemistry Research*, v. 50, n. 8, p. 4515–4533, 20 abr. 2011.

COSTA, A. L. H. et al. Analysis of the environmental and economic impact of fouling in crude preheat trains for petroleum distillation. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING IX, 2011, Crete Island. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning IX*. Crete Island: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2011.

COSTA, A. L. H. et al. Parameter Estimation of Fouling Models in Crude Preheat Trains. *Heat Transfer Engineering*, v. 34, n. 8–9, p. 683–691, 2013.

CRITTENDEN, B. D. et al. Crude Oil Fouling in a Pilot-Scale Parallel Tube Apparatus. *Heat Transfer Engineering*, v. 30, n. 10–11, p. 777–785, out. 2009.

DIAZ-BEJARANO, E.; COLETTI, F. Modelling shell-side crude oil fouling in shell-and-tube heat exchangers. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING XI, 2015, Enfield (Dublin), Ireland. *Proceedings*...: *International Conference On Heat Exchanger Fouling And Cleaning XI*. Enfield (Dublin), Ireland: M.R. Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2015.

DIAZ-BEJARANO, E.; COLETTI, F.; MACCHIETTO, S. Crude Oil Fouling Deposition, Suppression, Removal, and Consolidation—and How to Tell the Difference. *Heat Transfer Engineering*, v. 38, n. 7–8, p. 681–693, 2017.

DROSG, M. *Dealing with uncertainties*: a guide to error analysis. Berlin: Springer, 2007.

EBERT, W.; PANCHAL, C. B. *Analysis of Exxon crude-oil-slip stream coking data*. United States: Argonne National Lab., IL (United States), 1996.

FAN, Z. et al. Investigation of Fouling Mechanisms of a Light Crude Oil Using an Alcor Hot Liquid Process Simulator. *Energy & Fuels*, v. 24, n. 11, p. 6110–6118, 18 nov. 2010.

FAN, Z. et al. Fouling Characteristics of Hydrocarbon Streams Containing Olefins and Conjugated Olefins. *Energy & Fuels*, v. 25, n. 3, p. 1182–1190, 2011.

HONG, E.; WATKINSON, A. P. Precipitation and Fouling in Heavy Oil–Diluent Blends. *Heat Transfer Engineering*, v. 30, n. 10–11, p. 786–793, 2009.

ISHIYAMA, E. M.; PUGH, S. J. Considering In-Tube Crude Oil Boiling in Assessing Performance of Preheat Trains Subject to Fouling. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING X, 2013, Budapest, Hungary. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning X*. Budapest, Hungary: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2013.

ISHIYAMA, E. M. et al. The importance of scheduling and desalter control of preheat trains of crude distillation units: a case study. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING VIII, 2009, Schladming, Austria. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning VIII*. Schladming, Austria: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2009.

ISHIYAMA, E. M. et al. An industrial case study on retrofitting heat exchangers and revamping preheat trains subject to fouling. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING X, 2013, Budapest, Hungary. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning X*. Budapest, Hungary: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2013a.

ISHIYAMA, E. M. et al. Management of Crude Preheat Trains Subject to Fouling. *Heat Transfer Engineering*, v. 34, n. 8–9, p. 692–701, 2013b.

ISHIYAMA, E. M.; KENNEDY, J.; PUGH, S. J. Fouling Management of Thermal Cracking Units. Em: Heat Transfer Engineering. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING XI, 2015, Enfield (Dublin), Ireland. *Proceedings*...: *International Conference On Heat Exchanger Fouling And Cleaning XI*. Enfield (Dublin), Ireland: M.R. Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2015.

ISHIYAMA, E. M.; PATERSON, W. R.; WILSON, D. I. The effect of fouling on heat transfer, pressure drop and throughput in refinery preheat trains: optimisation of cleaning schedules. *In*: International Conference on Heat Exchanger Fouling and Cleaning XI, Tomar, Portugal. *Proceedings*... INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING XI. Tomar, Portugal: M.R. Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2007.

