PARTICULATE FOULING: CONTROLLING PROCESSES AND DEPOSIT STRUCTUTRE

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Fouling caused by a water-kaolin suspension in an annular heat exchanger was studied. Deposition seemed to be controlled by mass transfer for lower Reynolds numbers and by adhesion for higher Re. The data was satisfactorily described by the *generalized* model of Pinheiro.

The relative cohesion of the deposits was measured using a rotating cylinder apparatus, which also helped in confirming the existence of a *loose* and a *hard* layer in the kaolin deposits.

1. INTRODUCTION

Fouling can seriously affect the operating behaviour of sensible heat exchangers, boilers, condensers, evaporators, etc, since the deposits usually reduce the thermal efficiency of the equipment, increase the pumping costs and obstruct the flow passages.

Estimates of fouling costs in the United Kingdom and in the United States have been published [1,2] pointing out to about 0.25-0.30% of the Gross National Product; furthermore, at least in oil refineries, 30-40% of these costs can be assigned to energy losses [2].

In this paper an experimental study of particulate fouling caused by a water-kaolin suspension on copper tubes is reported, in which the effects of fluid velocity and the adhesion/cohesion characteristics of the deposits were examined.

2. FOULING MODELS

The basic theory of fouling follows the ideas suggested by Kern and Seaton [3], who assumed that deposits grow as a result of the competition between a deposition rate (ϕ_d) and a removal rate (ϕ_r). Considering that ϕ_d remains constant during the fouling process and that ϕ_r increases with the deposit thickness (or its thermal resistance), the authors obtained the following equation:

$$R_f = R_f^\infty [1 - \exp(-\beta t)] \qquad (1)$$

where R_f is the thermal resistance of the deposit at time t, R_f^∞ is the maximum (asymptotic) value of the R_f (at $t = \infty$) and β is the proportionality constant between ϕ_r and R_f:

$$\phi_r = \beta R_f \qquad (2)$$

According to Kern and Seaton, β will be directly proportional to the wall shear stress, which is a function of the square of the fluid velocity. It should be noted that for equal R_f, a higher value of β means that the removal of the deposit will proceed at a higher rate; hence, $1/\beta$ can be considered as a measure of the deposit resistance, or of its *cohesion*.

It can also be shown that:

$$\phi_d = \beta R_f^\infty \qquad (3)$$

The ideas of Kern and Seaton were further developed by several authors, mainly in what concerns the deposition term, although all the models maintained the basic form of equation 1. An unifying approach was presented by Pinheiro [4,5], involving the particulate fouling model of Watkinson and Epstein [6], the chemical reaction fouling model of Crittenden and Kolaczkowski [7] and the precipitation fouling model of Taborek et al [8]. The concepts of Cleaver and Yates [9,10] regarding the removal process were also considered in Pinheiro's *generalized* model. Its fundamental assumptions are the following (for the case of particulate fouling):

(i) The deposition process involves the transport of particles (by turbulent diffusion) to the deposition surface, followed by a surface interaction mechanism (adhesion). If u is the mean fluid velocity, C the suspension concentration, T_s the surface temperature and f the friction factor, the deposition rate will be:

$$\phi_d = \frac{C}{\dfrac{1}{k_1\sqrt{f}\,u} + \dfrac{ub}{k_0\exp(-E/RT_s)}} = \frac{C}{\rho_f\, k_f\left[\dfrac{1}{k_t} + \dfrac{1}{k_r}\right]} \qquad (4)$$

where E is an *activation energy* associated to the adhesion process, R is the ideal gas constant and k_0, k_1 are proportionality constants. k_t and k_r are the transport and the adhesion rate coefficients, while ρ_f and k_f are the density and

thermal conductivity of the deposit.

Parameter b depends essentially on the individual processes that control the deposition phenomenon:

- If the mass transfer step is much slower than the surface phenomena process ($k_t \ll k_r$), $\phi_d = (1/\rho_f k_f) k_t C$ and b = 0, which means that the deposition rate increases with fluid velocity.

- If the transport rate is much greater than the adhesion rate, then $\phi_d = (1/\rho_f k_f) k_r C$ and, as suggested by data from several authors [4], $0 \le b \le 1$. In this case, ϕ_d will decrease with increasing fluid velocity.

