

## FOULING IN THE CRUDE OIL DISTILLATION PREHEAT TRAIN: COMPARISON OF EXPERIMENTAL DATA WITH MODEL RESULTS

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### ABSTRACT

Losses due to heat exchanger fouling in the pre-heat train of the distillation unit are among the most important issues in the oil refining industry. Consequences are both economic and environmental: fouling increases both production and maintenance costs. Moreover it causes additional greenhouse gas. Hence, a better knowledge and accuracy of fouling modeling is needed in order to improve heat exchanger designs and network optimization.

The study compares crude oil fouling data obtained from several literature models to those obtained experimentally with a fouling rig which reproduces industrial operating conditions. The latter concerns at a small scale, the last heat exchanger of the preheat train located directly upstream of the furnace. Process fluids as well as the temperature level, the heat flux, the shear stresses and the fluid velocities are similar to industrial conditions.

Several empirical models are checked. Discrepancies between calculations and experimental values are analyzed to identify advantages and weaknesses of each model. Optimized parameters, identified from our experimental results, are finally proposed.

### INTRODUCTION

In an oil refinery, approximately 4 % of the energy contained in the crude is consumed by the furnace upstream of the atmospheric distillation column which is used to regulate the inlet temperature (Yeap et al., 2004). A network of heat exchangers – the preheat train – is used to reduce the amount of energy needed by recovering as much as possible the heat contained in the hot products of the distillation column. An efficient preheat train can recover up to 60 to 70 % of the energy needed for distillation (Yeap et al., 2004).

However, with time, unwanted materials (fouling) deposit on the heat transfer surfaces, affecting the heat transfer and fluid flow. Chemical reactions triggered by high temperatures are suspected to be responsible of fouling because it is particularly pronounced at the hot end of the preheat train. Throughout the train, a fall of 8 to 11 °C of the outlet pre-heated crude oil temperature per year is generally observed (Coletti and Macchietto, 2011). To maintain a constant temperature at the inlet of the

distillation column, the temperature drop is countered by burning extra fuel in the furnace.

This has not a negative effect on energy costs only but also has an environmental impact linked to additional emissions of greenhouse gases in the atmosphere (Müller-Steinhagen et al., 2009). A study by the US Department of Energy claims that reducing fouling in preheat trains and furnaces could lead to a saving of 15 % fuel oil (DOE, 2006).

Fouling in the preheat train is still an unsolved problem for decades (Taborek et al., 1972) since fouling mechanisms by oil remain unknown (Watkinson and Wilson 1997; Watkinson, 2007; Bennett et al., 2009).

Traditional methods of heat exchanger design based on empirical fouling factors of TEMA (1968) are not quite suitable (Chenoweth, 1995; Rabas and Panchal, 2000; Bennett et al., 2007). Indeed, they fail to correctly predict its dependence regarding of the operating conditions and crude oil composition.

To overcome the limitations imposed by the fouling factors, more complex models have been developed to describe fouling resistance by correlations depending on the operating conditions.

After reviewing the main fouling models of literature, the purpose of this article is to compare experimental data from an experimental fouling rig with models. The rig reproduces, the last and hottest heat exchanger of the preheat train located directly upstream of the furnace. The objective is to identify strengths and weaknesses of each model. The later perspective is to formulate an analytical model to predict fouling behavior according to operating conditions and heat exchanger geometry.

### FOULING MODELS

Several semi-empirical models have been established to express fouling rates based on measurable operating parameters. They are often based on experimental results from fouling rigs. In all cases, the models describe fouling in the inner portion of the tubes of shell-and-tube exchangers.

