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Heat exchanger fouling in the pre-heat train of a crude oil distillation unit

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HEAT EXCHANGER FOULING IN THE PRE-HEAT TRAIN OF A CRUDE OIL DISTILLATION UNIT

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HEAT EXCHANGER FOULING IN THE PRE-HEAT TRAIN OF A CRUDE OIL DISTILLATION UNIT

1. NOTATION AND UNITS

		<i>SI Unit*</i>
A_i	total surface area of inside of tubes	m^2
A_o	total surface area of outside of tubes	m^2
A_w	tube mean wall area	m^2
a_1	coefficient in Equation (5.3)	$m^2 s K/kg^2$
a_2	coefficient in Equation (5.3)	$K s/m W$
c_p	fluid specific heat capacity	$J/kg K$
C_b	bulk particulate concentration in Equation (5.3)	kg/m^3
C_{pb}	bulk precursor concentration, see Equation (5.1)	kg/m^3
C_{fi}	foulant concentration in Equation (5.2)	kg/m^3
C_w	wall particulate concentration in Equation (5.3)	kg/m^3
d	inside tube diameter	m
d_o	outside tube diameter	m
E_{act}	activation energy	kJ/mol
f	tube inside wall Fanning friction factor, $f = \frac{\tau_w}{\frac{1}{2}\rho v_a^2} = \frac{\Delta p d}{2\rho v_a^2 L}$	—
H_D	precipitation ratio, see Section 3.3.2	—
I_N	insolubility number, see Section 3.3.2	—
j	number of crude oils in a mixture	—
k_r	reaction rate constant in Equation (5.1)	m/s
L	tube length	m
Pr	Prandtl number, $= (c_p \eta / \lambda)$	—
p	pressure	$N/m^2 (Pa)$
Δp	frictional pressure drop	$N/m^2 (Pa)$

See footnote at the end of table.

		<i>SI Unit*</i>
r_w	wall thermal resistance, see Equation (2.1)	K/W
R	gas constant ($= 8.314 \times 10^{-3}$ kJ/mol K)	kJ/mol K
R_f	fouling factor	m ² K/W
R_{fi}	inside tube fouling factor	m ² K/W
R_{fo}	outside tube fouling factor	m ² K/W
Re	Reynolds number, $= \rho v_a d / \eta$	–
S_{BN}	solubility blending number, see Equation (3.3)	–
T_f	crude oil film temperature, defined in Section 4.3.2	K
T_s	surface temperature at tube inside surface, see Equation (3.1)	K or °C
T_w	tube inside wall temperature	K or °C
t	time	s
U_o	overall heat transfer coefficient based on tube outside surface area, A_o	W/m ² K
v_a	superficial axial flow velocity in the tubes (volume flowrate divided by bare tube inside flow area)	m/s
v_f	axial flow velocity in fouled tube	m/s
V_i	volume of the i th crude in a mixture	m ³
x_f	deposit thickness, see Equation (5.1)	m
α	constant in Equation (4.2)	m ² K/J
α_i	heat transfer coefficient related to inside of tube	W/m ² K
α_o	heat transfer coefficient related to outside of tube	W/m ² K
β	removal constant, see Equation (4.2)	–
β_f	foulant mass transfer coefficient in Equation (5.2)	m/s
β_p	precursor mass transfer coefficient in Equation (5.1)	m/s
η	dynamic viscosity	Pa.s
γ	factor in Equation (4.2)	(m ² /N) (m ² K/J)
λ	thermal conductivity	W/m K
λ_f	deposit thermal conductivity, see Equation (5.1)	W/m K

See footnote at the end of table.

SI Unit*

ϕ	helix angle for helical baffle, defined in Section 4.2.1.1	deg
ρ	crude oil density	kg/m ³
ρ_f	deposit density, see Equation (5.1)	kg/m ³
τ_w	wall shear stress	N/m ²

Subscripts

f	refers to foulant layer or variable under fouled conditions
i	refers to inlet or to counter for components of a mixture of crude oils
o	outlet
mix	mixture

* This Data Item is presented in terms of SI units only. This avoids the need to use specific non-SI units for E_{act} and R which are expressed in terms of mol; expressing these important quantities in terms of, say, lb mole is possible but unusual so the use of a consistent dual unit system has been avoided. For users wishing to work in British/US customary units, conversion factors for some of the quantities are given below.

Some conversion factors

Quantity	Given in → Gives ←	multiplied by → divided by ←	Gives Given in
Length	m	3.2808	ft
	m	39.370	in
Pressure	N/m ² (Pa)	0.000 145	lbf/in ² (psi)
	N/m ² (Pa)	0.020 88	lbf/ft ²
Power	W	3.411 8	Btu/h
Area	m ²	10.764	ft ²
Mass	kg	2.204 6	lb
Volume	m ³	35.315	ft ³
	m ³	6.289 8	US barrel
Energy	J	0.238 8	cal
	J	9.480×10^{-4}	Btu

Temperature conversions

$$^{\circ}\text{C} = \frac{5}{9}(^{\circ}\text{F} - 32)$$

$$^{\circ}\text{F} = \frac{9}{5}(^{\circ}\text{C}) + 32$$

$$\text{K} = ^{\circ}\text{C} + 273.15$$

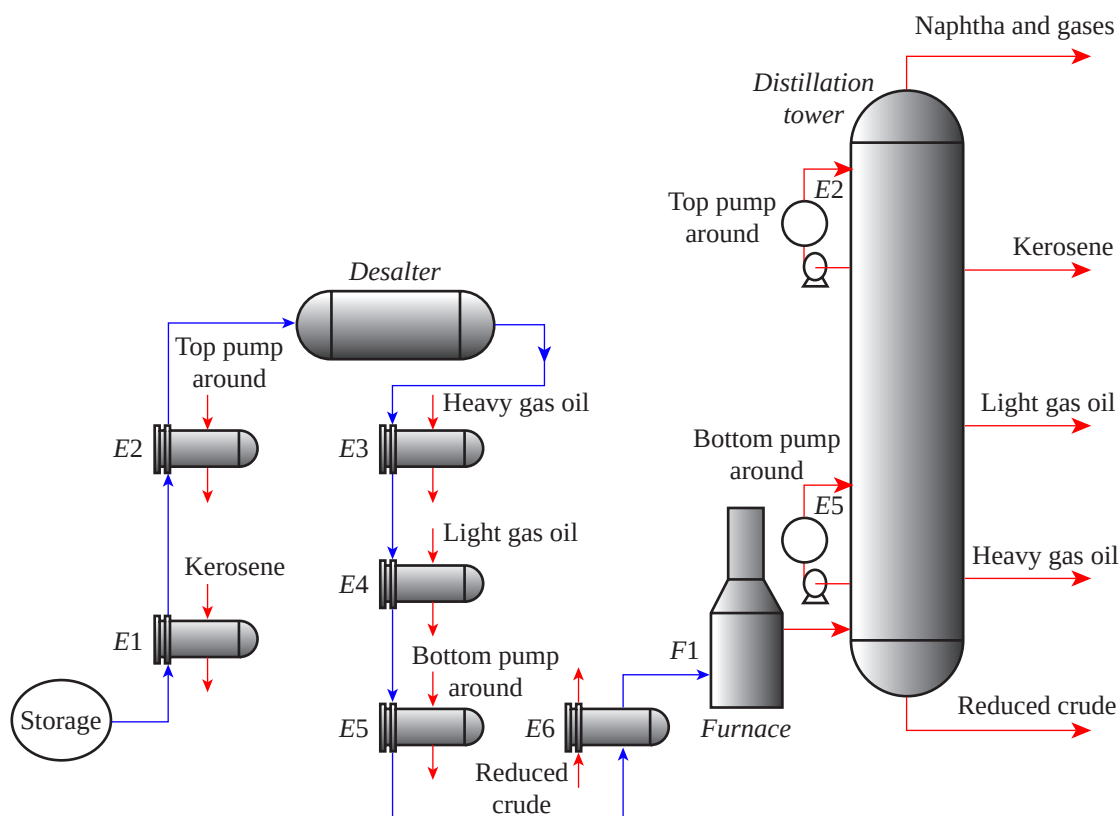
$$\text{R} = ^{\circ}\text{F} + 459.67$$

2. INTRODUCTION

2.1 Overview

This Data Item has been prepared as a result of a collaborative effort between ESDU and a group of oil refinery operators, heat transfer equipment manufacturers and chemical suppliers in recognition of the great importance of crude distillation unit fouling. The objective is to provide a practical user guide which will enable process engineers to review the impact of process conditions and equipment design changes on heat exchanger fouling rates in crude distillation units.

