CRUDE OIL FOULING DEPOSITION, SUPPRESSION, REMOVAL - AND HOW TO TELL THE DIFFERENCE

E. Diaz-Bejarano[1], F. Coletti[2] and S. Macchietto[1,2]

[1]Department of Chemical Engineering, Imperial College London, South Kensington Campus, London SW7 2AZ, UK
[2]Hexxcell Ltd., Imperial College Incubator, Bessemer Building Level 2, Imperial College London, London SW7 2AZ, UK,
s.macchietto@imperial.ac.uk

ABSTRACT

Crude oil fouling formation on heat transfer surfaces is often described as the result of two competing mechanisms: a deposition mechanism brings fouling species onto heat transfer surfaces and a deposition-limiting mechanism. There is uncertainty whether suppression (due inhibition of attachment or re-entrainment of foulant from the near wall region into the bulk) or removal of foulant already deposited is the overarching mechanism offsetting deposition. This is due not only to difficulties in experimentally identifying and isolating the key phenomena but also to the fact that effects are typically measured by monitoring thermal exchange rates (or resistance) alone.

In this paper, the question is addressed of whether it is conceptually possible to distinguish such phenomena, and if so, in which conditions. A recently developed deposit layer model and thermo-hydraulic model of a tube undergoing crude oil fouling are used to assess the response of the system when removal, suppression, ageing and consolidation are considered. It is shown that whilst suppression or removal lead to undistinguishable behavior during overall deposit growth, thermal and hydraulic responses will differ in certain conditions, for which an experimental procedure is suggested. Simultaneous consideration of thermal and hydraulic effects and accurate characterization of the deposit ageing and consolidation processes may allow one to unambiguously identify the dominant deposition-limiting mechanism.

INTRODUCTION

Crude oil fouling in the preheat train of refineries leads to significant costs and fuel consumption (Coletti et al. 2015). The complexity of crude oil composition and variation between feedstock makes it difficult to study fouling from a fully mechanistic approach. The main dependences of organic matter deposition on operating conditions are well known: fouling rate increases with temperature (thermal fouling) and decreases with flow velocity (or shear stress). The latter suggests the existence of a deposition-limiting mechanism related to mass transfer, shear forces or turbulence. A general approach, first introduced by Kern and Seaton (1959) is to quantify fouling rate as a competition between deposition and removal. The 'removal' term was initially introduced to explain the falling rate of fouling resistance (R_f) with time. Kern and Seaton assumed that the deposit is removed in chunks by effect of the shear action of the fluid (also called spalling or tearing off). Other removal mechanisms are thought to exist including dissolution or erosion (deposit finely removed by shear action) (Bohnet 1987; Somerscales and Sanatgar 1989; Bohnet et al. 1997). The dominant mechanism depends on the specific system under study.

Epstein (1983) noted that asymptotic or falling fouling rate data does not necessarily imply a partial removal of the deposit. He listed alternative mechanisms including the suppression of attachment by increasing flow velocity as a result of flow area blockage, the reduced transport due to formation of a thicker viscous sub-layer due to smoothing of the deposit surface, or the gradual weakening of wall catalysis effect as the deposit builds up. Epstein cites instances of fouling studies in different systems where no deposit removal has been proved, and others where removal has been observed.

In crude oil fouling, two main deposition-limiting mechanisms are generally acknowledged to be possible:

1. Suppression: inhibition of deposition by back diffusion of foulant, formed in the boundary by chemical reaction, to the bulk (Fig. 1(a)) or inhibition of deposition of particles by fluid dynamics or shear (Fig. 1(b)).
2. Removal: erosion/tearing off of the deposit by action of the shear stress, if the deposit is weak (Fig. 1(c)).

The complexity in separately characterizing deposition and deposition-limiting mechanisms led to the development of semi-empirical models to quantify a thermal fouling rate. The most popular type is the so called 'threshold' model, first proposed by (Ebert and Panchal 1995):

$$\frac{dR_f}{dt} = \alpha \, \text{Re}^\beta \exp\left(-\frac{E_f}{R_g T_{film}}\right) - \gamma \tau_w \qquad (1)$$

This semi-empirical approach lumps together physical and chemical phenomena involved in deposition using Re, Pr (in following modifications of the same equation) and some adjustable parameters (α, β, γ and activation energy, E_f). The deposition-limiting term (negative term on the right-hand side of Eqn. (1)) is assumed proportional to the

wall shear stress (τ_w). Ebert and Panchal (1995) defined it as the removal of foulant by diffusion or turbulent eddies from the thermal boundary layer to the bulk, therefore, as a suppression mechanism. These models are used to: a) Fit initial fouling rates to experimental data; and b) fit historical plant data and predict fouling. In design, the objective is to find operating conditions that prevent fouling. The existence of this threshold was proved experimentally (Knudsen et al. 1999).
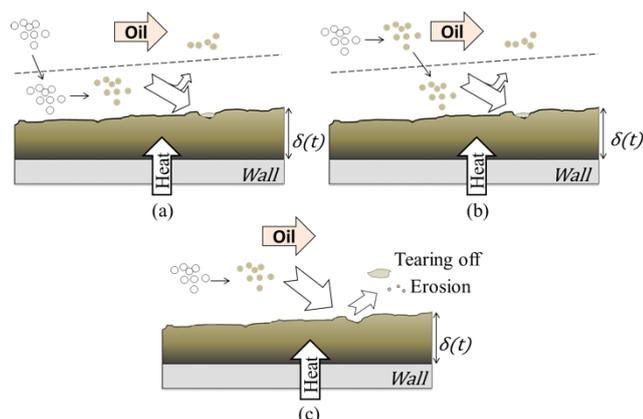

Fig. 1 Schematic representation of suppression (deposition by chemical reaction (a) and particles (b)) and removal (c). Ageing is shown as a gradual darkening. The dashed line indicates the limit of the thermal boundary layer (as in Panchal and Watkinson 1994).