ISHIYAMA, E. M.; PATERSON, W. R.; WILSON, D. I. The impact of ageing on fouling and cleaning: closing the fouling-cleaning loop. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING IX, 2011, Crete Island. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning IX*. Crete Island: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2011.

ISOGAI, S. et al. Measurement and Modeling for the Mitigation of Organic Crystallization Fouling. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING

AND CLEANING V, 2003, Santa Fe, New Mexico, USA. *Proceedings*...: *Heat Exchanger Fouling And Cleaning: Fundamentals And Applications*. Santa Fe, New Mexico, USA: 2003.

JAFARI NASR, M. R.; MAJIDI GIVI, M. Modeling of Crude Oil Fouling in Preheat Exchangers of Refinery Distillation Units. *Applied Thermal Engineering*, v. 26, n. 14–15, p. 1572–1577, 2006a.

JAFARI NASR, M. R.; MAJIDI GIVI, M. Application of Threshold Model with Various Tube Wall Temperatures for Crude Oil Preheat Train Fouling. *Iranian Journal of Chemistry and Chemical Engineering (IJCCE)*, p. 49–58, 2006b.

JEGLA, Z.; KOHOUTEK, J.; STEHLIK, P. Design and operating aspects influencing fouling inside radiant coils of fired heaters operated in crude oil distillation plants *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING IX, 2011, Crete Island. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning IX*. Crete Island: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2011.

JOSHI, H.; BRONS, G. Chemical Cleaning of Oil Refinery Heat Exchangers -- The Need for a Joint Effort. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING V, 2003, Santa Fe, New Mexico, USA. *Proceedings*...: *Heat Exchanger Fouling And Cleaning: Fundamentals And Applications*. Santa Fe, New Mexico, USA: 2003.

JOSHI, H.; HOEVE, F.; ZIJDEN, E. Applying technology advancements to improve heat exchanger economic and environmental performance in refinery crude preheat trains. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING VII, 2007, Santa Fe, New Mexico, USA. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning - Challenges and Opportunities*. Tomar, Portugal: M.R. Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2007.

JOSHI, H. M. Crude oil fouling field data and a model for pilot-plant scale data. *In*: International Conference on Heat Exchanger Fouling and Cleaning X, Budapest, Hungary. *Proceedings*... INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING X. Budapest, Hungary: H. Müller-Steinhagen, M.R. Malayeri and A.P. Watkinson, 2013.

JOSHI, H. M.; SHILPI, N. B.; AGARWAL, A. Relate Crude Oil Fouling Research to Field Fouling Observations. *In*: International Conference on Heat Exchanger Fouling and Cleaning VIII, Schladming, Austria. *Proceedings*... INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING VIII. Schladming, Austria: H. Müller-Steinhagen, M.R. Malayeri and A.P. Watkinson, 2009.

LANE, M. R.; HARRIS, J. S. Impact of mixing on crude oil fouling tests. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING XI, 2015, Enfield (Dublin), Ireland. *Proceedings*...: *International Conference On Heat Exchanger Fouling And Cleaning XI*. Enfield (Dublin), Ireland: M.R. Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2015.

LEMOS, J. C. et al. A Sliding Mixed-Integer Linear Programming Approach for the Optimization of the Cleaning Schedule of Crude Preheat Trains. *Heat Transfer Engineering*, v. 36, n. 7–8, p. 642–651, 2015.

LI, Y. et al. Formation of gum and deposits in an oxygenated naphtha stream. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING VII, 2007, Santa Fe, New Mexico, USA. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning - Challenges and Opportunities*. Tomar, Portugal: M.R. Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2007.

MACCHIETTO, S. et al. Fouling in Crude Oil Preheat Trains: A Systematic Solution to an Old Problem. Em: Heat Transfer Engineering, Schladming, Austria. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING VIII, 2009, Schladming, Austria. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning VIII*. Schladming, Austria: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2009.