Currently, most engineers use fixed values of fouling factor such as those specified by the Tubular Exchanger Manufacturers Association (TEMA)⁷⁰; little account is taken of the various factors which influence fouling and there is clearly a need for an alternative approach. However, it should be stated at the outset that the problem is very complex and there are limitations on the understanding of the detailed processes which occur. Nevertheless, as this Data Item shows, there is an increasing body of knowledge which can be used to improve the design and operation of the system to decrease fouling propensity



Sketch 2.1 Schematic of typical crude distillation unit (from Panchal and Huang Fu⁶⁰, 1998)

A typical crude distillation unit is illustrated in Sketch 2.1. The crude oil coming from storage tanks is normally fed to the crude distillation unit at ambient temperature. The crude is then heated to around 110-150°C before being contacted with water in the desalter (see Section 3.9). The main purpose of the desalter is to remove inorganic ionic species (chlorides, fluorides, sodium, *etc.*); a secondary purpose is to remove particulate matter or “sludge” (sand, corrosion products, *etc.*). Downstream of the desalter, the crude oil then flows through the pre-heat exchanger train, which typically heats it up to around 230-300°C before it enters the furnace (also called the “fired” or “process” heater).

It should be stressed that Sketch 2.1 shows a somewhat oversimplified version of a crude pre-heat train; most systems are much more complex. Note also that the heating medium for heat exchangers E3-E5 is not arranged in order of increasing temperature. This is not uncommon and is used to match heat load and to take account of other factors such as accessibility.

Chemical reaction fouling arising from thermal causes is usually low in the heat exchangers upstream of the desalter, though fouling and plugging due to solids and salt deposition sometimes cause problems. Most of the salts in the crude oil are eliminated by the desalter before the crude oil enters the hotter part of pre-heat exchanger train. The crude oil becomes hotter as it flows through the pre-heat train, so thermal fouling on the crude side of the exchangers becomes more serious. This fouling (coupled with fouling on the hot side of the exchangers which can also be a problem) leads to a decline in furnace inlet temperature which leads to an increase in heat input required to the furnace or a decrease in the crude unit throughput or a combination of both.

The economic and environmental significance of fouling is discussed in Section 2.2, and Section 2.3 explores in more detail the region of occurrence of fouling in the units. Section 2.4 gives a brief general background on fouling mechanisms in the particular context of crude distillation units. Section 3 discusses the factors influencing the fouling (temperature, flow velocity, oxygen, etc.) and Section 4 discusses fouling mitigation and control (chemical, mechanical and operational).

Section 5 discusses prediction of fouling including a survey of existing models. Section 6 describes a new methodology for identifying the regions of significant fouling within a crude pre-heat train and discusses computer programs developed by ESDU for identifying shell-and-tube heat exchanger configurations in which fouling is minimised: further guidance on these programs is given in Appendix A. Finally, Section 7 outlines the possibilities for further development.

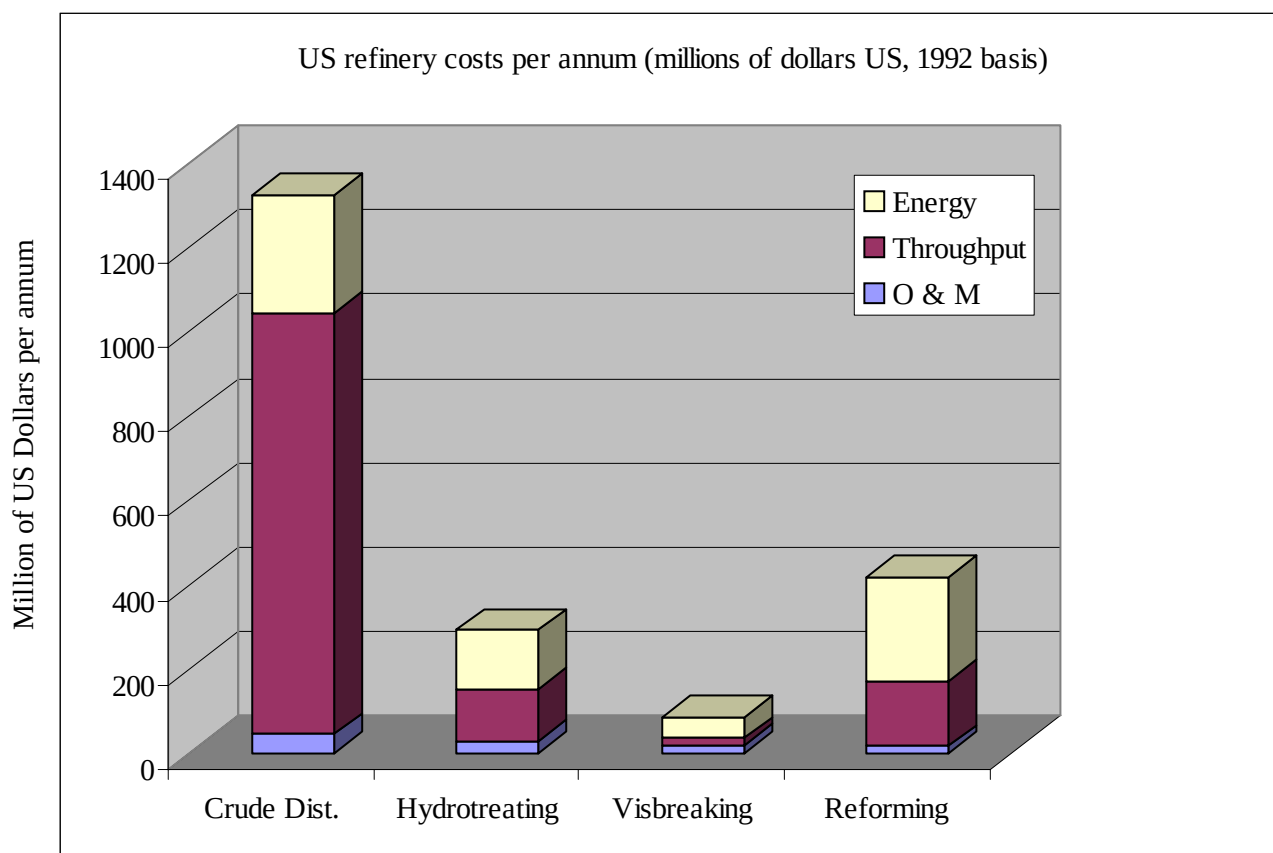
2.2 Economic and Environmental Significance of Fouling

Heat exchanger fouling in general is a major economic problem, accounting for 0.25% of the gross national product (GNP) in the highly industrialised countries. Müller-Steinhagen⁴⁹ (1995) estimates that the total cost of all heat exchanger fouling in the UK is of the order of \$2.5 billion US and the equivalent cost in the USA is \$14 billion US. Müller-Steinhagen⁴⁹ (1995) categorised the total fouling-related costs in four main areas:

- (1) **Energy costs and environmental impact.** In the case of the crude oil distillation unit, this corresponds to the additional fuel required for the furnace due to the failure of the pre-heat train to produce the required temperature for the separation process. In principle, energy losses due to increased pressure drop (pumping power) may also be significant. However, the normal practice is to oversize the pump and control the flow using valves. The use of more fuel leads to additional production of CO₂ with the associated environmental impact.
- (2) **Production loss during shutdowns due to fouling.** These are often considered to be one of the more significant costs but they are very difficult to estimate. If the pre-heat train throughput is furnace-limited, a typical 10% loss of production due to taking a heat exchanger out of service in a 100 000 US barrel /day plant would cost \$20 000 US per day (assuming \$2 per US barrel of marginal lost production). After shutdown, there is an additional cost due to out-of-specification production after production is restarted.
- (3) **Capital expenditure.** This includes excess surface area, costs for stronger foundations, provisions for extra space, increased transport and installation costs, costs of anti-fouling equipment and costs of installation of on-line cleaning devices and treatment plants and increased cost of disposal of the (larger) replaced bundles and, finally, the (larger) heat exchanger.