The debate regarding the mechanism limiting deposition in crude oil thermal fouling remains open. This is mainly a consequence of the lack of conclusive experimental proof. Crittenden et al. (1992) referred to trends in plant data and in situ observations that might suggest a removal mechanism: i) saw-tooth pattern of measured fouling resistance over time; ii) scatter in R_f versus time graphs potentially due to removal and re-deposition; and iii) observation of deposits accumulated at locations apart from the inside of the tube by visual inspection of heat exchangers open for cleaning. More recently Crittenden and collaborators reported negative fouling rates observed in a Batch Stirred Cell System (Young et al. 2011; Yang et al. 2011) in experiments alternating low stirrer speed with high speed periods. This has been interpreted by the authors as experimental evidence of partial removal of the fouling deposit. The authors acknowledge that a slower mass transfer mechanism might still exist, although shear removal would be dominant.

According to Watkinson and Wilson (1997) removal is usually less significant in organic systems than in other types of fouling. Wilson *et al.* (2005) referred to the lack of "evidence of deposit removal in crude oil fouling systems" to support suppression as the deposition-limiting mechanism, although the authors acknowledge that the mechanisms are not well understood and emphasize the importance of considering ageing (Wilson et al. 2009). The ageing process is considered to gradually change the initial fouling deposit (tarry or gel-like in consistency) to coke at high temperature. Ageing in crude oil fouling is believed to strengthen the deposits, as happens in polymerization systems, leading to the implicit conclusion that the removal becomes moredifficult, thus suppression is the more likely deposition-limiting mechanism.

A kinetic model of the thermal effect of ageing was postulated and implemented into lumped (Ishiyama et al. 2010) and distributed (Coletti et al. 2010) layer models and used to test the effect of ageing under industrially relevant conditions. This showed that ageing improves the thermal conductivity of the layer and contributes to an "apparent" falling rate and asymptotic time profile of the fouling thermal resistance. Consequently, both ageing and deposition-limiting (suppression and/or removal) mechanisms should be considered when studying crude oil fouling. However, ageing is often ignored in most fouling studies, which tend to focus on fitting threshold models to temperature and flow data. Estimation of fouling parameters considering ageing is limited to Coletti and Macchietto (2011).

Diaz-Bejarano et al. (2015) presented an extended representation and dynamic model of a fouling layer characterized by a single, continuous 2D distribution of compositions and other properties, and showed how it could be used to model deposition, cleaning (including condition-based cleaning) and any transitions between them with a single model. The model permits capturing the simultaneous thermal effects from changes in the deposit thickness and composition, and hydraulic effects resulting from flow area restriction. Here, the above models are used to investigate the effects previously described (suppression, removal and ageing) on the thermal-hydraulic response of a tube undergoing fouling, focusing on developed fouling layers rather than initial rates. Industrially relevant conditions and time scales are considered. The objective is to investigate whether it is conceptually possible to observe, measure and distinguish such phenomena, and if so, in which conditions.

APPROACH

A single heat exchanger tube is considered. The model comprises three domains: Tube-side, tube wall and fouling layer. The models and solution method are given elsewhere (Diaz-Bejarano et al. 2015) and are not reported here. The model is dynamic and distributed in space and evaluates fouling as function of local conditions.

This paper focuses on thermal fouling of organic materials, with the layer modelled as a two pseudo-components system: fresh material, or 'gel', and aged deposit, or 'coke'. The fresh deposit is assumed to be entirely composed of gel while coke is exclusively formed in-situ from the gel following a first order kinetic model. The thermal conductivity at each point in the layer is affected by the transformation of gel to coke (hence by the composition/temperature history at each point).

In a general formulation, the change in thickness is given by the contribution of deposition, suppression and removal, if all of them are well defined. Here we are interested in testing the deposition-limiting processes independently (assuming that one of them is clearly dominant).

First, we need to define the situation we wish to represent. It is assumed that a suppression mechanism occurs in the oil phase, before the foulant has settled on the surface, hence it does not interact with the deposit itself. If deposition exceeds suppression, material builds up on the surface contributing to the layer growth. Once deposited, the matter remains undisturbed by the fluid. If suppression equals or exceeds deposition, the layer stops growing (all approaching foulant is repelled to the bulk before attaching). The properties of the layer (even at the surface) are then only affected by thermal processes or reactions such as ageing. The local change in deposit thickness is:

$$\rho_l \frac{d\delta_l}{dt} = \max(0, n_{d,gel} - n_{s,gel}) \qquad (2)$$

where $n_{d,gel}$ and n_s are the deposition and suppression rates (mass fluxes) of gel, δ_l is the deposit thickness and ρ_{gel} is the density of gel. Negative fouling rates are not allowed.

A removal mechanism disrupts the deposit to a certain depth of the layer, depending on the shear forces, the properties of the layer and type of removal (e.g. a tearing off mechanism may affect greater portions of the layer than erosion, which would be rather superficial). As a result of the continuous deposition-removal, that portion of the layer is continuously renewed. Here, it is assumed that the depth of influence is small, i.e. the effect of removal is superficial (erosion), and removal can be simply captured as a boundary condition for the deposit layer. The local change in thickness is:

$$\rho_l \frac{d\delta_l}{dt} = n_{d,gel} - n_r \qquad (3)$$

where n_r is the removal rate (mass flux) and ρ_L is the density of the layer at the surface. The thickness of the layer may increase or decrease. If the thickness decreases, the net effect is that the top layer disappears and the underlying matter (with its own concentration-temperature history) is exposed to the removal mechanism.

If the depth affected by removal is significant compared to the total thickness of the layer, the single layer approach discussed may not be valid. A suitable model for this part of the layer should be defined (not considered here).

CASE STUDY: SUPPRESSION VS. REMOVAL

The objective of this case study is to investigate, using the model discussed, the phenomena limiting deposition (removal and suppression) and link them to measurable performance indicators, such as deposit thickness (hydraulic) and temperature (thermal). Deposit thickness, however, is difficult to measure directly. For a tube, pressure drop is taken as an indicator of fouling layer thickness. It should be noted that variations in deposit's roughness have been neglected. This assumption is a good approximation for the deposition-limiting mechanism here considered (suppression and removal by superficial erosion). However, the variation of this quantity may be important in other cases.

