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An Experimental Investigation and Prediction of Asphaltene Deposition during Laminar Flow in the Pipes Using a Heat Transfer Approach

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Abstract

In this study, asphaltene deposition from crude oil has experimentally and theoretically been studied using a test loop and an accurate temperature monitoring during a laminar flow. The effects of oil velocity and surface temperature on the thickness of asphaltene deposition were investigated. The results show that asphaltene deposition thickness increases by increasing surface temperature. As the oil velocity increased, less deposition was noticed in this experimental study. The thermal approach was used to describe the mechanisms involved in this process, and the results of data fitting showed that there was good agreement between the results of the proposed model and the measured asphaltene deposition rates. Moreover, the theoretical study of deposition process showed that the rate of asphaltene deposition was inversely related to velocity, which was proved by the experimental results.

Keywords: Asphaltene, Theoretical Model, Deposition Rates, Test Loop, Laminar Flow

1. Introduction

The formation of asphaltene deposition during petroleum production causes several operational issues such as total or partial blocking of pipeline and wellbore, changes in wettability, and damages to the equipment (Islam 1994). Generally, asphaltenes are regarded as a part of the crude oil that is insoluble in normal alkanes such as pentane and heptane but soluble in aromatics such as toluene and benzene. Moreover, asphaltenes are known for their highest molecular weight and most polar oil components (Ali, Al-Ghannam, 1981; Hirschberg et al., 1984). Asphaltenes composition, structure, and stability are dependent on its source, and the type of solvent used for the extraction of oil (Marlow et al., 1987; Groenzin, Mullins, 2001; Alboudwarej et al., 2002; Akbarzadeh et al., 2011; Akbarzadeh et al., 2007).

Asphaltene deposition is a widely identified problem that occurs during production operations in the petroleum industry and generates a large cost increase due to the necessity of prevention and removal of deposits. The precipitated and subsequently deposited asphaltenes can affect reservoir productivity

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and cause plugging in pipes and processing equipment. Asphaltenes are also known to facilitate the formation of stable water-in-oil emulsions and to contribute to the poisoning of catalysts in petroleum refining processes (Minssieux 1997; De Pedroza et al., 1996). Asphaltenes as colloids with various sizes or as individual molecules can be absorbed on solid surfaces by virtue of their phenolic and carboxylic weak acid groups (Kokal et al., 1995). The absorption and deposition of asphaltenes on a steel surface would restrict oil flow in transportation pipelines (Mochida et al., 1988; Faus et al., 1984). The remediation of asphaltene is very costly, which limits the production design of many asphaltenic crude oil reserves (Akbarzadeh et al., 2011; Akbarzadeh et al., 2009; Buckley, 2012; Hammami, Ratulowski, 2007). In many cases, the potential of organic solids depositions forces the field managers to rely mostly on chemical and mechanical remediation methods (Leontaritis, 2005; Minssieux, 1997; Peramanu et al., 1999; Chang, Scott Fogler, 1996).

Asphaltene deposition in pipelines is very complex, which depends on many parameters such as flow shear rate, surface type and characteristics, particle size and particle surface interactions. Although asphaltene precipitation is necessary, it is not sufficient for the deposition to interrupt fluid flow in the pipe because of blockage. After precipitation, asphaltene particles must move close to the surface and stick to it to be studied as a flow assurance problem in pipe lines. Therefore, a better understanding of the mechanism of solid deposition is required for proper design treatments, including the effect of pressure, temperature, composition, additives, and flow conditions. According to the literature on asphaltene deposition, few studies have been reported on asphaltene deposition in real pipe conditions (Alboudwarej, 2003; De Boer et al., 1995; Jamialahmadi et al., 2009; Wang et al., 2004; Zougari et al., 2006).

Broseta et al. (2000) and Wang et al. (2004) investigated asphaltene deposition in a capillary tube (Broseta et al., 2000; Wang et al., 2004). Pressure drop across the capillary tube was used as an indication of the extent of asphaltene deposition. The results were used to simulate the deposition of asphaltenes in porous media. Jamialahmadi et al. (2009) investigated the mechanisms of the deposition of flocculated asphaltene experimentally and theoretically under forced convective conditions and turbulence condition. The results showed that the rate of asphaltene deposition increased with increasing flocculated asphaltene concentration and temperature, while it decreased with increasing oil velocity (Jamialahmadi et al., 2009). Shirdel et al. (2012) studied the application of some particle depositions to predict the published experimental data. Based on the studies performed, the authors concluded that the deposition models satisfactorily agree with experimental data (Shirdel et al., 2012). One study using a fixed flow rate and changing ratios of heptane to two different petroleum fluids concluded that the amount of precipitating material was a strong contributor to the deposition behavior; when too little heptane was added to the petroleum fluid, very little deposition occurred; while the largest amount of heptane used, 50% by volume, the largest amount of deposition was generated (Hoepfner et al., 2013). In larger metal pipes (24 mm diameter), decreasing flow rate and increasing asphaltene content were both found to increase deposition, but the control parameters were varied only by a factor of two (Soltani Soulgani et al., 2009). Furthermore, there is disagreement regarding the uniformity of deposition along the axial direction in a pipe. Evidence has been presented to suggest both that deposition is uniform throughout a given length of pipe and that deposition occurs mainly near the pipe inlet (Hoepfner et al., 2013; Wang et al., 2004). “Washing technique” for quantitatively measuring the rate of asphaltene deposition during a laminar flow was used by Salimi et al. (2014) in a steel pipe. The results showed that thermal approach based on fouling method was applied to the experimental procedure, which showed good agreement between the predicted thickness and the measured value from the test loop (Salimi et al., 2013).