* 1 US barrel = 0.1589 m³.

- (4) **Maintenance costs.** This includes staff and other costs for removing fouling deposits and the cost of chemicals or other operating costs of anti-fouling devices. There is also an economic and ecological cost associated with disposal of cleaning chemicals after cleaning.



Sketch 2.2 Fouling costs in US refineries in 1992 US Dollars (estimates of van Nostrand *et al*⁷³ (1981) as adjusted for inflation by Panchal⁶⁰ (1995))

The cost of fouling in US refineries was estimated by van Nostrand *et al.*⁷³ (1981), whose figures were adjusted to account for inflation by Panchal⁶⁰ (1995). The resultant costs obtained by Panchal are illustrated in Sketch 2.2 which shows that the costs of fouling in the crude distillation units in US refineries was around 1300 million US dollars per annum. Making the same adjustments to the estimates by van Nostrand *et al.* of the total crude distillation unit fouling costs in the western world produces a figure of around 4500 million US dollars per annum. As will be seen from Sketch 2.2, the cost of fouling in the crude distillation unit is the dominant fouling cost in a refinery, hence the focus of the present study into this area. Some other points to note about the economic significance of fouling are as follows:

- (a) In the early days of refining, when refineries were built at or near oil fields, energy was in general very cheap; associated gas could often be used. However, when hydrocarbon products were transported over large distances before refining, energy costs became a very significant part (typically 25-50% depending on oil price) of the total costs which is not surprising when it is realised that energy utilisation for the whole refinery accounts for around 6% of the oil throughput. The energy utilisation in the crude unit accounts for 1-2% of the throughput.
- (b) Since fouling costs are so high, refinery operators should in principle take account of these costs when deciding to process a given crude. Often, it is not possible to do this explicitly and the availability of simple models for fouling would be a considerable aid in refinery management.

- (c) The importance of fouling in the pre-heat train can be judged from the estimate by Panchal and Huang Fu⁵⁸ (1998) that a loss of 1 deg C in the temperature of the crude oil entering the furnace in a 100 000 barrel/day crude unit equates to 450 kW and an energy cost of 40 000 US dollars per annum. Baudelet and Krueger⁵ (1999) estimate that an increase of furnace inlet temperature of 1 deg C is equivalent to 1 tonne of fuel saved per day, which may translate into additional costs of \$100 000 per year. The cost will depend on fuel costs and throughput.

If the temperature at the exit of the crude pre-heat train falls as a result of fouling, then extra fuel will be needed for the furnace and the combustion of this extra fuel will lead to an additional production of CO₂. A chart for determining costs due to a reduction in furnace inlet temperature is shown in Figure 1, where the lower chart shows the additional production of CO₂.

In view of the great economic significance of fouling in crude distillation units, it is perhaps surprising that the research effort attacking the problem is relatively small. This reflects the difficulty of defining and financing a research approach which is sufficiently generic to provide solutions to a wide range of cases. Crude oil is a very variable material and a set of actions in one case may be inappropriate in another. A major objective of the present exercise is to try to identify generic themes for research which can be pursued in the future.

2.3 Fouling Characteristics in a Simplified Crude Pre-heat Train

As discussed in Section 2.1, the most serious consequences of fouling are found upstream of the desalter (exchangers *E1* and *E2* in Sketch 2.1) and in the exchangers immediately upstream of the furnace (*E5* and *E6* in Sketch 2.1). The focus of this current document is on the latter zone. The fouling propensity for the desalted crude increases with increasing temperature. A useful compilation of heat exchanger fouling rates in a simplified crude train is given by Panchal and Huang Fu⁵⁸ (1998) and is shown in Table 2.1. In the case of multiple shell arrangements (heat exchangers *E2*, *E4*, *E5* and *E6*), two parallel crude oil streams are used for ease of cleaning. It will be noted that the fouling rate increases by an order of magnitude as the crude temperature increases between heat exchanger *E4* and heat exchanger *E6*. It should be emphasised that the increased temperatures and potential for corrosion may lead to a change in materials at the hot end of the pre-heat train; stainless steels (usually NiCr½Mo) are occasionally used, depending on the crude (*e.g.* for naphthenic crude).

2.4 Other Crude Unit Fouling Problems Not Covered in this Data Item

Though the main focus of attention of this Data Item is the pre-heat train, fouling is also important in other parts of the crude unit, as exemplified by the following:

- (1) Upstream of the desalter, plugging and/or corrosion may potentially occur as a result of salt deposition or asphaltene precipitation. This is not a frequent occurrence.
- (2) Pre-heat trains with low fouling are not usually associated with fouling problems in the furnace. However, it is sometimes thought that reducing fouling propensity in the heat exchangers will increase fouling propensity in the furnace. It should be pointed out that the foulant deposited in the exchangers is only a small proportion of the total foulant (asphaltenes, say) which flows through the system. Thus, in practice, there may be little interaction in fouling between the heat exchangers and the furnace.

Both inorganic and asphaltene fouling can occur in the furnace but it should be remembered that the furnace tubes have a larger diameter than those in the pre-heat exchangers and a given fouling layer would have less tendency to plug the tubes. Also, the temperature driving force is larger and is limited partially by radiation to the outer surface of the tubes, and the rate of heat transfer is

much less affected by the fouling on the inside of the tubes.

Nevertheless, fouling in heaters is significant in giving a closer approach to limitations in outside tube wall ("skin-point") temperatures. A 1 mm layer of coke can give a 10 deg C increase in metal temperatures at an average radiant section heat flux and 20 deg C increase at the point of maximum heat flux. The flux at different points in a heater for one-sided firing can vary by around a factor of 2.

- (3) Fouling in the kerosene/stripper reboiler may also present a problem. Fouling associated with nucleate boiling is more serious than fouling at higher qualities where nucleate boiling may be suppressed. The problems can be avoided under some circumstances by the use of steam stripping (that is, introducing steam as the bottom of the distillation column rather than using a reboiler).
- (4) Overhead condensers can also show significant propensity to fouling, though this may be mitigated by attention to detail in the desalting step. Fouling on the shellside of shell-and-tube exchangers or in the tubes of air-cooled heat exchangers (condensers) are two manifestations of this.
- (5) Fouling on the trays of the distillation column may be a problem.