The base case considers a single tube in an oil refinery heat exchanger. Geometric parameters of the tube, physical and fouling properties of the crude oil and operating conditions representative of typical values in oil refineries, are selected (Table 1). Uniform wall temperature (UWT) at the tube wall is assumed. The impact of other wall boundary conditions, such as uniform heat flux, is discussed elsewhere (Coletti et al. 2010). For UWT, the thermal impact of fouling is observed in the outlet oil temperature and heat duty. For alternative operation modes, such as uniform heat flux, the thermal impact would be observed in the wall temperature.

The deposition and deposition-limiting rates are calculated as a function of the local conditions based on an adaptation of the Ebert–Panchal threshold model (Diaz-Bejarano et al. 2015), where:

$$n_{d,gel} = \alpha' \mathrm{Re}^{-0.66} \mathrm{Pr}^{-0.33} \exp\left(-\frac{E_f}{R_g T_{film}}\right) \qquad (4)$$

$$n_{s,gel} = \gamma' \tau_w; \; n_r = \gamma' \tau_w \qquad (5)$$

This correlation was chosen for coherence with previous works (Coletti and Macchietto 2011). Values of the parameters are reported in various studies fitted to lab or plant data (Panchal et al. 1997; Yeap et al. 2004; Coletti and Macchietto 2011). Here, representative values for the 3 adjustable parameters were chosen (reported in their modified form in Table 1). A sufficiently high value of γ' was chosen to permit moving to non-fouling conditions within the typical allowable velocity range in heat exchangers (up to 3-3.5 m/s).

Table 1. Parameters for single tube model test

Parameter	Value	Parameter	Value
R_i(mm)	9.93	UWT (ºC)	270
R_o(mm)	12.70	Inlet T (ºC)	200
L(m)	6.1	M (kg/s)	0.3
α' (kg/m² s)	0.94	E_f (kJ/mol)	30
γ (kg/m² s Pa)	$1.2 \cdot 10^{-8}$	E_a (kJ/mol)	50
A_a (no ageing)(s⁻¹)	0	A_a (fast) (s⁻¹)	0.01
λ_{gel} (W/m K)	0.2	λ_{coke} (W/m K)	1.0

Threshold conditions are identified by solving the fouling equation (Eq. (2) or (3)) for change in thickness equal to zero. The case is considered of constant mass flowrate of oil, i.e. where flow velocity changes due to change in flow radius as a result of fouling build up or depletion. It is noted that the location of the threshold changes with flowrate.

The threshold curve is represented in terms of film temperature (T_{film}) versus average velocity (u). For the fouling parameters and inlet conditions in Table 1, the threshold loci are shown in Fig. 2 (dashed line). The variation of film temperature and velocity as a result of fouling (here referred to as T_{film}-u path) and the deposit thickness at the tube midpoint are also shown in Fig. 2 for the *no ageing* case. Starting with a clean tube, the tube operates on the fouling side of the Threshold (point A in Fig 2). As a result of fouling build-up, T_{film} drops quickly

initially, more slowly at long times. The velocity, on the other hand, increases at a greater rate at later stages (as flow radius decreases). The line gradually approaches the threshold but does not reach it after 1 year (B in Fig 2).
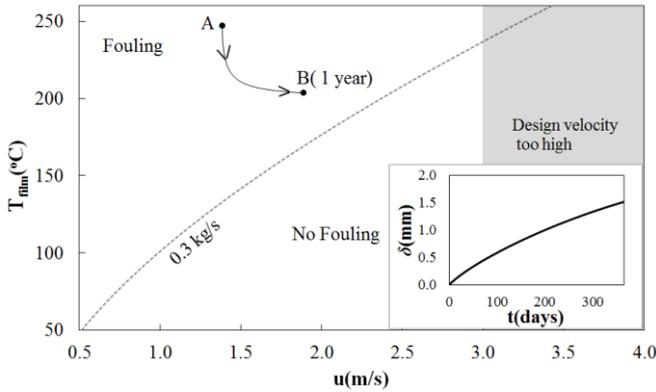

Figure 2. Threshold loci for constant mass flowrate (0.3 kg/s) (dashed line) and T_{film}-u path for growing fouling layer (no ageing) over a year at midpoint of tube (z = 3.05 m).

The form of Eqn. (2) and (3) suggests that suppression and removal lead to the same behavior when the net deposition rate is positive. The behavior is expected to be different when moving to conditions on the no-fouling side of the threshold. In order to investigate this point, the following operation schedule is considered: Period 1, fouling build up for 150 days (with M=0.3 kg/s); Period 2, high velocity period (M=0.6 kg/s) for 80 days, in order to move to the no-fouling side of the threshold; and Period 3, the velocity returns to fouling conditions (M=0.3 kg/s) for 50 days. This is similar to the experiments carried out in the Batch Stirred Cell system cited in the introduction (Young et al. 2011), but is applied here to a heat exchanger tube under operation conditions and time scales relevant to fouling in PHT heat exchangers. Ageing is expected to affect the thermal and rheological properties of the layer. Three cases are considered: a) No ageing; b) Fast ageing; c) Fast ageing with consolidation (hardening of the deposit).

No Ageing

During Period 1, the behavior of the system is the one shown in Fig. 2. Once the flowrate increases to 0.6 kg/s, operating conditions move over the threshold to no-fouling conditions (Fig. 3). The increase in flowrate moves the threshold line to the left, increasing the distance between the new operating conditions and the loci. Fig. 3 also shows in the inset the velocity profile over time. Key times at the beginning and end of each period are indicated: end of Period 1 (i), Period 2 (ii to iii) and Period 3 (iv to v). Plots for removal and suppression cases are overlaid. With the suppression model (Eq. 2), the system remains undisturbed during Period 2 (from ii to iii). Once the velocity is decreased, the system returns to the same point i as prior to the high velocity period (points i and iv overlap). With the removal model (Eq. 3), the T_{film}-u path shows a hysteresis cycle due to the decrease in thickness: T_{film} increases during Period 2 due to partial removal of the layer; as a result, the

system resumes from a less fouled situation at the beginning of Period 3.