A review of the existing literature on mechanisms and kinetics of asphaltene deposition from crude oil reveals that hardly any information is available on this subject. This is mainly because of the difficulty of using conventional approaches toward measuring the mass of asphaltene deposition per unit area as a function of time and determining the controlling mechanism and activation energy of the deposition process. The present research work is a mechanistic study of asphaltene deposition in a laminar flow to investigate the effects of oil velocity and temperature on the rate of asphaltene deposition using a thermal approach.

2. Thermal approach to deposition measurement

The total resistance to heat transfer from the flowing fluid in the pipes to the environment is consisted of convective heat transfer from the flowing fluid to the pipe wall, heat conduction through the pipe wall, including any insulation or deposited layer, and an appropriate heat-transfer resistance to the environment (e.g., water, air, or any other cooling fluid). If a layer of asphaltene deposit is formed in the pipe wall, a thermal resistance term is added to the total resistance for the heat transfer from the flowing fluid to the environment due to heat conduction through the asphaltene layer. Hence, the thickness can be determined from the measurements of relevant thermal parameters by solving the heat balance equation. Heat transfer from the internal flowing fluid to the outside environment is described by Chen et al. 1997 (Chen et al., 1997).

$$\frac{1}{U_b} = \frac{1}{h_o}\frac{r_o}{r_i - \delta_d} + \frac{r_o}{k_{dep}}\ln\frac{r_i}{r_i - \delta_d} + \frac{r_o}{k_p}\ln\frac{r_o}{r_i} + \frac{1}{h_g} \tag{1}$$

The total heat-transfer rate in the test section is given by:

$$Q = U_b A_b \Delta T_{lm} = m_o C_{po}(T_{o,in} - T_{o,out}) \tag{2}$$

where, $A_b = \pi d_0 L$, and ΔT_{lm} is the logarithmic mean temperature difference in the heat-exchange region. At the beginning of the experiment, asphaltene thickness is zero and the heat transfer equation is defined by:

$$\frac{1}{U_{bo}} = \frac{1}{h_o}\frac{r_o}{r_i} + \frac{r_o}{k_p}\ln\frac{r_o}{r_i} + \frac{1}{h_g} \tag{3}$$

If Equation 3 is subtracted from Equation 1, the following equation is obtained:

$$Y = \frac{1}{U_b} - \frac{1}{U_{b0}} = \frac{1}{h_o}\frac{r_o\delta_d}{r_i(r_i - \delta_d)} + \frac{r_o}{k_{dep}}\ln\frac{r_i}{r_i - \delta_d} \tag{4}$$

The overall heat-transfer coefficient, U_b, is calculated by Equation 2. Considering the regime for heat transfer, the inside convective heat-transfer coefficient for oil (h_o) is obtained using the Hausen correlation (Bergman and Incropera, 2011):

$$N_{Nu} = 3.66 + \frac{0.19[\mathrm{N}_{Pe}(\frac{d_w}{L})]^{0.8}}{1 + 0.117[\mathrm{N}_{Pe}(\frac{d_w}{L})]^{0.467}} \tag{5}$$

where, $N_{Pe}=N_{Re}N_{Pr}$, and $N_{Pr}=\mu Cp/k$; d_p is the diameter of the pipe open to flow. Finally, h_o is obtained using the below expression:

$$h_o = \frac{N_{Nu} k_W}{d_W} \tag{6}$$

3. Experimental procedure

A test loop is used to investigate asphaltene deposition inside the pipe using an Iranian asphaltenic crude oil. The fractions of saturates, aromatics, resins, and asphaltenes in the oil were obtained from the so-called SARA test depicted in Table 1. The physical properties of oil under various operating temperature are essential for the calculation of heat transfer coefficients and the mass of the asphaltene deposited on the surface. Figures 1 and 2 present the viscosity, density (using SVM 3000 viscometer), specific heat (Cragoe, 1929) and thermal conductivity (Stachowiak and Batchelor, 2013) of oil at different temperatures. As expected, the viscosity and thermal conductivity of the oil decrease as temperature is increased. Conversely, the specific heat increases with oil temperature, while the density remains almost constant.