Except for the last model, the fouling rate  $dR_f/dt$  of a heat exchanger can be modeled as a dynamic phenomenon (Kern and Seaton, 1959) where the deposited flow  $\Phi_d$

competes with the removal or suppression (see Wilson et al., 2005) flow  $\Phi_r$  (Eq. 1):

$$\frac{dR_f}{dt} = \Phi_d - \Phi_r \quad (1)$$

**Ebert and Panchal (1995) model (Eq. 2)**

$$\frac{dR_f}{dt} = \alpha Re^\beta \exp\left(-\frac{E_a}{RT_f}\right) - \gamma \tau \quad (2)$$

Re is the Reynolds number, R the gas constant.  $\alpha$ ,  $\beta$  and  $\gamma$  are constants from experiments.  $E_a$  is the activation energy and  $\tau$  the wall shear stress.  $T_f$  is used here to denote the film temperature. This is determined as follows (Eq. 3):

$$T_f = T_b + 0.55(T_w - T_b) \quad (3)$$

$T_w$  is the wall temperature and  $T_b$ , the bulk temperature. Based on the experimental data of Scarborough et al. (1979), the authors identified the following values:  $\alpha = 8.39 \text{ m}^2 \text{ K/J}$ ;  $\beta = -0.88$ ;  $\gamma = 4.03 \cdot 10^{-11} \text{ m}^2 \text{ K/J/Pa}$  and  $E_a = 68 \text{ kJ/mol}$ .

This model was developed using crude oil fouling data inside tubes of an industrial furnace. These values were obtained for only one crude and film temperatures between 360 and 420 °C. The model does not consider the change of the fluid properties regarding temperature or diversity of crude oil used in refineries.

**Ebert et al. (1999) model**

The authors modified the correlation of 1995 using the Prandtl number which can take into account the variations in the thicknesses of the hydraulic and thermal boundary layers during temperature changes. The proposed correlation has been established from five studies on experimental rigs (annular and tube flows) and three different types of crude from different origins. This new correlation is defined by Eq. 4:

$$\frac{dR_f}{dt} = \alpha Re^\beta Pr^{-0.33} \exp\left(-\frac{E_a}{RT_f}\right) - \gamma \tau \quad (4)$$

The optimum parameters obtained by Ebert et al. (1999) are:  $\alpha = 1.4 \cdot 10^{-2} \text{ m}^2 \text{ K/J}$ ;  $\beta = -0.66$ ;  $\gamma = 4.03 \cdot 10^{-11} \text{ m}^2 \text{ K/J/Pa}$  and  $E_a = 48 \text{ kJ/mol}$ .

This model is currently the most commonly used for industrial applications.

**Polley et al. (2010) model**

This model was developed considering that asphaltene precipitation plays a leading role in crude oil fouling. The model is given by Eq. 5:

$$\frac{dR_f}{dt} = \frac{K}{h_i} \exp\left(-\frac{E_a}{RT_f}\right) S \quad (5)$$

The constant K is a function of crude composition, temperature and geometry of the heat exchanger.  $h_i$  is the convective heat transfer coefficient inside the tubes and S is the probability of adhesion of the particles to the heat transfer surfaces. It varies between 0 and 1 and is calculated using Eq. 6 where j is an experimental constant:

$$S = 1 - \left(\frac{\tau - 2}{98}\right)^j \quad (6)$$

For this model, the authors revisit the concept of deposit removal: fouling is not a dynamic phenomenon which the deposition rate competes with the removal rate. They now consider that the thickness of the deposit depends on the ability of the precursors to reach the heat transfer surfaces. For that they introduce a probability term. The greater is the wall shear stress, the lower is the probability that asphaltene particles attach to the wall.

The experimental data used for this model are those from Ebert et al. (1999) and are completed with data collected in the refinery. Hence, the K parameter is assessed in the range  $2.78 \cdot 10^{-4}$  to  $0.11 \text{ s}^{-1}$ . Using Knudsen et al. (1999) data, parameters are:  $E_a = 44.3 \text{ kJ/mol}$ ;  $j = 0.5$  and  $K = 6.94 \cdot 10^{-2} \text{ s}^{-1}$ . These values will be used further and is named "initial model".

Some additional models which have not been considered in this paper exist. The Crittenden et al. (1992) model will not be compared because it does not consider the deposit removal. Models of Saleh et al. (2005) and Nasr and Givi (2006) are only applicable to laminar flows. As for the Epstein (1994) model, it only provides with the initial fouling rate from data that cannot be determined in this study.