The impact of the two deposition-limiting mechanisms on the measurable variables is shown in Fig. 4. After the threshold is crossed, with suppression the layer stops growing and the thickness is constant in Period 2 (Fig. 4(a)). With removal, the layer thickness decreases approximately 0.34 mm during the high flow period. There is a sudden decrease in outlet temperature in both cases as a larger amount of oil is heated (Fig. 4(b)).
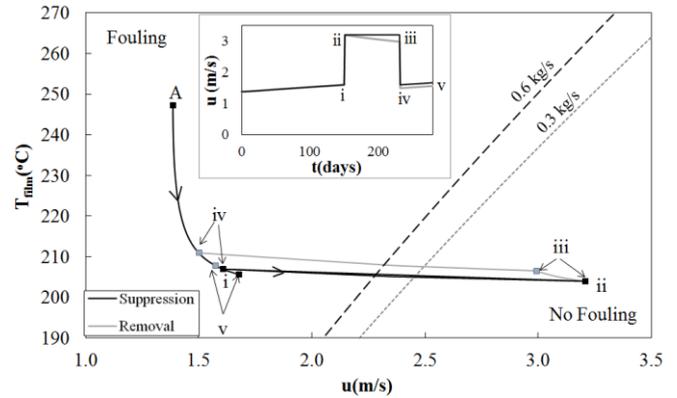

Figure 3. Threshold (dashed) and T_{film}-u path and velocity over time at midpoint of tube (z = 3.05 m) for no ageing. In the inset, velocity profile over time.

With suppression the outlet temperature remains constant, whilst with removal there is a 3.3ºC increase, as consequence of the reduction in thickness. As discussed, the impact of fouling on the thermal behavior is less significant at later stages; hence the effect on the outlet temperature is relatively small, but still noticeable.

The impact on pressure drop (Fig. 4(c)) is clearly distinguishable between the two cases: pressure drop stays constant with suppression and gradually decreases (about 4.3 kPa) with removal during the high flow period. Opposite to thermal effect, the effect on pressure is more significant at a late stage. For comparison with traditional methodologies, the tube-average fouling resistance (referred to the outer tube area) is also calculated as an indicator of the change in thermal performance. The fouling resistance (Fig. 4(d)) follows the same pattern as the thickness, which is expected since no ageing is taking place and the conductivity is radially uniform throughout the deposit.
In conclusion, if no ageing occurs, the deposition-limiting mechanism could be identified from either thermal (temperature, resistance) or hydraulic (ΔP) effects.

Fast Ageing

Organic matter is likely to undergo ageing under the operating conditions considered here. The previous numerical experiment is repeated with a fast ageing deposit (Table 1). This T_{film}-u path is shown in Fig. 5. The ageing process introduces hysteresis in the T_{film}-u path during the high flow period even with suppression and this effect is amplified with removal. This is due to the improved heat transfer properties as a result of conversion of gel to coke.
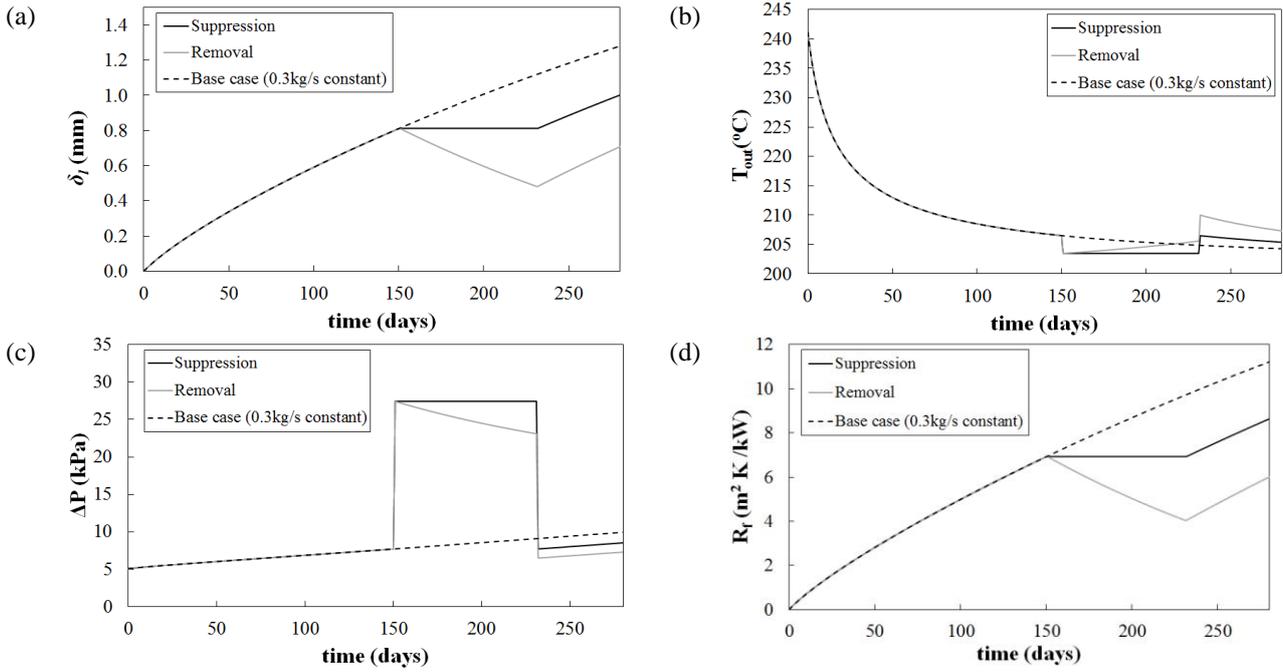
Figure 4. Time profiles of (a) thickness at midpoint of tube (z = 3.05 m), (b) outlet temperature, (c) pressure drop, (d) fouling resistance, during Periods 1 to 3 for a deposit without ageing, for suppression and removal.
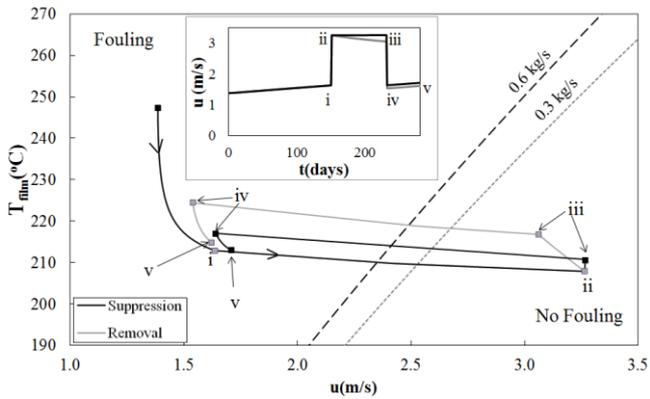