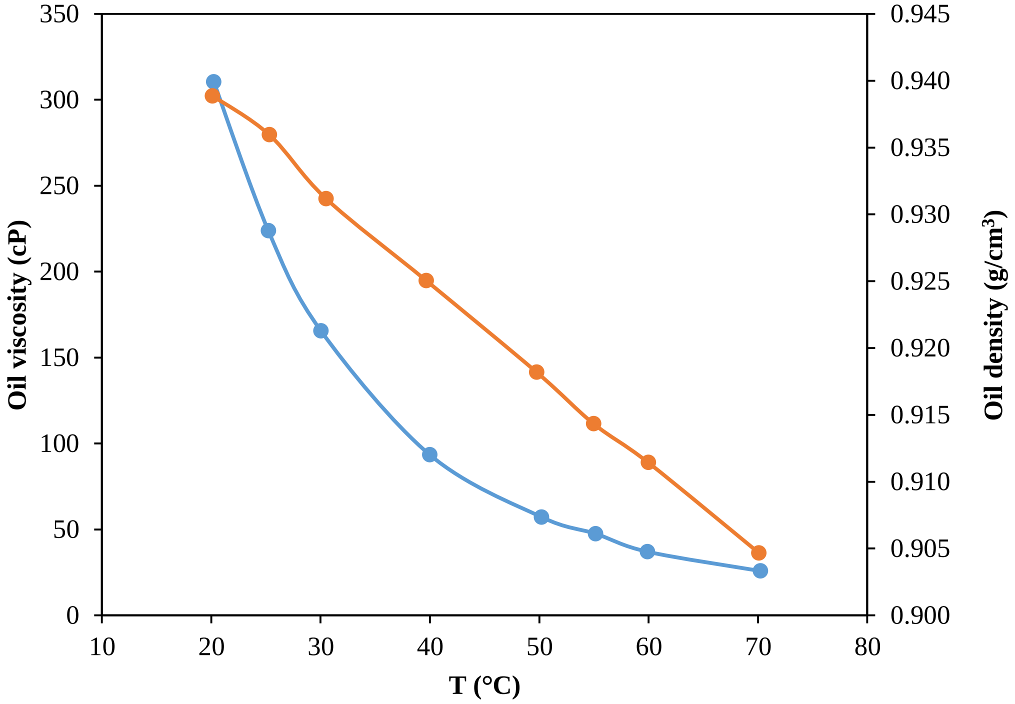

Figure 1
Variation of the oil viscosity and density against temperature.

One of the methods used to measure the amount of precipitated asphaltenes due to the solvent is the scaling method. Mathematical correlations of this method is very simple and do not need the oil specification. This method was first proposed by Rassamdana et al. (Rassamdana et al., 1996). The mass of precipitated asphaltene as a function of the volumetric dilution ratio (n-heptane/oil) for several temperatures measured in this work is shown in Figure 3. In this research, the concentration of flocculated asphaltene is 4.88 kg/m^3. The range of the experimental parameters and the proper range of model performance covered in this investigation are shown in Table 2.