## EXPERIMENTAL SETUP

### The fouling rig

The fouling rig described by Ratel et al. (2013) was designed to reproduce the operating conditions encountered at the end of refineries preheat train where fouling is promoted by high wall temperatures. This is a cascade loop which consists of five circuits (Fig. 1):

- Two closed circuits – each filled with approximately  $0.25 \text{ m}^3$  of fouling fluids – which exchange heat through the test section. The crude oil circuit (#3) plays the role of cold fluid and the atmospheric residue circuit (#2) is used as the hot fluid. These two circuits are designed and instrumented the same way. Two crudes (named Crude A and Crude B) are used for the experiments.
- Two closed circuits of utilities (#1 and #4) filled with THERMINOL® 72 in order to bring or to remove the heat to the previous circuits. Heat input to the loop is provided by a heater located on the circuit #1. It

regulates the THERMINOL® 72 output temperature so that it corresponds to the fixed set point. The set point and the flow rate are adjusted to achieve the desired temperature of crude at the test section inlet (from 200 to 300 °C). The duty is then transferred throughout the four heat exchangers of the rig, except the heat losses.

- A cooling circuit (#5) supplied with cooling water.

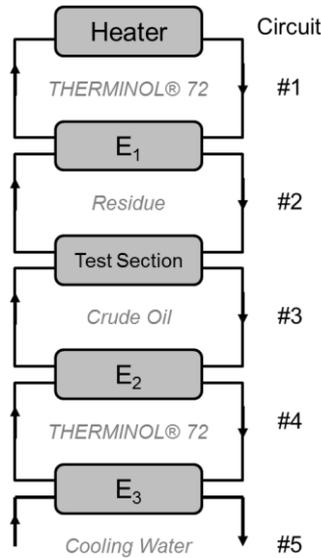


Fig. 1 Process flow diagram of the test rig.

The test section is a shell-and-tube heat exchanger because it is the most frequently encountered in refinery preheats trains. It is designed according to TEMA (1968) standards. The test section is designed to be as representative as possible of the heat exchangers used in the refinery. The heat flux, the shear stress and temperature levels are kept at the same value, as well as the tube diameter, thickness and materials. This small scale heat exchanger offers the same ranges of overall and local heat transfer coefficients than industrial heat exchangers and consequently shows the same film temperature levels.

The use of crude oil and residue in the test section and the same temperature, pressure and flow velocity ranges that those found at the end of preheating process make it possible to study fouling under representative industrial operating conditions.

To limit the fouling by autoxidation, all circuits are pressurized with nitrogen. The pressure is known as a parameter that can potentially influence fouling so the same pressure is hold in each circuit for the duration of the tests.

The operating conditions for data collection were chosen according to the semi-empirical model of Ebert et al. (1999). As shown in Fig. 2, experimental data are placed on either side of the threshold curve in order to characterize the behavior of the heat exchanger regarding the fouling.

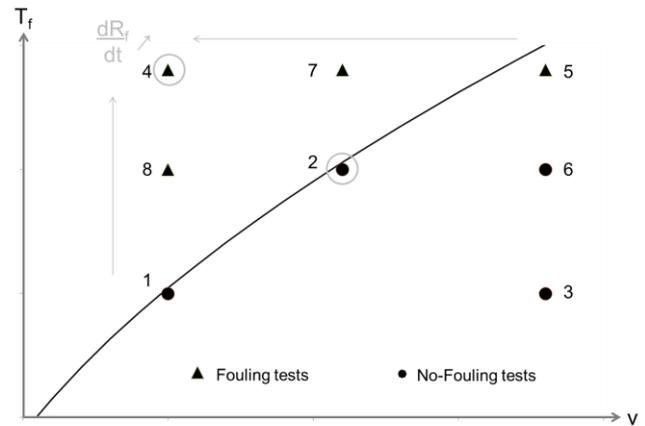


Fig. 2 Experimental plan and qualitative test results on the shell-and-tube heat exchanger (Crude A); circled tests were carried out with the two crudes.