Figure 5. Threshold (dashed) and T_{film}-u path and velocity over time at midpoint of tube (z = 3.05 m) for fast ageing. In the inset, velocity profile over time.

The concentration profiles at key times are shown in Fig. 6. With suppression, during Period 2 (ii-iii) (Fig. 6(a)) the thickness of fouling layer does not change, but the deposits undergo ageing (even at the layer surface) of gel to coke. With removal, a reduction of the thickness occurs in addition to ageing (Fig. 6(c)), causing a reduction in velocity and further reduction in thermal resistance. Once the flowrate returns to the initial value (point iv), the concentration at the layer boundary returns to that of fresh deposit. In Period 3, the layer grows again, but starting from different thickness with suppression (Fig. 6(b)) and removal (Fig. 6(c)), hence leading to different velocities and surface temperatures. This also causes a small increase in fouling rate in the removal case. As a result, the thickness growth in Period 3 (iv-v) is (slightly) larger with removal.
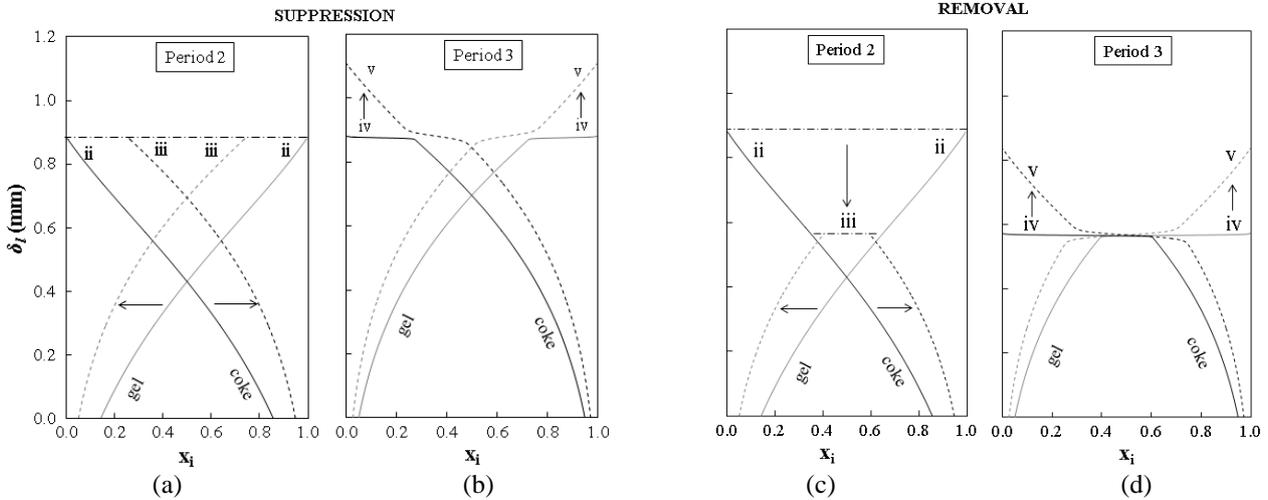

Figure 6. Volume fraction evolution with suppression (a,b) and removal (c,d) during Period 2 and 3 at midpoint of tube (z = 3.05 m) for fast ageing. Continuous and Dotted lines indicate start and end of the period, respectively.

During Period 3, a step in the concentration profile is shown (Fig. 6(b, d)) which separates the material built-up during Period 1 (older, close to the wall) and that built-up during Period 3 (newer, close to the surface).

The effects on the outlet temperature, R_f and pressure drop are shown in Fig. 7. The thickness and, hence, pressure drop profiles over time are very similar to those in the no-ageing (thickness profile is not shown here). The final thickness is slightly larger than in the "no ageing" case as a result of the improved conductivity due to ageing, which increases fouling rate.
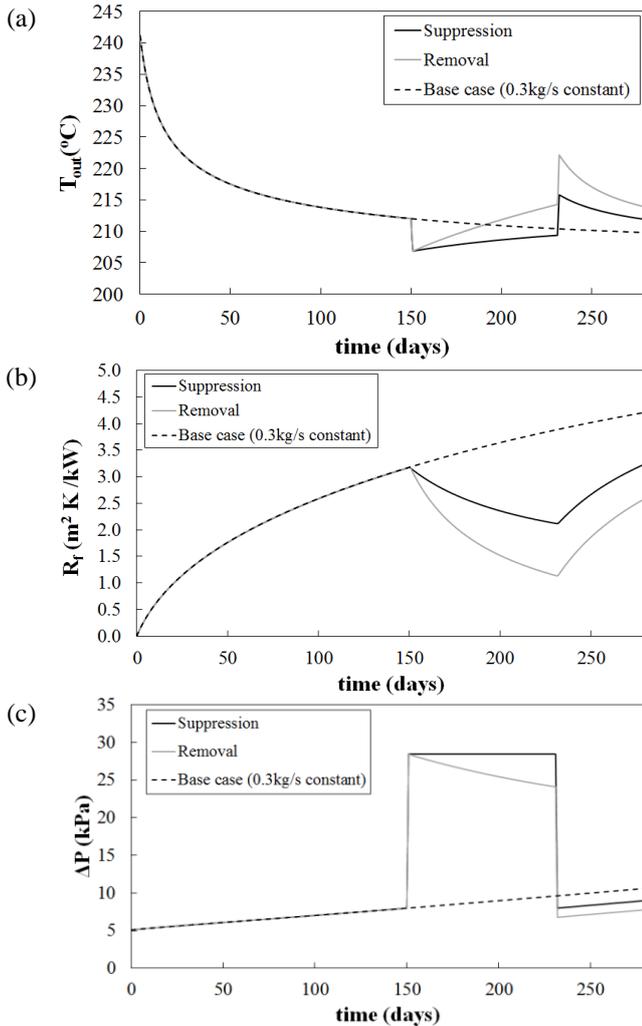