TABLE 2.1 OPERATING PARAMETERS AND FOULING RATES IN CRUDE DISTILLATION UNIT SHOWN IN SKETCH 2.1

Parameters		Heat exchangers						
		<i>E1</i>	<i>E2</i>	<i>E3</i>	<i>E4</i>	<i>E5</i>	<i>E6</i>	<i>F1</i>
Area, m ²		208	1152	208	523	1478	1344	13 239
Shells		1	4	1	2	4	4	
								
Tubes per shell		916	1256	907	911	1288	1464	
Tube i.d., mm		21.1	21.1	21.1	21.1	21.1	21.1	
Tubeside flow area, m ²		0.160	0.220	0.159	0.159	0.226	0.256	
Product stream		Kerosene	Top pump around	Heavy gas oil	Light gas oil	Bottom pump around	Reduced crude	
Crude flow, kg/h		489 000	489 000	489 000	489 000	489 000	489 000	
Product flow, kg/h		54 900	953 400	387 300	128 900	374 100	244 700	
Crude velocity, m/s		1.05	0.41	1.23	0.66	0.50	0.47	
Reynolds number		2673	2315	21 550	21 838	24 845	36 873	
Prandtl number		921	429	145	79	54	35	
Heat transfer coefficient, W/m ² K								
Tubeside		390	320	1230	980	910	1020	
Overall		340	280	860	690	590	540	
Average fouling rate, (m ² K/kW)/day		0.018	0.026	0	0.003	0.014	0.034	
Tubeside temp., C	Inlet	20	35	101	178	201	252	296
	Outlet	35	101	178	201	252	296	382
Shellside, temp., C	Outlet	45	168	241	180	209	256	
	Inlet	226	148	371	299	299	374	
Heat duty, MW		5	23	30	10	23	21	45
Pre-heat train total, MW						112	45	

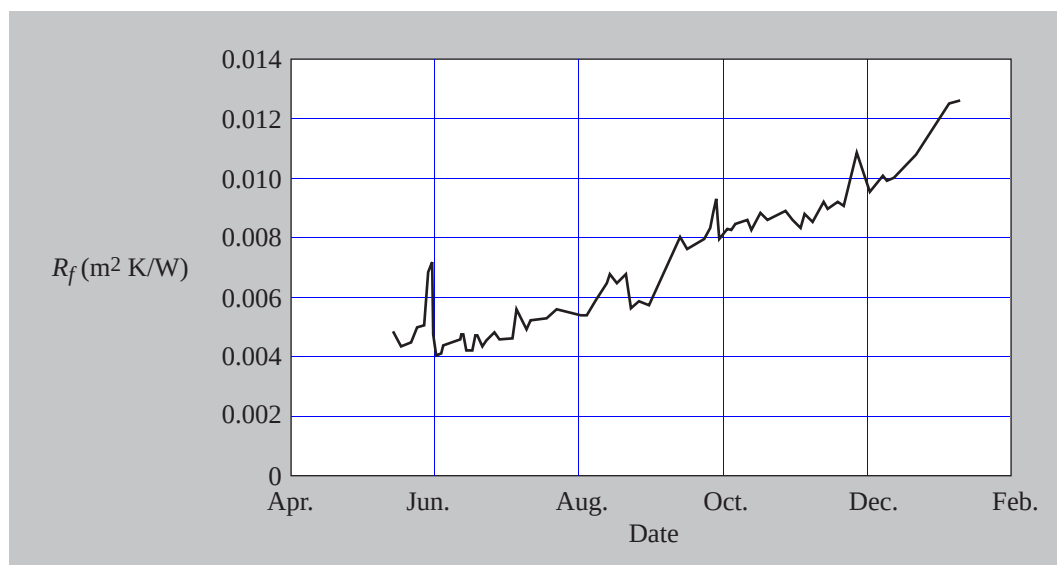
2.5 Generic Types of Fouling and their Significance for Heat Transfer and Pressure Drop in Crude Units

It is convenient at this point to define fouling resistance from the normal definition of overall heat transfer coefficient. The equation for overall heat transfer resistance has the form:

$$\frac{1}{U_o A_o} = \frac{1}{\alpha_o A_o} + \frac{R_{fo}}{A_o} + r_w + \frac{R_{fi}}{A_i} + \frac{1}{\alpha_i A_i} \quad (2.1)$$

where U_o is the overall heat transfer coefficient based on the outside surface area of the tubes, A_o ; α_i and α_o are the heat transfer coefficients for the inside and outside of the tubes (based on the inside and outside areas A_i and A_o), R_{fi} and R_{fo} are the fouling factors for the inside and outside of the tubes respectively and r_w is a tube wall thermal resistance. The wall resistance is given as $r_w = x_w / (\lambda_w A)$ for a flat plate geometry ($A = A_o = A_i$) where x_w is the wall thickness and λ_w is the thermal conductivity of the wall material. For a tubular system, $A_o/A_i = d_o/d$, where d_o is the outside tube diameter and d the inside tube diameter. In this case $r_w = d_o / (2\lambda_w A_o) \ln(d_o/d)$.

In what follows, the main focus is on fouling on the inside of tubes and is, hence, on R_{fi} rather than R_{fo} . The symbol R_f (that is, without the additional subscript i) should be taken to refer to inside-tube fouling unless otherwise stated.



Sketch 2.3 Example of variation of fouling factor with time in a typical pre-heat train heat exchanger (Jones and Bott³⁶, (1998)).

Jones and Bott³⁶ (1998) cite an example of fouling in a typical crude pre-heat train heat exchanger where the tubeside fluid was crude oil and the shellside fluid was atmospheric residue. The fouling factor (the sum of the fouling factor on both sides, though dominated by the crude oil side) was determined and varied with time as shown in Sketch 2.3. The fouling factor increased threefold over an eight month period. It is noteworthy that the fouling factor even at the beginning of the period was much greater than that recommended by the Tubular Exchangers Manufacturers Association⁷⁰ (TEMA). This underlines the problems which led to the generation of the present Data Item.

2.6 Influence of Fouling on Pressure Drop

The formation of deposits on heat transfer surfaces causes an increase in frictional pressure drop due to increased surface roughness and restricted cross-sectional flow area. According to Chenoweth¹⁴ (1987), more heat exchangers are taken out of service because of excessive pressure drop than because of reduced heat transfer. A rough estimate of the pressure drop inside a fouled tube can be made if it is assumed that the deposit is distributed evenly over the tube wall. The frictional pressure drop in cylindrical tubes is calculated from Equation (2.2)

$$\Delta p = 2f \frac{\rho L v_a^2}{d}, \quad (2.2)$$

where L is the total length of the tube run. The friction factor f may be obtained from charts or calculated from a variety of formulae. One of the most widely used is the Colebrook-White equation:

$$\frac{1}{\sqrt{4f}} = -0.86 \ln \left[\frac{\varepsilon/d}{3.7} + \frac{2.51}{Re \sqrt{4f}} \right] \quad (2.3)$$

where ε is the roughness height, d the tube inside diameter and Re the Reynolds number ($= \rho v_a d / \eta$). Equation (2.3) is implicit in nature and requires iterative solutions; an explicit equation which fits this expression is that of Chen¹³ (1979):

$$\frac{1}{\sqrt{4f}} = -2.0 \log \left\{ \frac{\varepsilon}{3.7065d} - \frac{5.0452}{Re} \log \left[\frac{1}{2.8257} \left(\frac{\varepsilon}{d} \right)^{1.1098} + \frac{5.8506}{Re^{0.8981}} \right] \right\}. \quad (2.4)$$

If the fouling factor, R_f , and the thermal conductivity, λ_f , of a layer of foulant deposits are known or can be estimated, the inside diameter of the flow area of a fouled tube, d_f , can be determined by Equations (2.5) and (2.6)

$$R_f = \frac{d \ln(d/d_f)}{2\lambda_f} \quad (2.5)$$

$$d_f = d \exp(-2\lambda_f R_f / d). \quad (2.6)$$

The friction factor for the fouled tube, f_f , can be obtained from either Equation (2.3) or Equation (2.4) by substituting d_f for d , $Re_f = \rho v_f d_f / \eta$ for Re (where $v_f = v_a (d/d_f)^2 = V_a (d/d_f)^2$) and by inserting a suitable value for ε . The choice of ε clearly depends on the particular fouling situation but a typical value of ε for a fouled surface might be around 0.1 mm. The pressure drop is then calculated from the expression:

$$\Delta p_f = 2f_f \frac{\rho L v_f^2}{d_f}. \quad (2.7)$$

In order to calculate d_f from Equation (2.6), a value of λ_f is required. Values of the thermal conductivity of fouling deposits are tabulated in the VDI - Wärmeatlas⁷⁴ (1994). Values are given for gas-oil deposits in the range 0.3–1.1 W/m K and for coke in the range 0.3–0.9 W/m K. For conservative estimates of the increase in pressure drop, therefore, the **higher** end of this range should be taken (a value of 1 W/m K, say).