Flow rates, temperatures, absolute and differential pressures are measured on each circuit. Temperature measurements are performed with Platinum probes (Pt100), except in the cooling circuit. The use of platinum probes provides higher measurement accuracy than those obtained with the thermocouples. These probes have, in addition, improved stability over time. To ensure the accuracy of these measurements, the calibration of these sensors is carried out before each test campaign.

All the circuits are insulated with Rockwool and Isolalu in order to limit heat losses.

### Fouling measurements

The setup of fouling in a heat exchanger results in the creation of an additional thermal resistance due to deposition of fouling materials on the heat transfer surfaces. This parameter depends on the fluid, the operating conditions and the geometry of the heat exchanger. It is calculated using Eq. 7:

$$R_f(t) = \frac{1}{U(t)} - \frac{1}{U(0)} \quad (7)$$

Wherein  $R_f$  corresponds to the fouling resistance,  $U(0)$  to the overall heat transfer coefficient at the initial time – i.e. at the beginning of a test – and  $U(t)$ , the overall heat transfer coefficient at the time  $t$ .

The continuous monitoring of temperatures and therefore overall heat transfer coefficient in the test section allows to detect the fouling resistance.

If fouling occurs, the overall heat transfer coefficient  $U(t)$  decreases with time as shown in Fig. 3. The value of the overall heat transfer coefficient at the initial time  $U(0)$  is determined by averaging the values of  $U(t)$  obtained in the  $U = f(t)$  curve plateau, during the stabilization phase and before the decrease of the heat transfer coefficient due to fouling (Fig. 3).

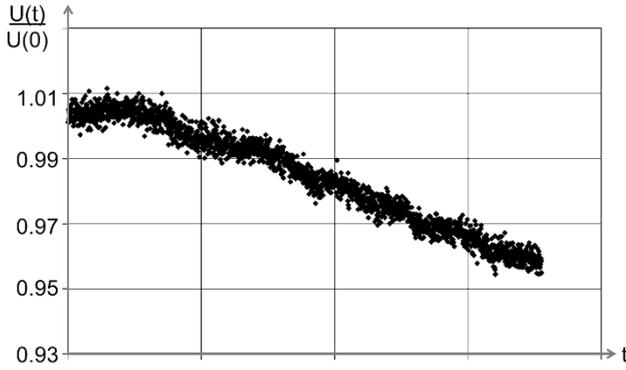


Fig. 3 Typical evolution of the overall heat transfer coefficient observed on the rig during a fouling test.

Finally, the fouling rate is calculated by the expression (Eq. 8):

$$\frac{dR_f}{dt} = \frac{1}{U(t_2)} - \frac{1}{U(t_1)} \quad (8)$$

$U(t_2)$  and  $U(t_1)$  are the overall heat transfer coefficient respectively calculated at the time instants  $t_2$  and  $t_1 < t_2$ .

The value of the overall heat transfer coefficient at the initial time  $U(0)$  determined experimentally has no influence on the calculation of the fouling rate.

## EXPERIMENTAL RESULTS

Ten tests were conducted with two different crudes (Crude A and Crude B) flowing through the tubes of the shell-and-tube heat exchanger.

The test with theoretically the highest fouling potential in the experimental plan (test #4 in Fig. 2), was carried out with the two crude oils. A higher fouling rate was detected during the Crude A test. This test confirms that the Crude A promotes fouling compared to the Crude B and shows that the rig is sensitive to changes in operating conditions as discussed previously by Ratel et al. (2013).

Crude A was therefore used for the subsequent tests, in order to amplify the fouling mechanisms.

Four tests (triangular shape in Fig. 2) led to detect a fouling resistance.

The comparison of fouling rates observed in these tests is consistent with trends of the literature. They are in the same order of magnitude ( $10^{-10} \text{ m}^2 \text{ K/J}$ ) than value observed by Coletti and Macchietto (2009) at the end of the preheat train of an ExxonMobil refinery and by Coletti et al. (2010). They are, moreover, comparable to those measured in French refineries. This observation shows that the rig is sensitive to changes in operating conditions.