Figure 7. Time profiles of (a) outlet temperature, (b) fouling resistance, and (c) pressure drop during Periods 1 to 3 for a deposit with ageing, with suppression and removal.

Regarding the thermal effect, the profiles for outlet temperature and fouling resistance are similar in shape with suppression and removal, with a more acute variation during Period 2 for removal. Apparent negative fouling rates are observed during Period 2 (high velocity) if fouling resistance is taken as indicator (Fig. 7(b)). The time profile of the fouling resistance presents a saw-tooth shape in both cases, and therefore this kind of response cannot be used in isolation as an indicator of removal, as suggested by Crittenden et al. (1992). Unless very precise

characterization of the ageing rate is available in advance, so as to accurately characterize the thermal behavior, it is necessary to use pressure drop measurements to unmask the dominant process limiting deposition.

Under industrial operating conditions ageing is thought to be relevant for long time scales (Coletti et al. 2010). Laboratory experiments are usually carried out under conditions that accelerate fouling, which may also accelerate ageing. As a result, the negative fouling rates (decline in fouling resistance) experimentally observed by Young et al. (2011), although likely to be due to removal, cannot be taken as unequivocal proof of such mechanism.

Removal with Consolidation by Ageing

The mechanical strength (i.e. resistance to removal) of the layer has been so far assumed to be unaffected by ageing. However, ageing is believed to harden the organic deposit, as discussed in the introduction. Some fouling models consider the resistance of the deposit to shear. Most models feature an inverse dependence of the removal term on the shear strength of the material and a direct proportionality to the shear stress, e.g. Bohnet et al. (1997) for crystallization fouling of calcium sulfate. A similar approach is often used to describe soil erosion by water (Knapen et al. 2007). These models usually consider two parameters: "erodibility", related to the strength of the soil (often considered inversely proportional to shear strength); and critical shear stress (τ_c), the minimum shear stress required to start eroding the soil.

In the case of thermal fouling, there is no reported data for any of the above parameters to characterize the strength of the deposit and the resistance to shear forces. For simplicity, a critical shear stress (τ_c) is considered and assumed to depend on the composition of the layer at the boundary. A removal model alternative to Eq. (5) is obtained by assuming a removal rate proportional to the difference between shear stress and this critical value:

$$n_r = \gamma' \left(\tau_w - \left(x_{gel} \big|_{r=Rflow} \tau_{c,gel} + x_{coke} \big|_{r=Rflow} \tau_{c,coke} \right) \right) \qquad (7)$$

Values of $\tau_{c,i}$ equal to 0 (no resistance to removal) and 20 kPa are assumed for gel and coke, respectively. During growth, the deposit at the boundary is entirely composed of gel and Eqn. 7 is reverted to the original expression for removal (Eqn. 5). As shown in Fig. 6(c), when the threshold line is crossed part of the top deposit is removed and the concentration at the boundary gradually becomes that of inner layers, i.e. with increased coke content. With consolidation, ageing implies an increase in τ_c. The impact of this consolidation on the deposit thickness and pressure drop is shown in Fig. 8. The results (dashed line) show an initial reduction in the thickness which is gradually cushioned until a plateau is reached. Whilst the first layer is easy to remove, the layers below present increasingly higher resistance to shear forces. At some point (with removal rate still far from a zero) the removal rate equals the deposition rate and the deposit thickness stabilizes. The thermal effect (outlet temperature and fouling resistance, not showed here) is intermediate between those with suppression and removal

in the previous case (Fig. 7). The gradual reduction in removal rate (as the critical shear stress increases) results in a displacement of the location of the threshold due to consolidation, shown in Fig. 9. During Period 2 (ii → iii), the concentration of gel at the surface becomes gradually lower (here only a few snapshots are shown) leading to threshold loci at progressively higher velocities. As in the previous case, measurement of thickness (or pressure drop) change allows distinguishing suppression from removal.
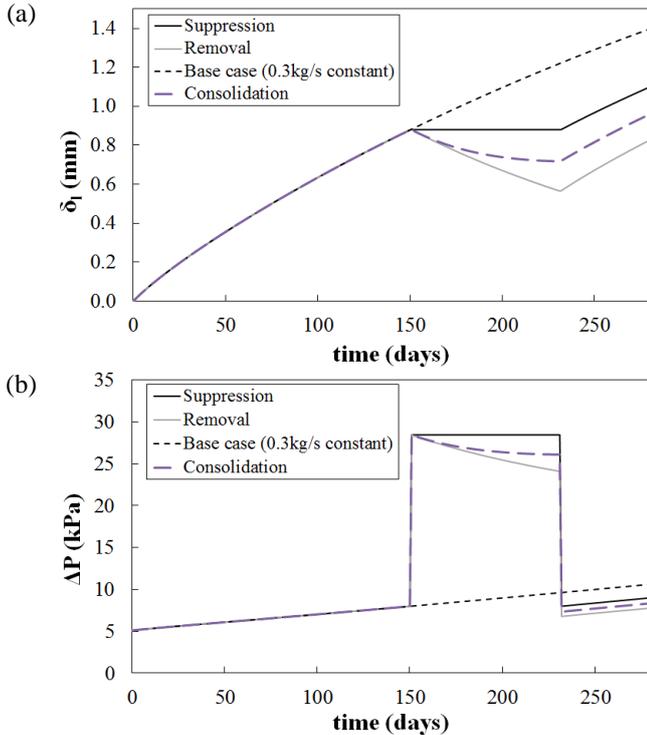