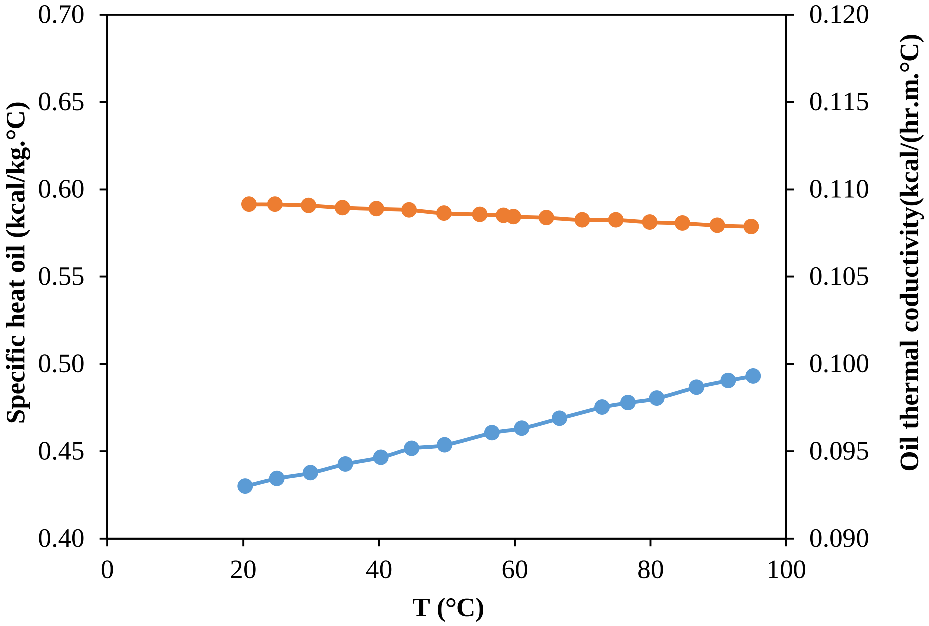

Figure 2
Variation of specific heat and thermal conductivity of the oil against temperature.

Table 1
Analysis of SARA.

Fractions	wt.%
Asphaltenes	13.84
Resins	13.46
Saturates	30.30
Aromatics	42.40

Table 2
Range of operating parameters.

Flow velocity	0.18-0.67 m/s
Surface temperature	50 °C
Asphaltene supersaturation concentration	4.88-6.2 kg/m^3
Bulk temperature	70-90 °C
Reynolds number	300-1100

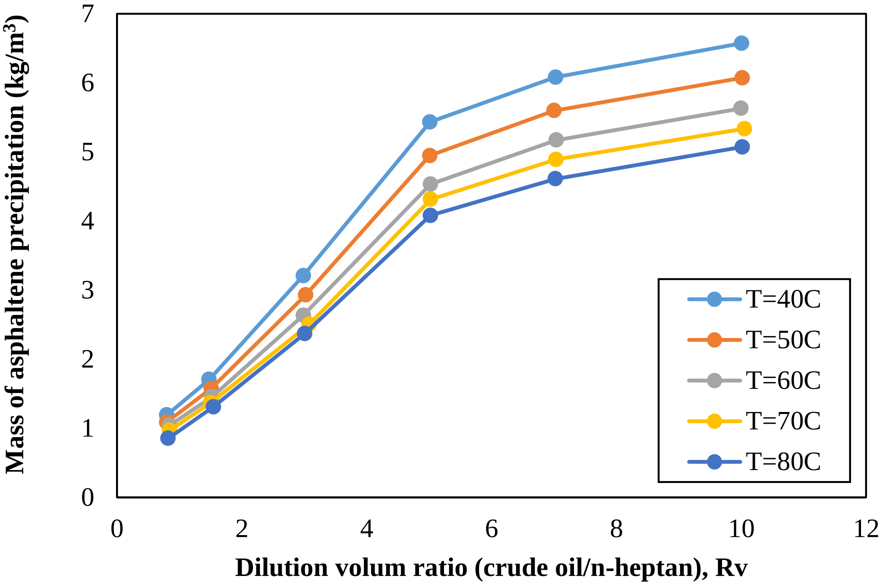

Figure 3
Variation of mass of asphaltene precipitation against dilution ratio at various temperatures.

3.1. Experimental apparatus

Figure 4 shows the schematic view of the deposition test apparatus which was used to measure the thickness of asphaltene deposition as a function of time in different conditions. The apparatus was made of a well-controlled temperature bath containing a long stainless steel tube in the shape of a coil. The temperature of the bath was maintained constant using a stirrer, and the long tube was equipped with accurate pressure transducers and thermocouples at several intervals, transferring all the information into a data acquisition system. The feed was prepared and transferred into feed storage and its temperature was maintained at a pre-set temperature prior to flow through the pump into the flowing loop.

The test section was made of a stainless steel tube with a length of 1 m and an inside diameter of 3.74 mm (seamless, Fitok Company) which was coiled and placed inside the bath. The bulk temperature of the oil was measured with K-type thermocouples which were located in the tank and mixing chambers before and after the test section. The temperature of the tube was controlled within ±0.1 °C by immersing it in the fluid bath. The absolute pressure at the outlet of the tube was controlled with a back pressure control regulator (model BP-66). The oil flow rate was controlled by a constant rate pump. A data acquisition system was used to monitor temperature differences in the system to be used for the thermal calculation procedure.

3.2. Error analysis

The experimental error for the measured mass of asphaltene deposition may be due to errors in the measurement of bulk temperature and bath temperature of the test section. The temperatures were measured with PT100 thermocouples located in the bulk of the oil flow before and after the test section. The inaccuracy in temperature measurements due to the calibration errors of the thermocouples may lead to a deviation of approximately ±0.1 K. The maximum error in the mass of asphaltene deposition measurements was estimated to be less than ±2.5% by taking into account the temperature and flow rate measurement errors.