MIRSADRAEE, A.; MALAYERI, M. R. Analysis of highly noisy crude oil fouling data using Kalman filter. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING XI, 2015, Enfield (Dublin), Ireland. *Proceedings*...: *International Conference On Heat Exchanger Fouling And Cleaning XI*. Enfield (Dublin), Ireland: M.R. Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2015.

MOZDIANFARD, M. R.; BEHRANVAND, E. Fouling at Post Desalter and Preflash Drum Heat Exchangers of CDU Preheat Train. *Applied Thermal Engineering*, v. 89, p. 783–794, out. 2015.

OLIVEIRA, J. A.; GAMBETTA, R.; PINTO, J. C. *Pega Ponto Versão 1.0.* Rio de Janeiro: [s.n.], 2006.

PANCHAL, C. B.; LJUBICIC, B. Designing two-phase flow heat exchangers for mitigating fouling. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING VII, 2007, Santa Fe, New Mexico, USA. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning - Challenges and Opportunities*. Tomar, Portugal: M.R. Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2007.

PETKOVIC, B.; WATKINSON, P. Fouling of a Heated Rod in a Stirred Tank System. *Heat Transfer Engineering*, v. 35, n. 3, p. 302–310, 11 fev. 2014.

SCHWAAB, M.; PINTO, J. C. *Análise de Dados Experimentais*: Fundamentos de Estatística e Estimação de Parâmetros. v.1. [S.l: s.n], 2007.

POLLEY, G. T. et al. Evaluation of Laboratory Crude Oil Threshold Fouling Data for Application to Refinery Pre-Heat Trains. *Applied Thermal Engineering*, v. 22, n. 7, p. 777–788, maio 2002.

POLLEY, G. T. et al. Extraction of Crude Oil Fouling Model Parameters from Plant Exchanger Monitoring. *Heat Transfer Engineering*, v. 28, n. 3, p. 185–192, 2007.

RAFEEN, M. S. et al. Crude oil fouling: petronas refineries experience. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING VII, 2007, Santa Fe, New Mexico, USA. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning - Challenges and Opportunities*. Tomar, Portugal: M.R. Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2007.

RATEL, M. et al. Investigation of fouling rates in a heat exchanger using an innovative fouling rig. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING X, 2013, Budapest, Hungary. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning X*. Budapest, Hungary: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2013.

RUSSELL, C. A.; CROZIER, S.; SHARPE, R. Observations from Heavy Residue Pyrolysis: A Novel Method To Characterize Fouling Potential and Assess Antifoulant Additive Performance. *Energy & Fuels*, v. 24, n. 10, p. 5483–5492, 21 out. 2010.

SAHIN, G. et al. Fouling Monitoring in Diesel Unit Preheat Exchangers. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING IX, 2011, Crete Island. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning IX*. Crete Island: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2011.

SALEH, Z. S.; SHEIKHOLESLAMI, R.; WATKINSON, A. P. Fouling Characteristics of a Light Australian Crude Oil. *Heat Transfer Engineering*, v. 26, n. 1, p. 15–22, 2005a.

SALEH, Z.; SHEIKHOLESLAMI, R.; WATKINSON, A. P. Blending effects on fouling of four crude oils. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING V, 2005, Kloster Irsee, Germany. *Proceedings*...: *Heat Exchanger Fouling And Cleaning*. Kloster Irsee, Germany: Hans Müller-Steinhagen, M. Reza Malayeri and A. Paul Watkinson 2005b.

SCHWAAB, M. et al. A New Approach for Sequential Experimental Design for Model Discrimination. *Chemical Engineering Science*, v. 61, n. 17, p. 5791–5806, set. 2006.

SMITH, A. D. Analysis of fouling rate and propensity for eight crude oil samples in annular test section. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING X, 2013, Budapest, Hungary. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning X*. Budapest, Hungary: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2013.

SMITH, A. D. et al. Translating Crude Oil Fouling Testing Rig Data to the Field: A Road Map for Future Research. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING XII, Aranjuez (Madrid), Spain. *Proceedings*... *International Conference On Heat Exchanger Fouling And Cleaning XII* Aranjuez (Madrid), Spain: H. U. Zettler, 2017.