Moreover, it was confirmed as expected that fouling increases when the temperature increases and when the fluid velocity decreases.

## DISCUSSION

### Comparison with literature models

Experimental results from the rig are compared with previously described literature models.

In order to identify the models which give results close to experimental values, models of Ebert and Panchal (1995 and 1999) and Polley et al. (2010) were plotted on the graph of Fig. 4. The curve of the Polley et al. (2010) model cannot be drawn for a zero fouling rate. Hence, all the curves are plotted for a fouling rate corresponding to the minimum fouling rate measured on the rig.

The Ebert and Panchal (1995) model seems to better represent the boundary between the fouling and non-fouling zones. To define their model, the authors used a medium density crude oil similar to the oils used for our experiments. This could explain the small discrepancy with the experimental results.

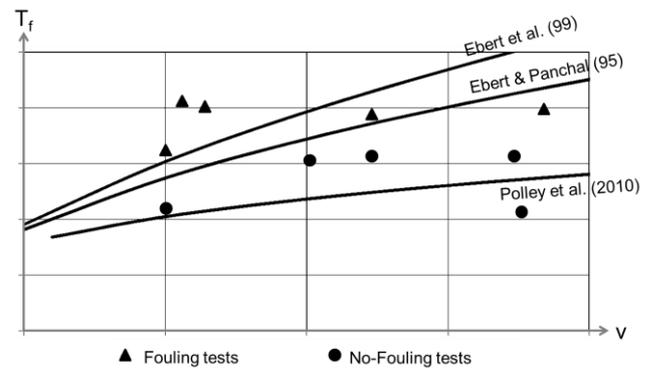


Fig. 4 Comparison of experimental results with fouling models; theoretically, the fouling points must be above the curve and no-fouling points, under or near the curve.

The Polley et al. (2010) model do not allow to separate the non-fouling area from the fouling area because most non-fouling points are located in the theoretical fouling zone.

A comparison was made for each model, between the experimental temperatures (film or wall depending on the model) and those predicted by the models. For each test, the film (or wall) temperature is calculated using the model and the experimental fouling resistance as illustrated by Eq. 9 for the Ebert et al. (1999) model.

$$T_f = \frac{-E_a}{R \ln \left( \frac{\frac{dR_f}{dt} + \gamma\tau}{\alpha Re^{-0.66} Pr^{-0.33}} \right)} \quad (9)$$

This value is then compared to the experimental temperature. The relative difference between these two temperatures is plotted for each test on Fig. 5.

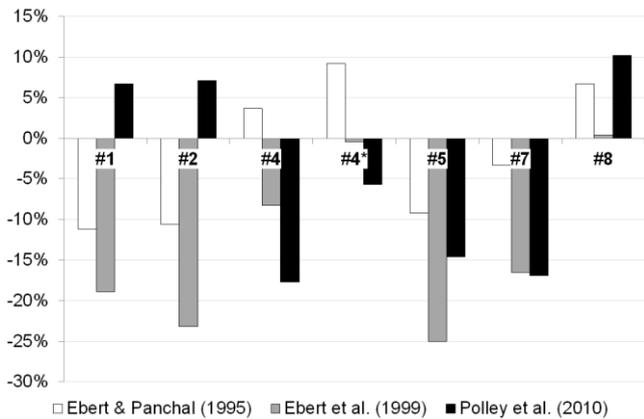


Fig. 5 Comparison between model predictions and rig values for Crude A (#4\* refers to Crude B).

In this column chart, a percentage of negative deviation means that the film temperature predicted by the model is greater than the experimental film temperature, for a same value of fouling rate.

Among the studied models, the Polley et al. (2010) correlation is the only one established from refinery data. Fouling of both sides of the heat exchanger is then predicted, as it is the case in the rig. Closer results to experimental values are therefore expected. The deviation between predicted and experimental film temperatures is indeed less than 20 %.