The effect of fouling on the shellside pressure drop can be estimated using Table 2.2, taken from Coulson *et al.*¹⁵, (1985).

TABLE 2.2 Ratio of Fouled to Clean Shellside Pressure Drop

Reciprocal of fouling factor (W/m ² K)	Shell diameter/baffle spacing		
	1.0	2.0	3.0
Laminar flow: $1/R_f = 6000$ $1/R_f = 2000$ $1/R_f < 1000$	1.06 1.19 1.32	1.20 1.44 1.99	1.28 1.55 2.38
Turbulent flow: $1/R_f = 6000$ $1/R_f = 2000$ $1/R_f < 1000$	1.12 1.37 1.64	1.38 2.31 3.44	1.55 2.96 4.77

2.7 Types of Fouling

The categorisation of fouling into various types has become fashionable, and a typical classification is that of Epstein²⁶ (1983) who suggests that the main mechanisms are those of crystallisation fouling, particulate fouling, chemical reaction fouling, corrosion fouling and biological fouling. He classifies knowledge in the area in terms of information available about initiation, transport, attachment, removal and ageing of the fouling layers. Epstein defines chemical reaction fouling as the formation of deposit at the heat transfer surface by chemical reaction, in which the surface itself is not a reactant. A characteristic of chemical reaction fouling is that, in common with other chemical reactions, the rate of reaction increases exponentially with temperature. The formation of polymeric material during transport of the crude through the heat exchanger was originally thought to be an important factor. This rate of formation was expected to be very sensitive to the presence of oxygen, as indeed it is. However, the **change** in the concentration of the polymeric material as the crude passes through the heat exchanger is small compared to the concentration of the material entering the heat exchanger and the formation process is not, therefore, dominant, as is confirmed by recent work. The crude oil organic fouling mechanism is a complex process, and is primarily due to the destabilisation of the asphaltene component of crude oil, its deposition at the tube wall and its subsequent degradation to a carbonaceous fouling layer.

Asphaltene is generally found in a disperse colloidal micellar form, with the asphaltene at the centre of the micelle, surrounded by a stabilising layer of resinous species. These species become increasingly saturated with distance from the micelle centre. At the edge of the micelle, the carbon/hydrogen ratio approximates to that of the bulk phase, and it is this gradually increasing saturation which affords solubility to the micelle.

The bulk phase contains paraffinic constituents which, under certain conditions, are capable of dissolving higher molecular weight hydrocarbons. The lightest of these paraffins contribute little to the overall carbon/hydrogen ratio of the hydrocarbon stream but are capable of removing many of the outermost resin species of the asphaltene micelle.

When the micelle comes into contact with paraffinic fractions, or when the temperature of the oil is raised, the outermost stabilising resins are removed by the paraffinic components and are liberated into the oil bulk. This leaves an exposed core of asphaltene which is no longer held in solution by a resinous shell. The asphaltene micelle, stripped of its solvating shell, has a low degree of saturation at its outer edge relative to that of the liquid phase. It is no longer stabilised in the oil bulk, and precipitation is more favourable.

The newly exposed asphaltene is highly polar and exerts attractive electrostatic forces upon other asphaltene species, resulting in the aggregation of asphaltene into a dense and insoluble particle. This aggregate may migrate from the turbulent bulk of the oil to the laminar sub-layer, where its residence time near the tube surface is vastly increased.

Deposition of destabilised asphaltene takes place by two consecutive processes. The first is the migration of the asphaltene aggregate to the metal surface, which may be due to gravitational, kinetic or electrostatic forces. At this point the thermodynamics of the aggregate-tube wall interaction dictate whether solvation will continue or whether the aggregate will be adsorbed onto the metal surface. The competition between adsorption and solvation is greatly influenced by the geometry and roughness of the surface at the point of contact.

If adsorption is favoured, the asphaltene aggregate is deposited on the metal surface. This begins at localised sites, where the thermodynamics of adsorption are the most favourable. With time, additional fouled sites continue to appear on the metal surface until a coating of asphaltene is produced. At high tube temperatures, this asphaltene layer undergoes degradation, through polymerisation and dehydrogenation mechanisms, to progressively more carbonaceous species.

The chemistry of asphaltene degradation involves a myriad of chemical reactions occurring simultaneously. The following are some of the major reactions types involved.

- C-H, C-C bond cleavage to form reactive free radicals,
- molecular rearrangement,
- thermal polymerisation,
- aromatic condensation,
- elimination of side chains and hydrogen.

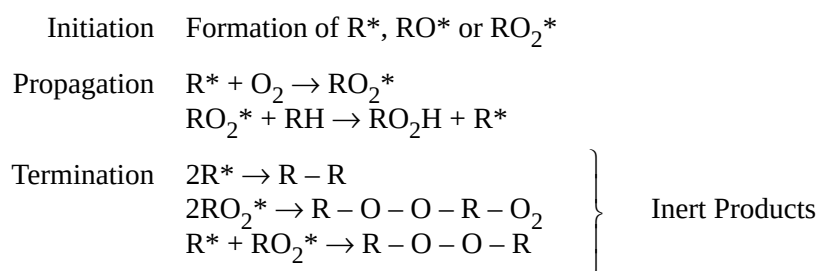
An important step in the early stages of coking is thermal rearrangement, where either an initial component or a rearranged entity serves as the building block in the further stages of degradation. One of the factors which makes coking so complex is the presence of so many polymerisation sites in an asphaltene molecule. The polymerisation processes for asphaltene can produce either condensed or non-condensed polymers, which vary considerably in structure.

As the fouling layer degrades to coke it becomes hard and brittle, and begins to fracture, leaving small cracks in the foulant surface. These cracks behave as energetically favourable sites for deposition in much the same way as the roughness of a clean surface. Asphaltene deposition continues at these sites and the fouling process begins again.

Studies have indicated that the rate of fouling in crude oil systems increases exponentially with temperature in a manner analogous to chemical reaction fouling. However, as will be seen from the above, the process is subtly different to a chemical reaction process; the destabilisation of the asphaltenes is very dependent on temperature (hence the approximately exponential law) but is also influenced by many other factors, and in particular by the crude composition.

Organic foulants are either already present in the crude or are formed through a number of pathways, the most common of which are autoxidation, pyrolysis, condensation and polymerisation. Autoxidation has been the subject of numerous scientific papers, with a summary being given by Braun and Hausler⁹ (1976). The processes are extremely complex and it is beyond the scope of the present Data Item to give details.

However, it might be helpful to give a summary of some typical reactions that take place. These occur in three stages as follows:



Here, the * denotes a free radical and the initiation of the process results from the formation of such free radicals by catalysts, thermal reactions between oxygen and an olefin, redox reactions, *etc.* The larger molecular species formed as inert products in the termination phase may, for example, form part of the depositing species. However, it should be stressed that, in normal crude pre-heat train situations, the oxygen content is usually quite low and the formation of new polymers by reactions of the above type is likely to be of low significance. The situation is dominated by the asphaltenes already present in the crude, the fouling process being governed by their destabilities and aggregates as discussed above.