SMITH, A. D.; JOSHI, H. M. A critical look at the use of activation energy in crude oil fouling models. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING XI, 2015, Enfield (Dublin), Ireland. *Proceedings*...: *International Conference On Heat Exchanger Fouling And Cleaning XI*. Enfield (Dublin), Ireland: M.R. Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2015.

SRINIVASAN, M.; WATKINSON, A. P. Fouling of Some Canadian Crude Oils. *Heat Transfer Engineering*, v. 26, n. 1, p. 7–14, 2005.

STEPHENSON, T. et al. Thiophene Mitigates High Temperature Fouling of Metal Surfaces in Oil Refining. *Fuel*, v. 139, p. 411–424, 2015.

TAY, F. H.; KAZARIAN, S. G. Study of Petroleum Heat-Exchanger Deposits with ATR-FTIR Spectroscopic Imaging. *Energy & Fuels*, v. 23, n. 8, p. 4059–4067, 20 ago. 2009.

VENDITTI, S. et al. Developing characterisation methods for foulants deposited in refinery heat exchangers. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING VIII, 2009, Schladming, Austria. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning VIII*. Schladming, Austria: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2009.

WANG, W.; WATKINSON, A. P. Iron Sulphide and Coke Fouling from Sour Oils: Review and Initial Experiments. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING IX, 2011, Crete Island. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning IX*. Crete Island: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2011.

WANG, W.; WATKINSON, A. P. DEPOSITION FROM A SOUR HEAVY OIL UNDER INCIPIENT COKING CONDITIONS: EFFECTS OF SURFACE MATERIAL AND TEMPERATURE. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING X, 2013, Budapest, Hungary. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning X*. Budapest, Hungary: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2013.

WANG, Y. et al. A Review of Experimental Measurement and Prediction Models of Crude Oil Fouling Rate in Crude Refinery Preheat Trains: Measurement and Models of Crude Oil Fouling. *Asia-Pacific Journal of Chemical Engineering*, p. 607-625, 2015.

WATERS, A. J.; AKINRADEWO, C. G.; LAMB, D. Fouling: implementation of a crude preheat train performance monitoring application at the Irving Oil Refinery. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING VIII, 2009, Schladming, Austria. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning VIII*. Schladming, Austria: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2009.

WATKINSON, A. Comparison of Crude Oil Fouling Using Two Different Probes. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING V, 2003, Santa Fe, New Mexico, USA. *Proceedings*...: *Heat Exchanger Fouling And Cleaning: Fundamentals And Applications*. Santa Fe, New Mexico, USA: 2003.

WATKINSON, A. P. Deposition from Crude Oils in Heat Exchangers. *Heat Transfer Engineering*, v. 28, n. 3, p. 177–184, 2007.

WATKINSON, A. P.; FAN, Z.; PETKOVIC, B. Extending fouling concepts from heat exchangers to process equipment. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING IX, 2011, Crete Island. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning IX*. Crete Island: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2011.

WATKINSON, A. P.; LI, Y.-H. Fouling characteristics of a heavy vacuum gas oil in the presence of dissolved oxygen. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING VIII, 2009, Schladming, Austria.

Proceedings...: *International Conference on Heat Exchanger Fouling and Cleaning VIII*. Schladming, Austria: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2009.

WILSON, D. I. et al. Ageing: looking back and looking forward. Em: Proceedings of International Conference on Heat Exchanger Fouling and Cleaning VIII, Schladming, Austria. *Anais*... *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING VIII, 2009, Schladming, Austria. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning VIII*. Schladming, Austria: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2009.

WILSON, D. I.; ISHIYAMA, E. M.; POLLEY, G. T. Twenty Years of Ebert and Panchal—What Next? *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING XI, 2015, Enfield (Dublin), Ireland. *Proceedings*...: *International Conference On Heat Exchanger Fouling And Cleaning XI*. Enfield (Dublin), Ireland: M.R. Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2015.