(ii) The removal rate is a function of the hydrodynamic forces acting on the deposit and of the cohesive properties of the latter. As Cleaver and Yates [9] pointed out, there will be a *minimum fluid velocity*, or shear stress, necessary for removal to occur. Based on experimental observations of several authors [11,12,13], Pinheiro assumed that the structure of the deposit is not uniform, consisting of (at least) two layers: the inner one, near the wall, is more hard and adherent, while the outer one, in contact with the fluid, is composed by more or less loose particles and is, thus, more easily removable by the fluid. Hence the *minimum fluid velocity* will be determined by the resistance to removal of the outer layer: the higher the cohesion of this layer, the higher will be u_{min}.

ϕ_r is taken as proportional to the shear stress, i.e., to $f u^2$, but considering, as Taborek et al [8] did, that higher velocities originate harder deposits, ϕ_r will also vary inversely with u^a, where a is an empirical parameter depending on the cohesive properties of the deposit. The removal term in Pinheiro's model is then:

$$\phi_r = k_2 f (\frac{u}{u_{min}})^{2-a} R_f \qquad (5)$$

where it is probable that $0 < a \le 2$ (4). k_2 is a proportionality constant.

Table 1 summarizes the dependence of ϕ_d, β and R_f^∞ on the fluid velocity, as given by the model of Pinheiro; in this table, the Blasius correlation for the friction factor ($f \sim u^{-0.25}$) was taken into account.

Table 1 Effects of fluid velocity on fouling, as predicted by Pinheiro's model.

Controlling Process	ϕ_d	β	R_f^∞
Transport (Turb. diffusion)	$(u)^{0.875}$	$(u)^{1.75-a}$	$(u)^{-0.875+a}$
Adhesion	$(u)^{-b}$	$(u)^{1.75-a}$	$(u)^{-1.75+a-b}$

3. EXPERIMENTAL EQUIPMENT AND METHODS

3.1 Annular Heat Exchanger

An horizontal annular exchanger has been used

consisting of a 2 meters long external perspex tube (ID=36 mm) and a removable inner copper tube (OD=25 mm) heated by an electrical resistance placed inside it. Thermocouples were located in the internal upper wall of the copper tube at five different positions in the axial direction (A,B,C,D,E), the distances from the inlet of the fluid being: A-20 cm; B-45 cm; C-78.5 cm; D-111.5 cm; E-145 cm. Fluid temperatures were also measured at the same five positions. Pressure drop was monitored.

The water-kaolin suspension (particle mean equivalent diameter = 7 microns) was cooled and stirred in a 250 liters vessel, pumped through PVC tubes to the test section and back to the reservoir. Voltage and current intensity were registered in order to evaluate the heat flux. Therefore, overall heat transfer coefficients could be determined at each one of the five positions, knowing the local wall and fluid temperatures as well as the heat flux.

All fouling tests were run at constant heat flux (3000 W/m^2), constant kaolin concentration (2.2 kg/m^3), constant bulk water temperature (12°C) and constant pH (7.5). Samples of fluid were periodically withdrawn for analysis of kaolin concentration and suspension pH. After the tests, the thickness of the deposits was measured using a micrometer inserted in an appropriate electrical circuit, following a technique described by Harty and Bott [15] and adapted by Melo and Pinheiro [16].

3.2 Rotating Cylinder Apparatus

This apparatus has a removable inner cylinder (2 cm long, OD=25 mm) placed inside an outer rotating perspex cylinder (8 cm long, ID=36 mm). A Variac controls the input voltage to the alternative motor connected to the outer cylinder, the rotation speed of the latter being measured with a stroboscope.

Samples of the fouled copper tubes obtained in the annular exchanger tests were used as the inner cylinder in the apparatus. Before placing them inside the device, filled with water at pH $\simeq$ 7.5, they were dried and weighed. The outer cylinder was then rotated at low velocity for 3 minutes, after which the sample was removed, again dried and weighed. This procedure is repeated at increasing speeds of rotation in order to measure the effect of hydrodynamic forces on the deposit.

The hydrodynamic force can be evaluated using the correlations developed by Wendt [17] - also referred by Visser [14] - that relate the rotation speed to the shear stress at the surface of the inner cylinder.