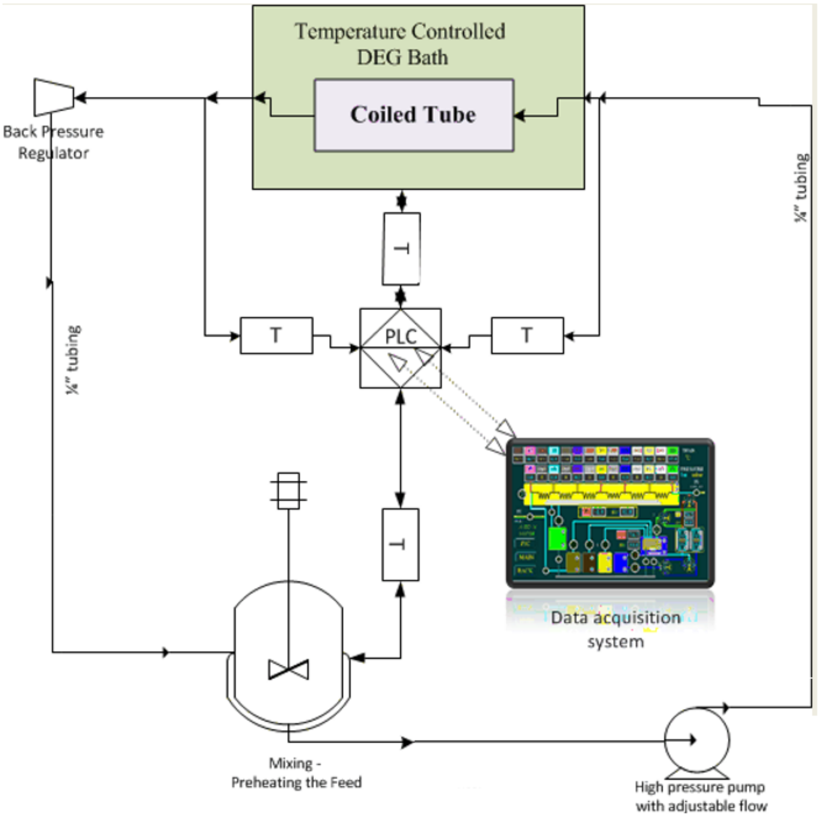

Figure 4
A schematic of the experimental apparatus.

4. Results and discussion

4.1. Effect of wall temperature

To study the effect of wall temperature on asphaltene deposition, three different tests at different wall temperatures were performed at a constant oil velocity, oil temperature, and asphaltene concentration. The thickness of asphaltene deposition is plotted at different times in Figures 5 which shows that the deposition thickness increases at higher wall temperatures. Figure 6 illustrates the linear correlations between the rate of asphaltene deposition and surface temperature. These results indicate that the rate of deposition of asphaltene particles on the wall surface depends strongly on the wall temperature.

If the logarithm of the deposition rate is plotted versus the inverse of wall temperature (Figure 7), a linear correlation is found. This behavior is similar to the fouling phenomenon happens in the heat exchanges pipes, which shows the exponential increase of the fouling thickness by increasing the surface temperature (Crittenden et al., 1992; Eaton and Lux, 1984; Saleh et al., 2005; Srinivasan and Watkinson, 2005).

The solubility of asphaltene plays an important role in crude oil deposition. Generally, the solubility of asphaltene in crude oil increases with an increase in temperature (Figure 3). A complex relationship between asphaltene solubility and temperature has been reported by Lambourn and Durrieu (1983), in which the solubility of asphaltene increased with temperature (Lambourn and Durrieu, 1983). At high bulk temperatures, the asphaltene is in the form of solution in crude oil, and the deposition (fouling) rate is low, whereas at low bulk temperatures, asphaltene precipitates out from crude oil, and the deposition rate is high. On the other hand, the main effect of surface temperature on the rate of asphaltene deposition on the surface is through the diffusivity of asphaltene particles and the attachment rate constant of the process. Moreover, according to Figure 3, changes in the amount of precipitated asphaltene in oil decrease very low with increasing temperature.

Thus, comparing these two phenomena indicates that the effect of surface temperature on the formation of deposition is more than reducing precipitation with temperature (Salimi et al., 2013; Jamialahmadi et al., 2009; Salimi et al., 2016).