WILSON, D.; POLLEY, G.; PUGH, S. Ten years of ebert, panchal and the 'threshold fouling' concept. Em: Proceedings of the Heat Exchanger Fouling and Cleaning - Challenges and Opportunities, Kloster Irsee, Germany. *Anais*... Em: EXCHANGER FOULING AND CLEANING VI. Kloster Irsee, Germany: H. Müller-Steinhagen, M. R. Malayeri, and A. P. Watkinson, 2005.

YANG, M. et al. Modelling Fouling Induction Periods. *International Journal of Thermal Sciences*, v. 51, p. 175–183, 2012.

YANG, M. et al. Mitigation of Crude Oil Fouling by Turbulence Enhancement in a Batch Stirred Cell. *Applied Thermal Engineering*, v. 54, n. 2, p. 516–520, 2013.

YANG, M.; O'MEARA, A.; CRITTENDEN, B. D. Determination of crude oil fouling thresholds. *In*: INTERNATIONAL CONFERENCE ON HEAT EXCHANGER FOULING AND CLEANING IX, 2011, Crete Island. *Proceedings*...: *International Conference on Heat Exchanger Fouling and Cleaning IX*. Crete Island: Malayeri, H. Müller-Steinhagen and A.P. Watkinson, 2011.

YEAP, B. L. et al. Mitigation of Crude Oil Refinery Heat Exchanger Fouling Through Retrofits Based on Thermo-Hydraulic Fouling Models. *Chemical Engineering Research and Design*, v. 82, n. 1, p. 53–71, 2004.

YEAP, B. L. et al. Retrofitting Crude Oil Refinery Heat Exchanger Networks to Minimize Fouling While Maximizing Heat Recovery. *Heat Transfer Engineering*, v. 26, n. 1, p. 23–34, 2005.

YOUNG, A. et al. Characterisation of Crude Oils and Their Fouling Deposits Using a Batch Stirred Cell System. *Heat Transfer Engineering*, v. 32, n. 3–4, p. 216–227, 2011.

APPENDIX A – SUMMARY OF DIFFERENT APPROACHES IN THE LITERATURE

Table 20- Summary of information obtained in the literature.

Author	Rf x t	dRf/dt x T	ln(dRf/dt) x 1/T	ln(A) x Ea	Others
Yang et al. (2011)	x	x	x		
Mozdianfard and Behranvand (2015)		x			
Bennett et al. (2009)	x	x	x	x	
Costa et al. (2013)	x				
Snirivasan and Watkinson (2005)	x				dRf/dt x 1000/T
Saleh et al. (2005a)	x				(dRf/dt)V^n p^m x 1000/T
Watkinson (2003)	x				dRf/dt x 1000/T
Polley et al. (2007)	x				
Crittenden et al. (2009)			x	x	
Hong and Watkinson (2009)	x				dRf/dt x 1/T
Young et al. (2011)	x	x	x	x	
Watkinson and Li (2009)	x				dRf/dt x 1000/T
Yang et al. (2012)	x				
Barrie et al. (2013)				x	ln(dRf/dt) x 1000/T
Ishiyama et al. (2011)	x				
Smith (2013)	x		x		
Joshi (2013)	x				
Smith and Joshi (2015)					ln(dRf/dt) x 1000/T
Lane and Harris (2015)	x				
Yang et al. (2013)	x				
Jafari Nars and Majidi Givi (2006b)			x		

Jafari Nars and Majidi Givi (2006c)		x	
Wang et al. (2015)	x		
Assomaning et al. (2000)	x		
Watkinson (2007)	x		dRf/dt x 1000/T
Fan et al. (2010)			ln(dRf/dt) x 1000/T
Wilson et al. (2009)	x		
Saleh et al. (2005b)	x		
Isogai et al. (2003)	x		
Rafeen et al. (2007)	x		
Li et al. (2007)	x		
Andersson et al. (2009)	x		
Ishiyama et al. (2009)	x		
Coletti and Macchietto (2011)	x		
Ishiyama et al. (2013b)	x		
Ishiyama et al. (2013a)	x		
Ishiyama and Pugh (2013)	x		
Coletti et al. (2015)	x		
Diaz-Bejarano et al. (2017)	x		
Ishiyama et al. (2015)	x		
Brignone et al. (2015)	x		

Source: The author, 2019.