4. RESULTS AND DISCUSSION

4.1 Controlling Processes

Using clean surfaces, overall (U_0) and convective (h_0) heat transfer coefficients were determined by the Wilson method [18]. U_0 and h_0 are related by:

$$\frac{1}{U_0} = R_w + \frac{1}{h_0} \qquad (6)$$

where R_w is the thermal resistance between the point where the wall thermocouple is located and the clean deposition surfaces.

During the fouling tests, the overall heat transfer coefficient changes as the deposit builds-up. If, at a given time t, the overall coefficient is U and h is the convective coefficient, the thermal resistance of the deposit at the same time (R_f) will be:

$$R_f = \frac{1}{U} - \frac{1}{U_0} + \frac{1}{h_0} - \frac{1}{h} \qquad (7)$$

Supposing that there are no blockage effects (which in the present case is practically true because the thickness of the deposits is much smaller than the annulus diameter), changes in h will be due to changes in the roughness of the deposition surface as the particles adhere to it. Melo and Pinheiro [19] described a technique, based on pressure drop measurements, that can be used to evaluate the relationship between h and h_0 at the solid/fluid interface in a fouled annulus.

Fouling curves (R_f versus t), similar to the one shown in Figure 1, were then obtained from tests conducted at Reynolds numbers (Re) 2300, 2760, 4140, 6900 and 11040. By fitting equation 1 to the data, values of R_f^∞ for positions A,B,C,D, and E were determined as a function of Reynolds numbers (see Figure 2); final thickness (y_f) versus Re curves were also drawn (see Figure 3) indicating the same trends.

Values of ϕ_d were calculated with equation 3 and are shown, as a function of Re, in Figure 4 for position D and E, and in Figure 5 for positions A and B.

Figure 6 shows the *resistance to removal* of the deposits ($1/\beta$) as a function of Reynolds numbers for positions A,D and E.

Striking differences between the amount of fouling obtained at the various positions in the exchanger can be readily noted in Figures 2 and 3. Previous studies [20] have shown that for the lower Reynolds numbers (below 7000, more or less), positions A,B and, C are in a developing laminar flow region where the removal forces are weaker, which explains the thicker deposits obtained in this entrance region. In the fully developed non-laminar region (positions D and E), R_f^∞ (or y_f) increases with fluid velocity up to Re $\simeq$ 4000, the trend being inverted for higher Re values. A possible explanation for this behaviour, at D and E, is the following:

- For low fluid velocities (Re < 4000), mass transfer rates are small and the transport of particles can be the process controlling deposition. In this case, increasing the Reynolds numbers will not only increase ϕ_r, but also ϕ_d (see Figure 4.). If, simultaneously, there is an increase in the cohesion of the deposit (see Figure 6), thicker deposits will be obtained as the Reynolds number grows. This feature is predicted by Pinheiro's model when a > 0.875 (see table 1).
- For higher fluid velocities, a change in control will occur between Re > 4000 and Re $\simeq$ 6000, and the deposition process will

be governed by adhesion. Consequently, ϕ_d ceases to increase with Re, while the fluid removal forces continue to grow overcoming the cohesion effect. Figure 4 shows that, as predicted by Pinheiro's model, ϕ_d decreases with increasing Reynolds numbers.

In what concerns the upstream positions, laminar effects will prevail as far as those points remain in a developing flow region. In such a case, an increase in the velocity will not result in a significant increase in the transport rate, because molecular (and not turbulent) diffusion will be the mass transfer mechanism. Furthermore, gravitational effects can also play a role in the laminar region and, if so, transport rates will be independent of fluid velocity (see Figure 5). In any case if the shear stress is increased, the final amount of deposit will decrease, as shown in Figures 2 and 3 for positions A and B.

For the higher range of Reynolds numbers, similar trends for all the five positions can be observed in Figures 2 and 3. At A and B, however, the curves in Figure 5 (ϕ_d versus Re) seem to indicate that mass transfer still controls depositions for Re > 6000; but, as the cohesion of the deposits (Figure 6, curve A) decreases, lower thicknesses are obtained as Re is increased.

The model of Pinheiro was fitted to the data of Figure 2, 4 and 6 using the average values of D and E, in order to test the proposed fluid velocity dependencies for the two limiting cases of deposition control presented in Table 1.