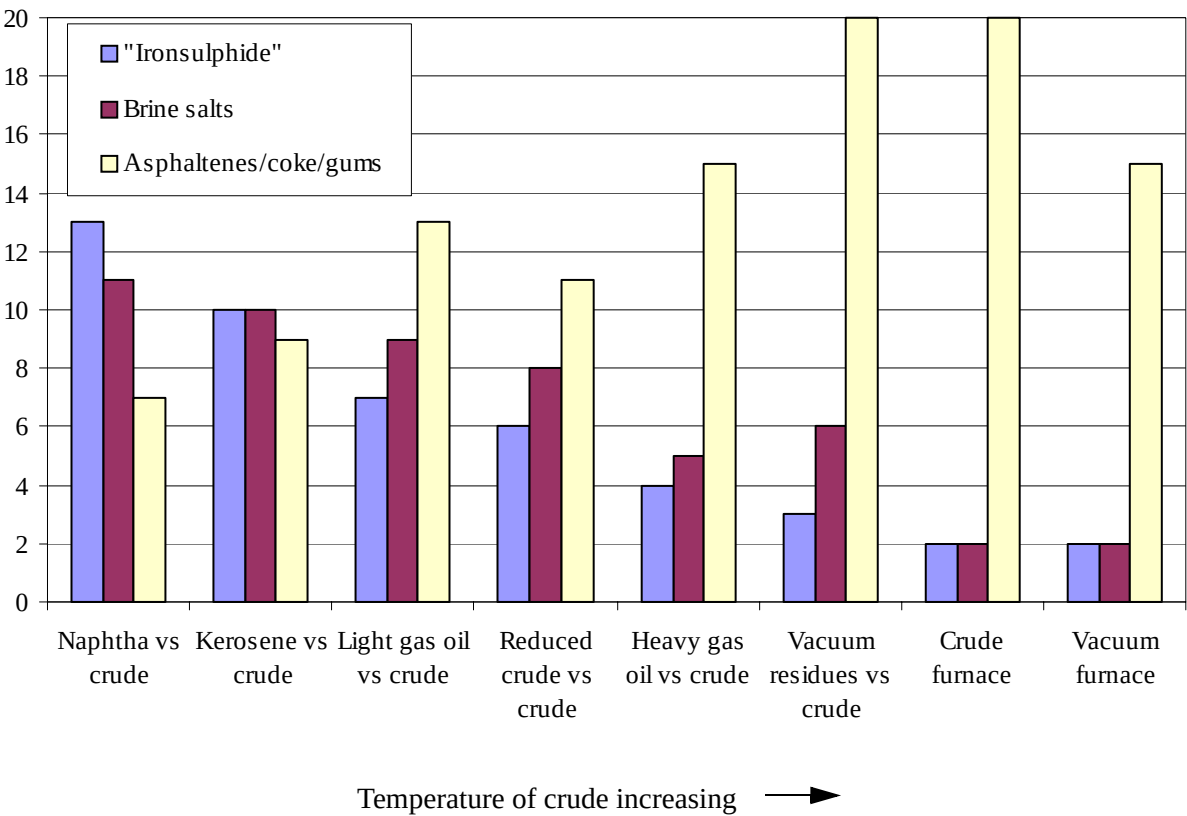
Though there is a vast literature on fouling, specific studies relating to crude oil fouling are fewer in number. These include studies of fouling in operational heat exchangers and also studies on fouling with crude oil streams in experimental apparatus. Table 2.3 summarises these studies.

TABLE 2.3 STUDIES OF CRUDE OIL FOULING

Authors	System	Maximum wall temperature or wall temperature range (°C) studied
Weiland <i>et al.</i> ⁸⁰ (1949)	Pre-heat train	310
NACE report ⁵² (1970)	Pre-heat train	400
Scarborough <i>et al.</i> ⁶⁶ (1979)*	Single-heated tube	365-475
Latos and Franke ⁴⁴ (1982)*	Hot wire probe	175-400
Lambourne and Durrieu ⁴² (1983)	Pre-heat train	374
Eaton and Lux ²⁴ (1984)*	Heated probe	287
Dickakian and Seay ²² (1988)*	Single-heated tube	232-266 (bulk)
Crittenden <i>et al.</i> ²¹ (1992)	Pre-heat train	250
Crittenden <i>et al.</i> ²⁰ (1993)*	Single heated tube (with inserts)	197-218
Ebert and Panchal ²⁵ (1995)	Heated probe	376-467
Asomaning and Watkinson ³ (2000)	Heated probe	160-240

* These measurements were taken with a recirculation system which may give results which are unrepresentative of the real (once-through) situation.

It should be emphasized that the nature of the fouling process will vary from heat exchanger to heat exchanger in the pre-heat train. Fouling in the earlier (upstream of the desalter) exchangers in the train will be dominated by inorganic fouling (brine salts and iron sulphide, *etc.*) whereas the later exchangers (downstream of the desalter) will have a preponderance to fouling by organic foulants (asphaltenes, *etc.*). A diagram illustrating this change is that of Lemke⁴⁵ (1999) which is reproduced here as Sketch 2.4; note that the crude temperature increases from left to right. The first two exchangers are upstream of the desalter.



Sketch 2.4 Occurrence of types of foulant in pre-heat train heat exchangers (20 = very likely, 0 = very unlikely) (Lemke⁴⁵ (1999))

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3. FACTORS INFLUENCING FOULING

3.1 Effect of Surface Temperature

A frequently used relationship for the effect of surface temperature on fouling rate is of the Arrhenius type as follows:

$$\frac{dR_f}{dt} = A \exp\left(\frac{-E_{act}}{RT_s}\right) \quad (3.1)$$

where A is a parameter fitted to the data (it has the units $\text{m}^2 \text{K/J}$), E_{act} is an activation energy (typically expressed in kJ/mol) and T_s is the *absolute* temperature of the interface between the fluid and the deposit surface (K). Equation (3.1) implies that a plot of $\ln(dR_f / dt)$ against $1/T_s$ would be a straight line, from which the values of E_{act} and A may be determined. In applying Equation (3.1), it is implicit that all other factors (velocity, geometry, fluid composition) should remain constant. Crittenden *et al.*²¹ (1992) fitted the following equation to data for light crude oil:

$$\frac{dR_f}{dt} = 4.9 \times 10^{-7} \exp\left(\frac{-33}{RT_w}\right) . \quad (3.2)$$

A variety of activation energies have been observed for crude oils. As implied in Equation (3.2), Crittenden *et al.*²¹ obtained a value of 33 kJ/mol for light crude oils; their value for heavy crude oils is 21 kJ/mol . Scarborough *et al.*⁶⁶ (1979) and Eaton and Lux²⁴ (1984) obtained values of 53 and 36 kJ/mol respectively for crude oil feedstocks. It should be stressed that these values are based on *wall* temperatures (as measured by thermocouples in the wall); it should be remembered that the temperature varies away from the wall and this will affect the situation. The temperature falls from its value at the wall, T_w , to its value at the interface, T_s , and then decreases further to its value in the bulk fluid. It should also be stressed that Equations (3.1) and (3.2) are for situations in which the fluid flow conditions are constant; specifically, the experiments were carried out at constant velocity and the effect of variation of velocity was not covered. Velocity effects are discussed in Section 3.2. There is also some discussion in the literature about whether T_w or T_s is the appropriate temperature to use in flowing system. Ebert and Panchal²⁵ (1995) use the “film temperature” (T_f) (that is, the mean of the bulk and wall temperature) in their analysis of fouling rates; this approach is dealt with in more detail in Section 4.3.2.

3.2 Effect of Flow Velocity

Typically, in a crude pre-heat train, fouling decreases with increasing velocity. Care has to be taken in the interpretation of data on the effect of flow velocity on crude oil fouling. For a given bulk temperature and heat flux, the principal effect of increasing velocity may be to *decrease the fouling rate due to the fact that the heat transfer coefficient increases and the wall temperature therefore decreases*. On the other hand, if the fouling rate is controlled by mass transfer to the wall region of species present in the bulk fluid, then the mass transfer coefficient from the bulk to the near-wall region will increase with increasing velocity and this may give rise to an increase in fouling rate with increasing velocity.

For a situation in which the fouling rate for a given interface temperature *decreases* with increasing velocity, the influence of velocity may be manifested in two ways:

- (1) If the deposit is weak, then the shear stress at the wall (which increases with increasing velocity) may give rise to an erosion of the fouling layer which offsets the deposition of the foulant.