APPENDIX B – COMPUTATION OF PROPERTIES OF CRUDE OIL

Procedure for the determination the crude oil physical properties and some correlations necessary for calculations throughout this work.

Specific mass, heat capacity, dynamic viscosity and thermal conductivity (POLLEY et al., 2002):

$$\rho = 917 - 0.833T_c \tag{53}$$

$$C_p = 1940 + 3T_c \tag{54}$$

$$\mu = 0.0985\, e^{\left(\frac{406}{T_c}\right)} \tag{55}$$

$$\lambda = 0.145 - 0.0001T_c \tag{56}$$

Reynolds and Prandtl numbers:

$$Re = \frac{Dh\, v\, \rho}{\mu} \tag{57}$$

$$Pr = \frac{\mu\, Cp}{\lambda} \tag{58}$$

Friction factor for the annular section, Nusselt number from Gnielinski's correlation for the turbulent regime and the heat transfer coefficient:

$$f = \frac{0.178}{Re^{0.1865}} \tag{59}$$

$$Nu = \frac{\left(\frac{f}{8}\right)(Re - 1000)\, Pr}{\left(1 + \left(12.7\left(\frac{f}{8}\right)^{0.5}\left(Pr^{\frac{2}{3}} - 1\right)\right)\right)} \tag{60}$$

$$h = \frac{\lambda\, Nu}{Dh} \tag{61}$$

APPENDIX C – COMPARISON OF THE MODELS OF THE FOULING RATE

Table C 1 – Comparison of the models of the fouling rate used in this work.

	Pol	**NG**	**EPM**	**Ma**	**Wan**	**EPM**	**Flu**	**Yea**
Pol	--	β is a parameter; $-Pr^{-0.33}$ in first term; T_f instead T_w in first term; $Re^{0.4}$ instead $Re^{0.8}$ in second term.	β is a parameter; T_f instead T_w in first term; τ_w instead $Re^{0.8}$ in second term.	$Re^{-0.35}$ instead $Re^{-0.8}$ in first term; T_f instead T_w in first term; $Re^{0.35}$ instead $Re^{0.8}$ in second term.	$Re^{-0.35}$ instead $Re^{-0.8}$ in first term; T_f instead T_w in first term.	β is a parameter; $-Pr^{-0.33}$ in first term; T_f instead T_w in first term; τ_w instead $Re^{0.8}$ in second term.	$-Re^{-0.8}$ and $-Pr^{-0.33}$ in first term; $+\frac{1}{h_i}$ in first term; T_f instead T_w in first term; τ_w instead $Re^{0.8}$ in second term.	$-Re^{-0.8}$ and $-Pr^{-0.33}$ in first term; $+f, v, T_w^{\frac{2}{3}}, \rho^{\frac{2}{3}}, \mu^{-\frac{4}{3}}$ and $+\frac{1}{1+\beta\, v^3 f^2 \rho^{-\frac{1}{3}} \mu^{-\frac{1}{3}} T_w^{\frac{2}{3}}}$ in first term; $v^{0.8}$ instead $Re^{0.8}$ in second term.
NG		--	$+Pr^{-0.33}$ in first term; τ_w instead $Re^{0.4}$ in second term.	β kept constant; $+Pr^{-0.33}$ in first term; $Re^{0.35}$ instead $Re^{0.4}$ in second term.	β kept constant; $+Pr^{-0.33}$ in first term; $Re^{0.8}$ instead $Re^{0.4}$ in second term.	τ_w instead $Re^{0.4}$ in second term.	$-Re^{\beta}$ in first term; $+\frac{1}{h_i}$ in first term; τ_w instead $Re^{0.4}$ in second term.	$-Re^{\beta}$ in first term; $+f, v, T_w^{\frac{2}{3}}, \rho^{\frac{2}{3}}, \mu^{-\frac{4}{3}}$ and $+\frac{1}{1+\beta\, v^3 f^2 \rho^{-\frac{1}{3}} \mu^{-\frac{1}{3}} T_w^{\frac{2}{3}}}$ in first term; T_w instead T_f in first term; $v^{0.8}$ instead $Re^{0.4}$ in second term.