The Ebert et al. (1999) model tends to overestimate the film temperature values (negative deviation). The predicted fouling rate is then underestimated by the model. Most of the differences are around 20 %. This model has been established for fouling in the tubes solely. Our experimental values used for the comparison are higher than the predicted values since fouling occurred on the both sides of the heat exchanger.

The Ebert and Panchal (1995) model predicts the film temperature with a deviation of less than 20 % for all tests. However, this correlation was established from a single medium density crude. It does not use the Prandtl number, which takes into account the evolution of the boundary layers which differs depending on whether the crude is heavy, light or medium. Thus, this model is applicable for medium crudes only, such as Crude A and Crude B; it could be less accurate to provide a fouling rate for light and heavy crude but this parameter was not investigated in this study.

## Model parameters estimation

### *Ebert et al. (1999) model*

This model was developed from experimental data where only tubes undergo fouling whereas fouling was set up on both sides of the shell-and-tube heat exchanger tested

on the rig. Model parameters must be adapted to represent this fouling due to both crude and residue.

According to Bories and Patureaux (2003), the parameter  $\alpha$  depends on the fluid and the parameter  $\gamma$  relates to the heat exchanger geometry. The activation energy can also be adapted. It depends on the reactions that occur during the fouling phenomenon, that is to say, the crude origin and composition.

The parameters optimization was performed using the least squares method on fouling rates, so that the predictions of this model are as close as possible to the experimental results.

The graph in Fig. 6 shows the percentage of deviation between the experimental values and the predictions of the Ebert et al. (1999) model, with the initial and optimized parameters. This comparison is performed between experimental film temperature values and the values predicted by the model using the experimental fouling rates.

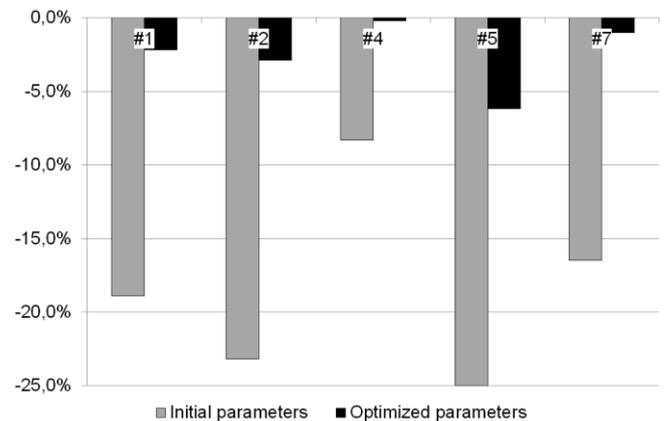


Fig. 6 Ebert et al. (1999) model with optimized parameters compared to the experimental values.

The parameters optimization significantly reduces the deviation between model and experimental results for all tests. Indeed, the average difference between film temperatures predicted by the model and the experimental values is initially 20 %; once optimized, it is no more than 2 %. The initial model overestimates the film temperature values (negative deviation) more than the modified model. For the same temperature, the prediction of fouling rates is therefore more accurate after optimization.

Figure 7 shows the experimental fouling rates, and those predicted by both the initial model and optimized model. Uncertainty bars on the experimental values appears on this graph. Apart from the test #5, the fouling rates predicted by the optimized model are included in the error bars.

### *Polley et al. (2010) model*

Parameter  $K$  of Polley et al. (2010) model depends on the heat exchanger geometry. For the shell-and-tube heat exchanger of the rig, this parameter was assessed to

$3.61 \cdot 10^{-3} \text{ s}^{-1}$ . The activation energy can also be changed; it depends on the crude origin. Data from a refinery preheat train were used for this model. Fouling due to the two fluids circulating in the heat exchanger is modeled.