- (2) If the foulant material is formed in the fluid boundary layer adjacent to the hot surface (where the formation rate may be highest), then this formed foulant may diffuse back into the bulk fluid. An increase in velocity would increase the rate of mass transfer of the foulant and may reduce the fouling rate.

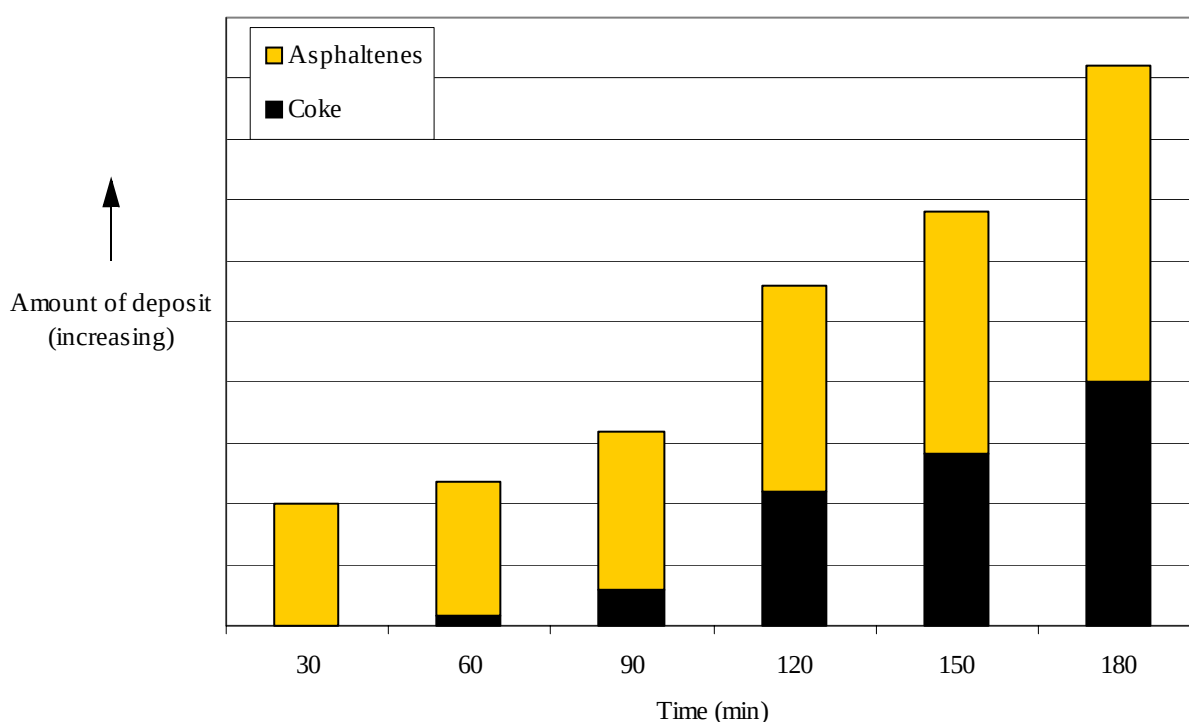
It is not surprising, therefore, that the results for the effects of velocity on the fouling of crude oils are rather inconclusive (Nelson⁵⁴ (1969), Scarborough *et al.*⁶⁶ (1979), Dickakian and Seay²² (1988) and Crittenden *et al.* (1992)). However, it seems probable that fouling can be avoided by having a sufficiently high wall shear stress; we shall return to this point in Section 4.3.2.

An effect associated with the effect of velocity is that of *residence time*. There is some evidence that, in passing through a heat exchanger, the crude tends to “cook” and, in large heat exchangers, the behaviour of the material may be different near the entrance to that near the exit. Both temperature and time may therefore be important variables.

3.3 Effect of Oil Properties

3.3.1 Crude composition

Crude oil is an extremely variable material. A crucial factor governing fouling is the amount of *asphaltenes* present in the crude. “Heavy” crudes tend to have higher asphaltene contents and are more susceptible to fouling. Asphaltene is the highly condensed aromatic material which gives many oils their black colour. Ashphaltenes have a molecular weight ranging from 1000 to 300 000 g/mol and are defined as a class of crude oil constituents insoluble in non-polar solvents such as pentane, hexane or heptane but soluble in solvents such as pyridine, carbon disulphide, carbon tetrachloride, toluene or benzene (Watkinson⁷⁷ (1992)). Dickakian and Seay²² (1988) studied the effect of asphaltenes on thermal fouling. They were able to characterise the deposits formed on the heat surfaces at various times, showing that the deposits were initially asphaltene precipitates which were then carbonised on the surface into an infusible coke. A typical time sequence is shown in Sketch 3.1.



Sketch 3.1 Effect of asphaltenes on fouling (Dickakian and Seay²² (1988)).

Dickakian and Seay²² (1988) concluded that most crude oil fouling in heat exchangers proceeds in the following manner:

- (1) Incompatibility between the asphaltenes and the oil initiates precipitation of some asphaltenes.
- (2) The precipitated asphaltenes adhere to the hot metal surface.
- (3) The asphaltenes then carbonise into infusible coke, which produces an irreversible lowering of heat transfer coefficients which is unrecoverable without off-line cleaning.

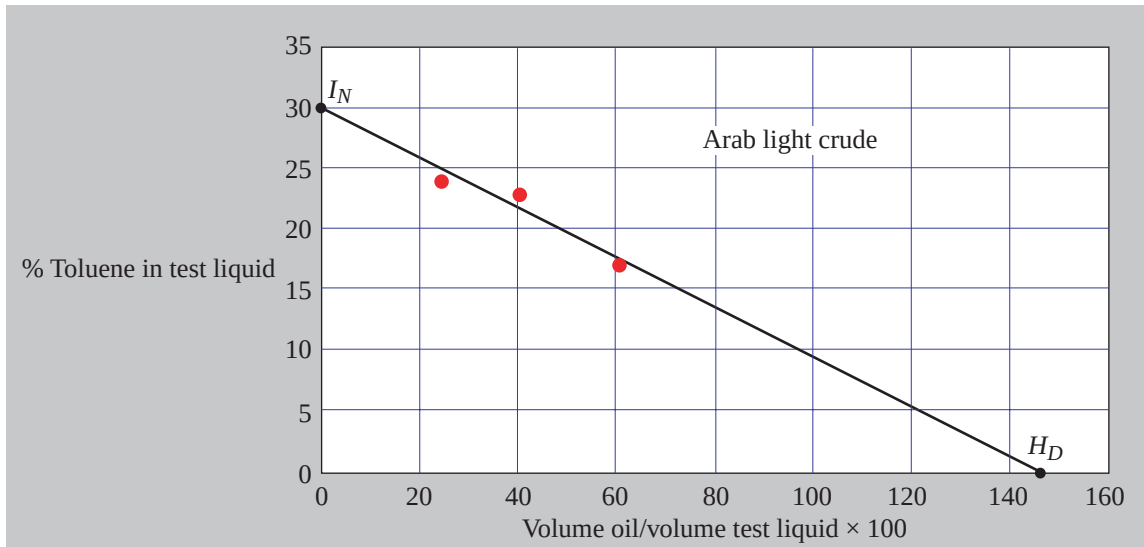
Thus, asphaltenes may be regarded as the most important crude oil component in governing fouling. Important factors relating to asphaltenes are their combination of low solubility and high thermal reactivity. The asphaltenes have the highest thermal reactivity of any fraction of a crude oil. While soluble, asphaltenes react to form lower molecular weight products but when insoluble the major thermal reaction product is coke (Wiehe⁸¹ (1993)).