Figure 8. Time profiles of (a) thickness at midpoint of tube (z = 3.05 m), (b) pressure drop, during Periods 1 to 3 for a deposit without ageing, with suppression, removal and removal with consolidation by ageing.
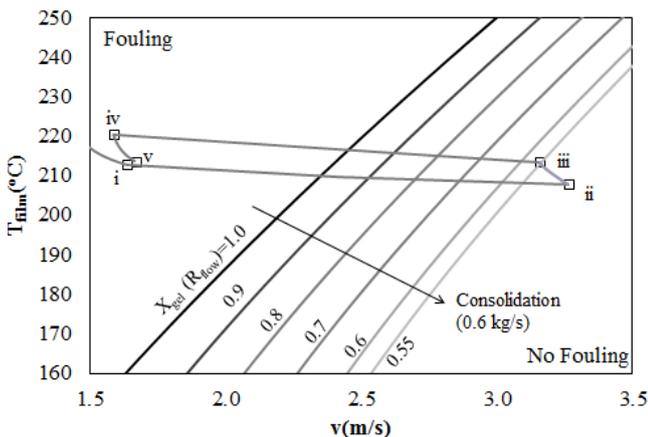

Figure 9. Threshold displacement at midpoint of tube (z = 3.05 m) due to consolidation

Here, representative values of τ_c were arbitrarily chosen within a reasonable range (Knapen et al. 2007), to provide negligible resistance to removal of gel, significant resistance to removal of coke and show the effect of consolidation.

Experimental characterization of deposits is required to find the actual dependence on concentration and characteristic values for fresh and aged deposit. With fast consolidation, the reduction in thickness produced could be small and unnoticeable in the measurements, and again it may not be possible to distinguish between suppression and removal based on hydraulic responses alone. Characterization of the ageing process and its effect on the properties of the deposit would be, again, required to decouple effects and becomes a pre-requisite in understanding the dominant deposition-limiting mechanism. Hardening of the deposit due to ageing does not necessarily imply the deposition-limiting mechanism to be suppression, since removal could be still possible if there is a superficial (erosion type) process.

CONCLUSIONS

The phenomena potentially limiting deposition in crude oil thermal fouling have been considered. A dynamic, distributed first-principles layer model (Diaz-Bejarano et al. 2015) was used to simulate the impact of suppression and removal on measurable thermal and hydraulic performance of a heat exchanger tube. Removal was assumed to be of superficial type (erosion). A sequence of three periods was considered: 1) deposit build up; 2) increase in flowrate moving to the no-fouling side of the threshold; 3) flowrate reduction to the original value with resumption of fouling. The simulations were performed for three cases: no ageing, ageing, and ageing with deposit consolidation.

It was shown that during a deposit layer growth phase, removal and suppression have the same effect and are indistinguishable. However, removal and suppression mechanisms give different responses during the high velocity period when velocity is increased to cross the threshold curve *after some initial fouling build up*. The results show that thermal effects may be unequivocally taken as a measurement of fouling rate (i.e. as an indicator of the dominant deposition-limiting mechanism) only when the deposit does not age. When ageing takes place, however, it contributes to the change in the observed thermal behavior. As a result, trends in temperature and fouling resistance show similar shape during the high speed period, with the changes being more pronounced in case of removal, and simultaneous consideration of thermal and pressure drop effects is necessary to correctly interpret the data. Mechanical consolidation by ageing, if very acute, may hinder the ability to resolve deposition-limiting mechanism even when hydraulic effects are measured. From this analysis, it follows that the typical arguments used in literature to support one or other of the deposition-limiting hypotheses are not sufficiently conclusive.

The work presented here shows the risks of studying fouling merely based on thermal effects, and particularly on thermal resistances. To unambiguously distinguish the underlying phenomena, the following is required:
1. Accurate measurement over time of thermal performance and deposit thickness (directly or by measuring pressure drop in the case of a tube).
2. Characterization of ageing and its impact on the properties of the deposit, with special attention to its thermal conductivity and rheology (i.e. strength).

ACKNOWLEDGMENTS

NOMENCLATURE

A_a Ageing pre-exponential factor, 1/s
E_a Ageing activation energy, J/mol
E_f Fouling activation energy, J/mol
L Tube length, m
M Mass flowrate, kg/s
n Mass flux, $kg/m^2 s$
Pr Prandtl number, $C_p \mu / \lambda$, dimensionless
R Radius, m
R_f Fouling resistance referred to outer tube area, $m^2 K/W$
R_{flow} Radius at the fouling layer-fluid interfase, m
R_g Ideal gas constant, 8.314J/molK
r Radial coordinate, m
Re Reynolds number, $\rho u d_o / \mu$, dimensionless
t Time, s
T Temperature, K
T_{film} FilmTemperature, K
u Linear velocity, m/s
UWT Uniform wall temperature, K
x volume fraction, -
z Axial coordinate, m
α, β, γ Constants in Eq. (1)., $m^2 K/J$, - , $m^2 K/J Pa$
α' Modified Deposition constant, $kg/ m^2 s$
γ' Modified suppression constant, $kg/ m^2 s Pa$
δ_l Deposit thickness, m
ΔP Pressure drop, Pa
λ Thermal-conductivity, W/mK
ρ Density, kg/m^2
τ_c Critical Shear stress, N/m^2
τ_w Shear stress, N/m^2

Subscript
i inner
coke aged organic deposit
gel fresh organic deposit
l layer
d deposition
o outer
r removal
s suppression

REFERENCES

Bohnet, M., 1987. Fouling of heat transfer surfaces. *Chemical Engineering Technology*, 10, pp.113–125.