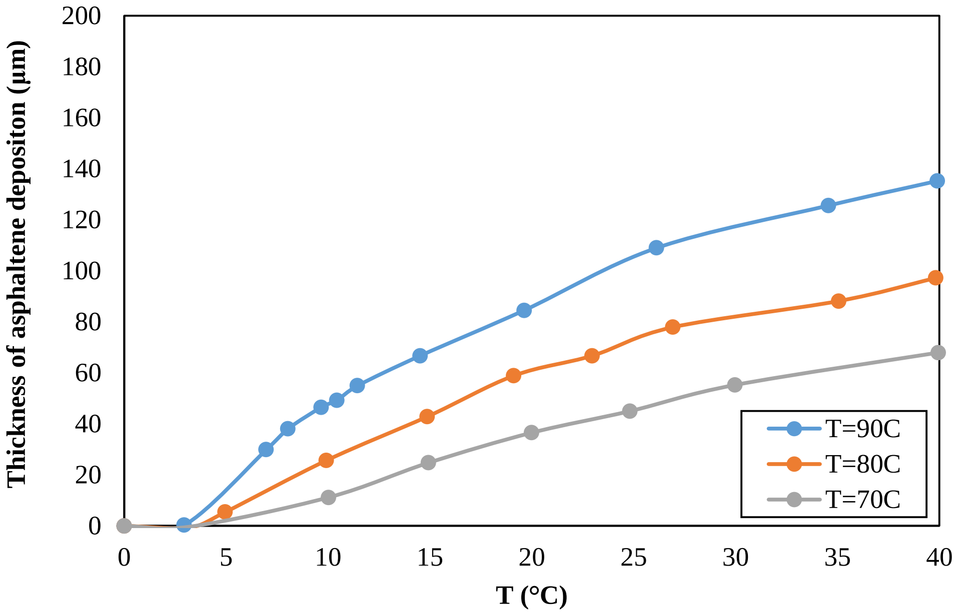

Figure 5
Thickness of asphaltene deposit as a function of time, showing the effect of the bath temperature.

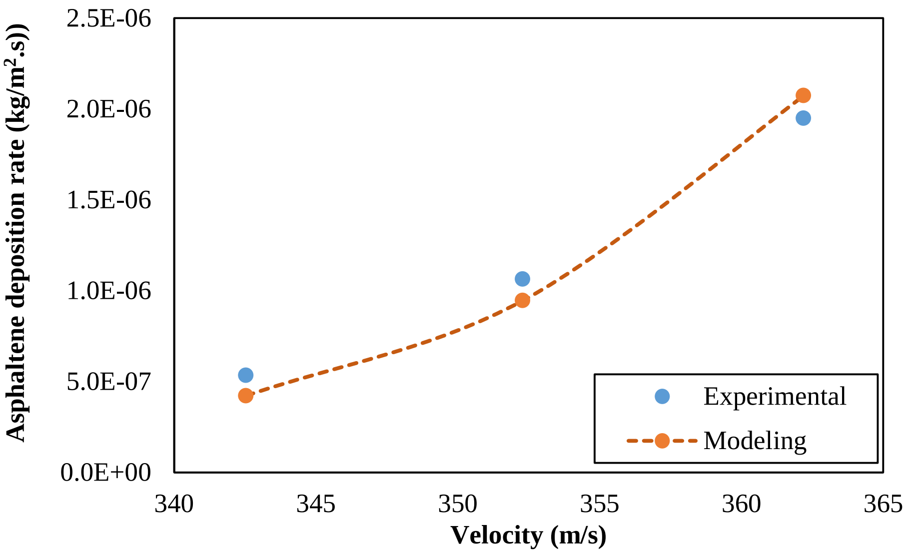

Figure 6
Variation of asphaltene deposition rate against wall temperature.

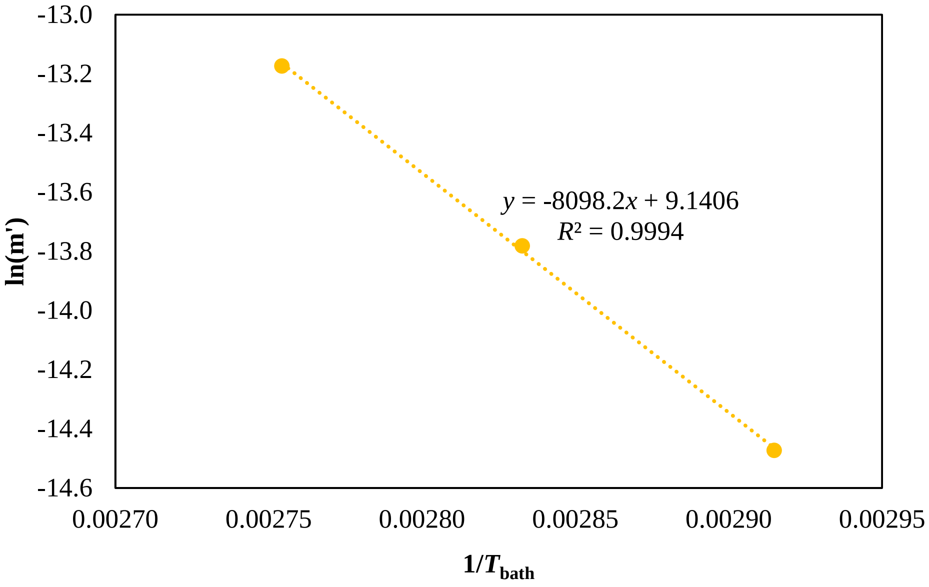

Figure 7
Logarithm of asphaltene deposition rate versus the inverse of wall temperature.