3.3.2 Crude mixing

In practical situations, another factor which is of great importance is the presence in the crude feedstock of *oily slops* and *oily sludge*. Oily slops can, for instance, be re-run, off-specification material of various types. The addition of slops to the feed stream is seen as a means of recovering a difficult material; this may cause additional fouling problems. Oily sludge on the other hand is material which arises from the bottom of crude storage tanks and can also be a major source of problems in the crude pre-heat train. One oil company eliminated the sludge by using settling systems and this gave much longer operation times. The problem still arises as to what to do with the separated material. Oily sludge or slops can also give problems in the desalter by inhibiting demulsification and causing loss of crude to the aqueous outlet stream. A typical operator reported up to 4% loss of crude oil by this means. Again, there is a considerable problem of disposing of the oil-contaminated aqueous stream.

Another important factor in managing crude oil fouling is that of *mixing* of various crudes. Mixing crude types prior to their entry into the heat exchanger system can have a dramatic effect on fouling behaviour. Crude oils are broadly categorised as *parafinic* or *asphaltenic*. Mixing typical parafinic crudes or condensate and asphaltenic crudes can cause the asphaltenes to precipitate, giving rise to high fouling factors. This may limit the amount of condensate that can be mixed with the crude blend.

Mixing of crude oils can cause the precipitation of asphaltenes and hence result in rapid fouling. In a series of papers, Wiehe^{85,86,87} (1999 a, b & c) and Wiehe and Kennedy⁸⁸ (1999) demonstrated how the incompatibility of crude oils can be predicted based on simple laboratory measurements. As defined earlier in this section, asphaltenes are substances that are insoluble in heptane but soluble in toluene. Two parameters can be defined to characterise a crude blend, namely the Insolubility Number, I_N and the Solubility Blending Number, S_{BN} . Numerical values for both these parameters are estimated if the oil under investigation is mixed with a certain amount of test liquid consisting of a heptane/toluene mixture, and the minimum amount of toluene in this mixture to avoid asphaltene precipitation is determined. The percentage of toluene in the heptane/toluene mixture which is required to inhibit asphaltene depositions varies with the ratio of the volume of oil tested to the volume of the heptane/toluene mixture with which it is tested. Results are obtained which are typified by those shown in Sketch 3.2. The oil/mixture volume ratio is expressed as a percentage with equal volumes of oil and mixture being represented as 100%.



Sketch 3.2 Evaluation of compatibility numbers for Arab Light Crude
(Wiehe and Kennedy⁸⁸, 1999, Wiehe, 1999^{85, 86, 87} (a, b and c))

As will be seen from Sketch 3.2, the data can be fitted by a straight line. The intercepts on the y and x axes are denoted as I_N and H_D respectively. I_N is defined as the **Insolubility Number** and represents the percentage of toluene in the hexane/toluene test fluid which would be required to prevent asphaltene precipitation in an infinitesimal volume of the oil being tested. The parameter H_D represents the ratio of oil to heptane volume required for precipitation to occur when the oil is contacted with an increasing volume of pure heptane. Wiehe and co-workers define the Solubility Blending Number, S_{BN} , in terms of I_N and H_D as follows:

$$S_{BN} = I_N \left(1 + \frac{100}{H_D} \right). \quad (3.3)$$

The value of S_{BN} will vary from crude to crude and a value for a crude mix, $S_{BN,mix}$, can be obtained on a volumetric average basis, as demonstrated by Wiehe⁸⁶ (1996b). Thus, for a mixture of j crude oils, the value of $S_{BN,mix}$ can be obtained from

$$S_{BN,mix} = \frac{\sum_{i=1}^j V_i S_{BN,i}}{\sum_{i=1}^j V_i} \quad (3.4)$$

where V_i is the volume of crude i in the mixture and $S_{BN,i}$ is Solubility Blending Number for this crude.

For compatibility (that is, for the avoidance of asphaltene precipitation), the Solubility Blending Number of the mixture must be higher than the largest Insolubility Number of any component in the mixture:

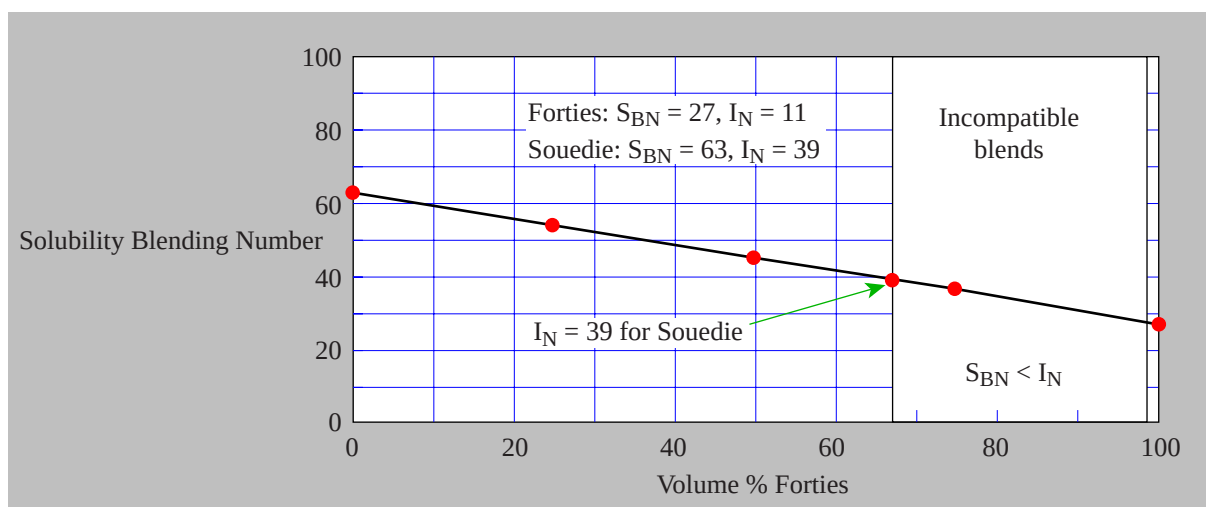
$$S_{BN, mix} > I_{N, max} \quad (3.5)$$

The application of this method is demonstrated by Wiehe⁸⁶ (1999b) for blends of Souedie and Forties crudes. The following blending and incompatibility numbers have been determined in laboratory experiments:

Forties: $S_{BN} = 27$, $I_N = 11$.

Souedie: $S_{BN} = 63$, $I_N = 39 = I_{N,max}$.

For both crudes alone, $S_{BN,i} > I_{N,i}$; that is, no asphaltene precipitation will occur. However, if the two crudes are mixed, the results outlined in Sketch 3.3 are found:



Sketch 3.3 Asphaltene solubility during blending of Souedie and Forties crudes

If Forties crude is added to a certain amount of Souedie crude, a compatible mixture is produced until the Solubility Blending Number of the mixture reaches the value of the larger of the two Insolubility Numbers, that is, 39. This occurs at a Forties crude concentration of about 67%. Further addition of Forties crude will result in asphaltene precipitation until almost pure Forties crude is reached.

Sketch 3.3 also shows the importance of the blending order. If, for example, a 50% mixture of these two crudes is produced by adding Forties Crude to Souedie crude, asphaltene precipitation will not occur. If, however, the same mixture is produced by adding Souedie crude to Forties crude, the mixing process will move through the incompatibility region and asphaltenes would be precipitated. While the precipitated asphaltene will ultimately dissolve again for Souedie crude concentrations above 33%, this dissolution process may take long enough to produce deposits in the crude distillation unit. This procedure has been verified in commercial crude pre-heat application.

3.4 Effect of Oxygen

If oxygen is present in the crude, then free radical polymer-forming reactions may occur of the type discussed in Section 2.7. Thus, oxygen has a *potentially* dramatic effect on fouling rate and has been the subject of a number of investigations.

Asomaning *et al.*² (1995) attempted to simulate the effect of oxygen on hydrocarbon fouling by recirculating a solution of indene in kerosene over a heated probe. Before placing over the probe, the mixture was saturated with a mixture of air and nitrogen at various concentrations of air. As will be seen from Sketch 3.4, there was an induction period after which the fouling factor rose very rapidly, the induction period being