In the lower range of Reynolds numbers, the dependence of ϕ_d on u is close to the usual turbulent diffusion relationship (exponent = =0.8).

Parameters a and b were also evaluated. The results of the fitting procedure are presented in Table 2 and appear to support reasonably well the proposed model, although the values of a and b are, in some cases, beyond the limits suggested by Pinheiro.

4.2 Deposit Characteristics

Macroscopically, the kaolin deposits that were obtained in the heat exchanger were similar to a continuous layer of white paint, although a greater thickness could be easily observed in the entrance region. Samples of the fouled copper tubes were examined on the scanning electron microscope, showing a surface layer of loose particles and a more compact structure in the inner zones of the deposit.

Figure 7 shows the results of the removal experiments carried out in this apparatus using samples of the annular exchanger fouled tubes at position D.

These results seem to indicate that:
- The deposit obtained with Re = 4140 has a higher cohesion than the one obtained with Re = 2300, confirming the results of Figure 6 and the explanation suggested in 4.1 for the increase in R_f^∞ with fluid velocity.
- All the curves in Figure 7 present a similar shape, showing a *transition point* above which the fluid velocity has a minor effect on the amount of deposit removed. This *transition*
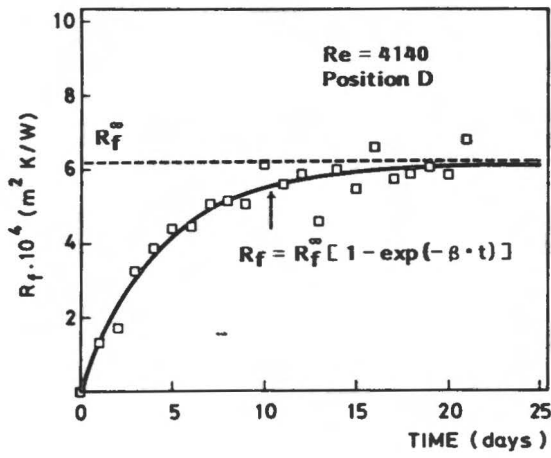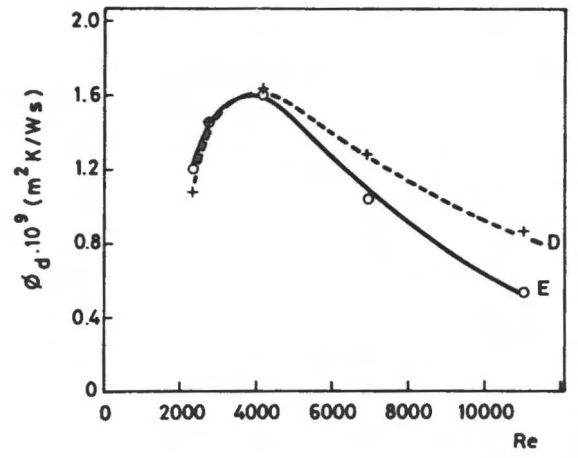
Figure 1 Typical fouling curve

$$R_f = R_f^\infty [\, 1 - \exp(-\beta \cdot t)\,]$$

Re = 4140
Position D

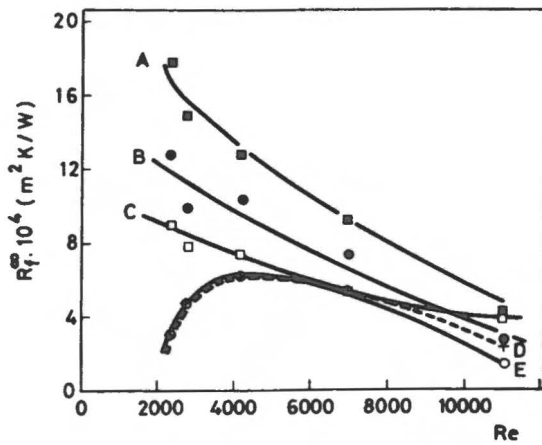Figure 4 Deposition rate versus Reynolds No.