4.2. Effect of oil velocity

In this section, the effect of velocity, in the range of 0.23 to 0.67 m/s, on the deposition thickness is presented. Figure 8 shows the rate of deposited asphaltene on the surface for different oil velocities measured using the heat transfer approach. The deposition rates obtained from the slope of these curves is plotted as a function of oil velocity. This clearly indicates that the deposition rate decreases significantly when the oil velocity increases. Figure 9 shows the deposition rate ($kg/m^2.s$) at various velocities; it clearly presents a linear trend. This correlation could be used for this oil to find the critical fluid velocity for less deposition on the pipe wall.

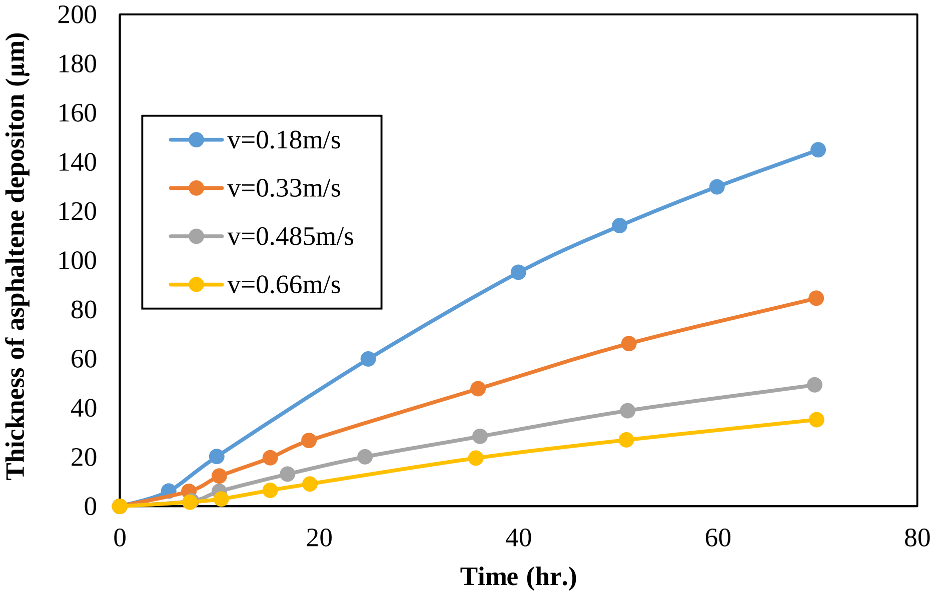

Figure 8
Thickness of asphaltene deposit as a function of time, showing the effect of oil velocity (T_{bath}=70 °C).

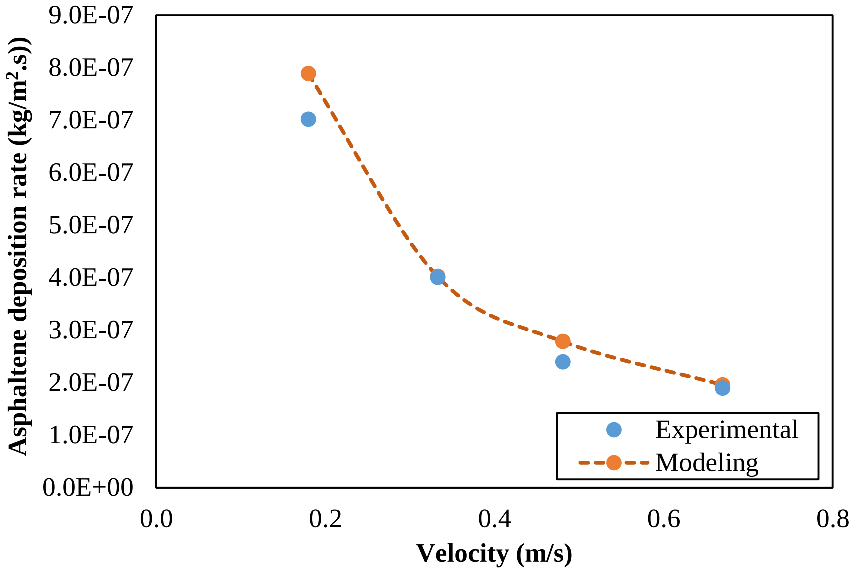

Figure 9

Variation of asphaltene deposition rate against velocity (T_{bath}=70 °C).