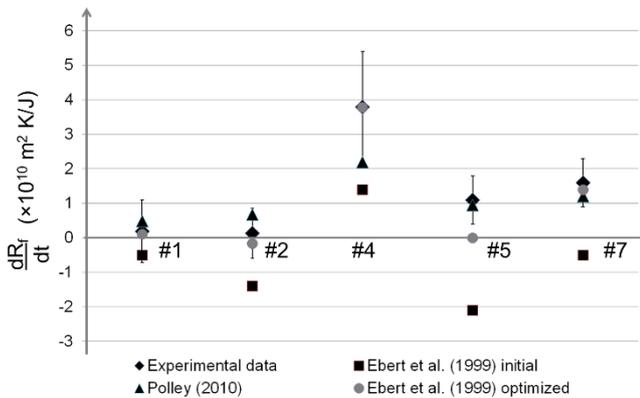


Fig. 7 Comparison of fouling rates predicted by the Ebert et al. (1999) and Polley (2010) models with experimental values.

The graph on Fig. 7 compares the experimental fouling rates with the predictions of the initial model. The values predicted by the initial model are within the error bars of the experimental results. Unlike the Ebert et al. (1999) model, a parameter optimization is not necessary. This model has been obtained from refinery values, considering fouling due to both crude and residue. Hence, it is consistent with the fact that predicted fouling rates are comparable with the experimental values. The results also confirm that the values obtained on the rig are close to the values measured in the refinery.

## CONCLUSIONS

1. The shell-and tube heat exchanger used in the test section reproduces the behavior of the one located at the end of refinery preheat train;
2. The Polley et al. (2010) model predicts the film temperatures and thus fouling rate with better accuracy. This is consistent with the fact that this model was established from refinery data for fouling from both sides of the exchanger. However, the feedback is low and adaptation of the  $K$  parameter to the heat exchangers geometry is not specified in the literature;
3. Once optimized, most of the values predicted by the Ebert et al. (1999) model are close to the experimental values and included in the measurement uncertainties. The optimization can consider fouling by the residue as the initial model considers only a fouling due to the crude.
4. This study point out that models prediction capability is limited to experimental devices that have produced data. A better understanding of the impact of the crude origin and heat exchangers geometry on fouling and therefore on model parameters will be interesting to perform an analytical model.

The next step of this work is to study the impact of geometry on the fouling trends. A shell-and-tube heat exchanger designed according to Bennett and Nesta (2004) recommendations (no-foul method) is being tested and will be compared to existing models.

## NOMENCLATURE

|       |   |
|-------|---|
| $E_a$ | activation energy, J/mol  |
| $h$   | heat transfer coefficient, $\text{W}/\text{m}^2/\text{K}$         |
| $j$   | model constant, dimensionless                                     |
| $K$   | model constant, $\text{s}^{-1}$                                   |
| $Pr$  | Prandtl number, dimensionless                                     |
| $R$   | gas constant, $\text{J}/\text{mol}/\text{K}$                      |
| $Re$  | Reynolds number, dimensionless                                    |
| $R_f$ | fouling resistance, $\text{m}^2 \text{K}/\text{W}$                |
| $S$   | asphaltene adhesion probability, dimensionless                    |
| $T$   | temperature, $\text{K}$   |
| $t$   | time, $\text{s}$  |
| $U$   | overall heat transfer coefficient, $\text{W}/\text{m}^2/\text{K}$ |
| $v$   | fluid velocity, $\text{m}/\text{s}$                               |

## Greek

|           |  |
|-----------|--|
| $\alpha$  | model constant, $\text{m}^2 \text{K}/\text{J}$           |
| $\beta$   | model constant, dimensionless                            |
| $\gamma$  | model constant, $\text{m}^2 \text{K}/\text{J}/\text{Pa}$ |
| $\gamma'$ | model constant, $\text{m}^2 \text{K}/\text{J}$           |
| $\tau$    | wall shear stress, $\text{Pa}$                           |
| $\Phi$    | specific mass flow, $\text{m}^2 \text{K}/\text{J}$       |

## Subscript

|     |            |
|-----|------------|
| $b$ | bulk       |
| $d$ | deposition |
| $f$ | film       |
| $i$ | inner      |
| $r$ | removal    |
| $w$ | wall       |

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