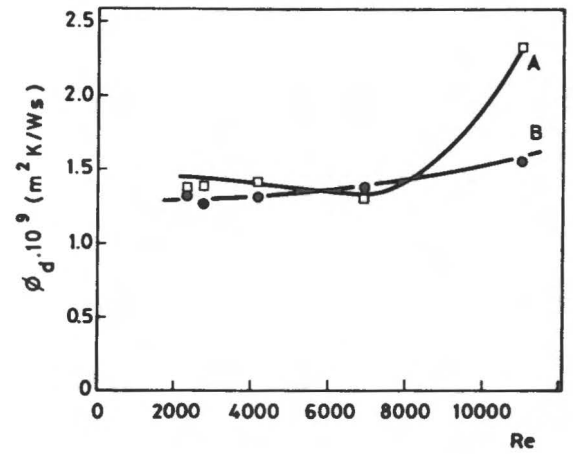Figure 2 R_f^∞ versus Reynolds No.

Figure 5 Deposition rate versus Reynolds No.
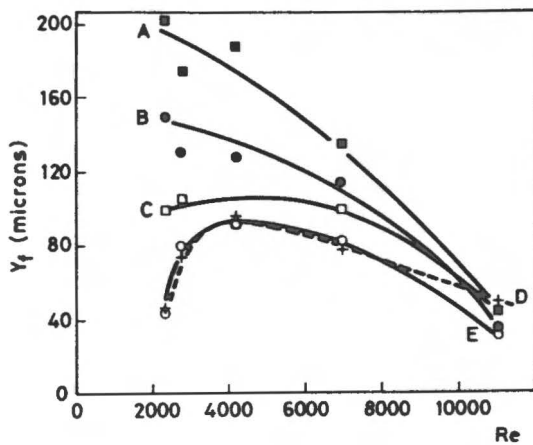

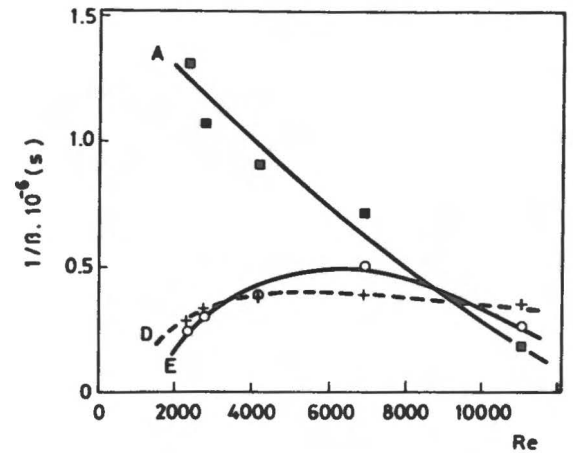Figure 3 Final thickness versus Reynolds No.

Figure 6 Resistance to removal versus Reynolds No.

Table 2 Fitting of Pinheiro's model to data from the annular heat exchanger
(average of positions D and E).

Range of Reynolds No.	Process controlling deposition	Effects of fluid velocity			Parameters	
		ϕ_d	β	R_f^∞	a	b
2300- 3300	Mass transfer (turbulent diffusion)	$u^{0.72}$	$u^{-0.74}$	$u^{1.43}$	2.3-2.5	0
6900-11400	Adhesion	$u^{-1.03}$	$u^{0.64}$	$u^{-1.62}$	1.1-1.2	1

point could represent, according to the hypothesis of a two-layer structure in the deposits, the transition from the *loose* to the *hard* layer. Hence, the deposit obtained with the lowest Reynolds number (Re = 2300) would contain around 50% of hard layer, while the deposits obtained with Re = 4140 and Re = 6900 would contain a higher proportion of hard layer (about 70%). For Re = 6900, the hard layer appears to have a lower cohesion than for Re = 4140, and the inverse seems to occur with the loose layer.