EPM			--	β kept constant; $Re^{0.35}$ instead τ_w in second term.	β kept constant; $Re^{0.8}$ instead τ_w in second term.	$-Pr^{-0.33}$ in first term.	$-Re^{\beta}$ and $-Pr^{-0.33}$ in first term; $+\frac{1}{h_i}$ in first term.	$-Re^{\beta}$ and $-Pr^{-0.33}$ in first term; $+ f, v, T_w^{\frac{2}{3}}, \rho^{\frac{2}{3}}, \mu^{-\frac{4}{3}}$ and $+\frac{1}{1+\beta\, v^3 f^2 \rho^{-\frac{1}{3}} \mu^{-\frac{1}{3}} T_w^{\frac{2}{3}}}$ in first term; T_w instead T_f in first term; $v^{0.8}$ instead τ_w in second term.
Ma				--	$Re^{0.8}$ instead $Re^{0.35}$ in second term.	β is a parameter; $-Pr^{-0.33}$ in first term; τ_w instead $Re^{0.35}$ in second term.	$-Re^{-0.35}$ and $-Pr^{-0.33}$ in first term; $+\frac{1}{h_i}$ in first term; τ_w instead $Re^{0.35}$ in second term.	$-Re^{-0.35}$ and $-Pr^{-0.33}$ in first term; $+ f, v, T_w^{\frac{2}{3}}, \rho^{\frac{2}{3}}, \mu^{-\frac{4}{3}}$ and $+\frac{1}{1+\beta\, v^3 f^2 \rho^{-\frac{1}{3}} \mu^{-\frac{1}{3}} T_w^{\frac{2}{3}}}$ in first term; T_w instead T_f in first term; $v^{0.8}$ instead $Re^{0.35}$ in second term.

Wan					--	β is a parameter; $-Pr^{-0.33}$ in first term; τ_w instead $Re^{0.8}$ in second term.	$-Re^{-0.35}$ and $-Pr^{-0.33}$ in first term; $+\frac{1}{h_i}$ in first term; τ_w instead $Re^{0.8}$ in second term.	$-Re^{-0.35}$ and $-Pr^{-0.33}$ in first term; $+f, v, T_w^{\frac{2}{3}}, \rho^{\frac{2}{3}}, \mu^{-\frac{4}{3}}$ and $+\frac{1}{1+\beta\, v^3 f^2 \rho^{-\frac{1}{3}} \mu^{-\frac{1}{3}} T_w^{\frac{2}{3}}}$ in first term; T_w instead T_f in first term; $v^{0.8}$ instead $Re^{0.8}$ in second term.
EPM						--	$-Re^{\beta}$ in first term; $+\frac{1}{h_i}$ in first term.	$-Re^{\beta}$ in first term; $+f, v, T_w^{\frac{2}{3}}, \rho^{\frac{2}{3}}, \mu^{-\frac{4}{3}}$ and $+\frac{1}{1+\beta\, v^3 f^2 \rho^{-\frac{1}{3}} \mu^{-\frac{1}{3}} T_w^{\frac{2}{3}}}$ in first term; T_w instead T_f in first term; $v^{0.8}$ instead τ_w in second term.

Flu							--	$-\frac{1}{h_i}$ in first term; $+ f, v, T_w^{\frac{2}{3}}, \rho^{\frac{2}{3}}, \mu^{-\frac{4}{3}}$ and $+\frac{1}{1+\beta\, v^3 f^2 \rho^{-\frac{1}{3}} \mu^{-\frac{1}{3}} T_w^{\frac{2}{3}}}$ in first term; T_w instead T_f in first term; $v^{0.8}$ instead τ_w in second term.
Yea								--

Source: The author, 2019.