Bohnet, M., Augustin, W. and Hirsch, H., 1997. Influence of fouling layer shear strength on removal behaviour. In T. R. Bott, ed. *Understanding Heat Exchanger Fouling and its Mitigation.* New York: United Engineering Foundation and Begell House, pp. 201–208.

Coletti, F.. Ishiyama, E.M., Paterson, W.R., Wilson, D.I. and Macchietto, S., 2010. Impact of Deposit Aging and Surface Roughness on Thermal Fouling : Distributed Model. *AIChE J*, 56(12), pp.3257–3273.

Coletti, F., Joshi, H.M., Macchietto, S. and Hewitt, G.F., 2014. Introduction to Crude Oil Fouling. In F. Coletti and G. F. Hewitt,

eds. *Crude Oil Fouling: Deposit Characterization, Measurements, and Modeling.* Boston (USA): Gulf Professional Publishing.

Coletti, F. and Macchietto, S., 2011. A Dynamic, Distributed Model of Shell-and-Tube Heat Exchangers Undergoing Crude Oil Fouling. *Ind & Eng Chem Res*, 50(8), pp.4515–4533.

Crittenden, B.D., Kolaczkowski, S.T. and Downey, I.L., 1992. Fouling of Crude Oil Preheat Exchangers. *Trans IChemE, Part A, Chem Eng Res Des*, 70, pp.547–557.

Diaz-Bejarano, E., Coletti, F. and Macchietto, S., 2015. A new dynamic model of crude oil fouling deposits and its application to the simulation of fouling-cleaning cycles. . in press: *AIChE J*, DOI: 10.1002/aic.15036.

Ebert, W.A. and Panchal, C.B., 1995. Analysis of Exxon crude-oil-slip stream coking data. In C. B. Panchal, ed. *Fouling Mitigation of Industrial Heat-Exchange Equipment*. San Luis Obispo, California (USA): Begell House, pp. 451–460.

Epstein, N., 1983. Thinking about Heat Transfer Fouling : A 5 × 5 Matrix. *Heat Transf Eng*, 4(1), pp.43–56.

Ishiyama, E.M., Coletti, F., Macchietto, S., Paterson, W.R. and Wilson, D.I., 2010. Impact of Deposit Ageing on Thermal Fouling : Lumped Parameter Model. *AIChE J*, 56(2), pp.531–545.

Kern, D.Q. and Seaton, R.E., 1959. A theoretical analysis of thermal surface fouling. Brit. Chem. Eng., 4(5), pp.258–262.

Knapen, A., Poesen, J., Govers, G., Gyssels, G. and Nachtergaele, J., 2007. Resistance of soils to concentrated flow erosion: A review. *Earth-Science Reviews*, 80(1-2), pp.75–109.

Knudsen, J.G., Lin, D. and Ebert, W.A., 1999. The determination of the threshold fouling curve for a crude oil. In T. R. Bott et al., eds *Understanding Heat Exchanger Fouling and its Mitigation*. New York: Begell House, pp. 265–272.

Panchal, C.B., Kuru, W.C., Liao, C.F., Ebert, W. A. and Palen, J.W., 1997. Threshold conditions for crude oil fouling. In T. R. Bott, ed. *Understanding Heat Exchanger Fouling and its Mitigation*. Lucca, Italy: Begell House, pp. 273–281.

Panchal, C.B. and Watkinson, A.P., 1994. *Development of an Analytical Model for Organic-Fluid Fouling*. Technical report. Argonne National Laboratory, U.S. Department of Energy.

Somerscales, E.F.C. and Sanatgar, H., 1989. Investigation of the Deposit Removal Processes on a Fouled Surface. In R. Shah, ed. *Proceedings of the 1989 National Heat Transfer Conference, Heat Transfer Equipment Fundamentals, Design, Applications and Operating Problems*. Philadelphia (USA): ASME, pp. 261–271.

Watkinson, A.P. and Wilson, D.I., 1997. Chemical reaction fouling: A review. *Exp Therm & Fluid Sci*, 14(4), pp.361–374.

Wilson, D. I., Ishiyama, E. M., Paterson, W. R. and Watkinson, A. P., 2009. Ageing: Looking back and looking forward. In H. Müller-Steinhagen, M. R. Malayeri, and A. P. Watkinson, eds. *Int. conf. on heat exchanger fouling and cleaning VIII*. Schladming, Austria, pp. 221–230.

Wilson, D.I., Polley, G.T. and Pugh, S.J., 2005. Ten Years of Ebert, Panchal and the "Threshold Fouling" Concept. In H. Müller-Steinhagen, M. R. Malayeri, and A. P. Watkinson, eds. *6th International Conference on Heat Exchanger Fouling and Cleaning - Challenges and Opportunities*. Kloster Irsee (Germany): ECI Symposium Series, pp. 25–36.

Yang, M., O'Meara, A. and Crittenden, B.D., 2011. Determination of crude oil fouling thresholds. In M. R. Malayeri, H. Müller-Steinhagen, and A. P. Watkinson, eds. *Proceedings of International Conference on Heat Exchanger Fouling and Cleaning*. Crete Island, Greece, pp. 1–6.

Yeap, B.L., Wilson, D. I., Polley, G. T. and Pugh, S., 2004. Mitigation of crude oil refinery heat exchanger fouling through retrofits based on thermo-hydraulic fouling models. *Chem Eng Res and Des*, 82(1), pp.53–71.

Young, A. et al., 2011. Characterization of Crude Oils and Their Fouling Deposits Using a Batch Stirred Cell System. *Heat Transfer Engineering*, 32(3-4), pp.216–227.