• A value of u_{min} can be determined in the case of Re = 6900. Extrapolating the curve to 0% removal, the minimum speed of rotation will be 8 rot/s , corresponding to a mean fluid velocity of 0.09 m/s [17]. This is much lower than the mean fluid velocity in the annular heat exchanger when Re = 6900 (u = 0.79 m/s), showing that the removal of the loose layer is easily accomplished.
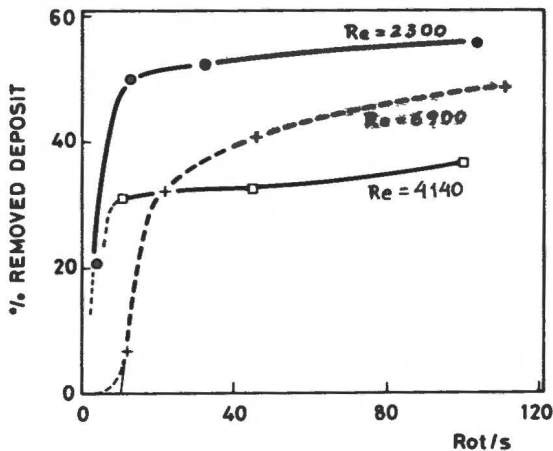

Figure 7 Results from the concentric cylinders apparatus

• The hard layer is practically unremovable at the operating conditions of the heat exchanger. In fact, by means of the correlation given by Wendt [17] it was possible to calculate the velocity of rotation of the outer cylinder that is necessary to produce a shear stress equal to the one used in the annular heat exchanger in each test. The following values were

estimated: 29 rot/s for Re = 2300, 52 rot/s for Re = 4140 and 78 rot/s for Re = 6900.

Using samples of the fouled tubes at position A, curves similar to the ones in Figure 7 were also obtained; however, the deposits in this entrance region contain only 20% - 40% of hard layer.

5. CONCLUSIONS

Particulate fouling tests were carried out in an annular heat exchanger at several Reynolds numbers using a water-kaolin suspension. The data can be satisfactorily described by the *generalized* model of Pinheiro, and the following conclusions can be drawn up for the case of non-laminar fully developped flow:

- At low Reynolds numbers, mass transfer controls the deposition process and, since the cohesion of the deposits is relatively high, the thickness of the deposits increases with the Reynolds numbers.

- At higher fluid velocities, adhesion is the controlling step, resulting in a decrease in the amount of deposit as Re is increased.

Tests carried out in a rotating cylinder apparatus supported the hypothesis concerning the relative cohesion of the deposits and, furthermore, confirmed the predicted existence of a *loose* and a *hard* layer in the deposits, the latter being a substancial fraction (50-70%) of the total mass of the deposit.

In the entrance zone of the heat exchanger, where a laminar developing flow exists, the results show a quite different behaviour, R_f^∞ always decreasing with increasing Reynolds numbers.

NOMENCLATURE

a,b   - empirical parameters in Pinheiro's model
C   - suspension concentration (kg/m^3)
E   - activation energy for adhesion (J/kg/mole)

f	-	friction factor of the inner surface of the annulus
h	-	convective heat transfer coefficient $(W/m^2\ k)$
h_0	-	convective heat transfer coefficient, clean wall $(W/m^2\ k)$
k_0, k_1, k_2	-	proportionality constants
k_f	-	thermal conductivity of the deposit $(W/m\ k)$
k_r	-	adhesion coefficient (m/s)
k_t	-	transport coefficient (m/s)
R	-	ideal gas constant $(J/kg\ mole\ k)$
Re	-	Reynolds number
R_f	-	fouling resistance at time t $(m^2\ k/W)$
R_f^∞	-	asymptotic fouling resistance, at time $t = \infty$ $(m^2\ k/W)$
R_w	-	thermal resistance between the wall thermocouple and the clean surface $(m^2\ k/W)$
t	-	time (s)
T_s	-	surface temperature (k)
u	-	mean fluid velocity (m/s)
u_{min}	-	minimum fluid velocity necessary for removal to occur (m/s)
U	-	overall heat transfer coefficient $(W/m^2\ k)$
U_0	-	overall heat transfer coefficient, clean wall $(W/m^2\ k)$
y_f	-	final thickness of the deposit (microns)
β	-	constant in equation 1 (s^{-1})
ϕ_d	-	deposition rate $(m^2\ k/J)$
ϕ_r	-	removal rate $(m\ k/J)$
ρ_f	-	density of the deposit (kg/m^3)

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Heat Transfer 1986

Proceedings of
The Eighth International Heat Transfer Conference
San Francisco, CA U.S.A.

IHTC 1986

Volume

6

General Papers

Edited by C. L. Tien, V. P. Carey, and J. K. Ferrell

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The International Scientific Committee
and **The U.S. Scientific Committee**