It is shown that the deposition rate strongly depends on the flow velocity, which has direct effects on both deposition and removal rates through the hydrodynamic effects such as eddies and shear stress at the surface. Furthermore, an increase in velocity increases the heat transfer coefficient and thus reduces the wall and film temperatures.

On the other hand, if the deposition rate is controlled by the mass transfer of the asphaltene flocculation from the bulk fluid to the surface, the velocity increases the mass transfer coefficient from the bulk fluid to the surface, and deposition rate increases too.

4.3. The model

The mass transfer coefficient for a laminar flow can be calculated from the Sieder–Tate correlation (Bergman and Incropera, 2011) and the Chilton and Colburn analogy by replacing the Nusselt and Prandtl numbers by the Sherwood and Schmidt numbers (Equation 8):

$$Sh = 1.86\left(Re.Sc.D/L\right)^{1/3}\left(\mu/\mu_w\right)^{0.14} \tag{8}$$

After some mathematical manipulations, Equation 9 is simplified to:

$$\beta = 12\left(v^{1/3}.D_{Diff}{}^{2/3}\right).\left(\mu/\mu_w\right)^{0.14} \tag{9}$$

For submicron particles, the Brownian diffusion coefficient can be determined from the Stokes–Einstein equation (Bott, 1995):

$$D_{Diff} = \frac{k_B T}{3\pi\mu d_p} \tag{10}$$

In Equation 10, viscosity difference in bulk and near the wall is low, so the ratio $(\mu/\mu_w)^{0.14}$ is almost constant and negligible. Substituting Equation 10 into Equation 9 results in:

$$\beta \propto v^{1/3} d_p^{-2/3} \tag{11}$$

For a constant average particle size, Equation 11 is simplified to:

$$\beta \propto v^{1/3} \tag{12}$$

The experimental results show that, for this case, the deposition rate decreases at higher velocities. It is known that the shear stress on the wall is directly proportional to the fluid velocity, which increases the erosion of the deposited layer.

The deposition of the asphaltene particles to the surface takes places after the asphaltene particles from the bulk of the oil move towards the surface. By replacing the mass transfer coefficient, *β*, and the term of the probability in overall mass transfer formulation, Equation 13 will be obtained (Jamialahmadi et al., 2009):

$$\dot{m} = k_t C_{Ab} = S_p \beta C_{Ab} \tag{13}$$

where, k_t is a transport coefficient. If it is assumed that all asphaltene particles arrived at the tube surface adhere to here, one may consider C_{AS}=0. Asphaltene particles adhere to the surface if the physicochemical adhesive forces between the asphaltene particles and the surface overcome the drag forces exerted near the surface (Watkinson, 1968):

$$S_p = S_o \frac{F_{adh}}{F_{drag}} \tag{14}$$

where, S_o is a constant. The adhesion forces, F_{adh}, generally obey an Arrhenius-type trend which depends on the wall temperature. The drag force, F_{drag}, is a function of drag coefficient, cross-section of the asphaltene particles in the flow direction, oil density, and oil velocity:

$$F_{drag} = C_p A_p \rho v^2 \tag{15}$$

Therefore, a number of approximations of C_D have been suggested in the literature (Hoey et al., 2012) as a function of *Re* Number. For *Re* Numbers smaller than 1000, C_D is given as follows (Allen and Raabe, 1982):

$$C_D = 30\ Re^{-0.625} \tag{16}$$

Substituting Equation 16 into Equation 15 results in:

$$F_{drag} = 30\,\mathrm{Re}^{-0.625} A_p \rho v^2 \tag{17}$$

After some mathematical manipulations, Equation 17 is simplified to:

$$F_{drag} \propto v^{1.375} \tag{18}$$

The adhesion forces generally obey an Arrhenius-type expression and may be written as:

$$F_{adh} = F_a \exp\left(\frac{E_a}{RT}\right) \tag{19}$$

After substituting Equations 18 and 19 in Equation 14 and some mathematical manipulations, Equation 20 is obtained:

$$S_p \propto \frac{\exp\left(\frac{E_a}{RT}\right)}{v^{1.375}} \tag{20}$$

and substituting Equations 20 and 12 in Equations 13, and 21, and assuming asphaltene concentration constant, one may obtain:

$$\dot{m} \propto \frac{\exp\left(\frac{E_a}{RT}\right)}{v^{1.04}} \tag{21}$$

In this equation the deposition rate is proportional to the inverse of velocity, and an increase in velocity results in a reduction in the deposition rate. Good agreement is seen between the experimental results and Equation 20, as shown in Figure 10.

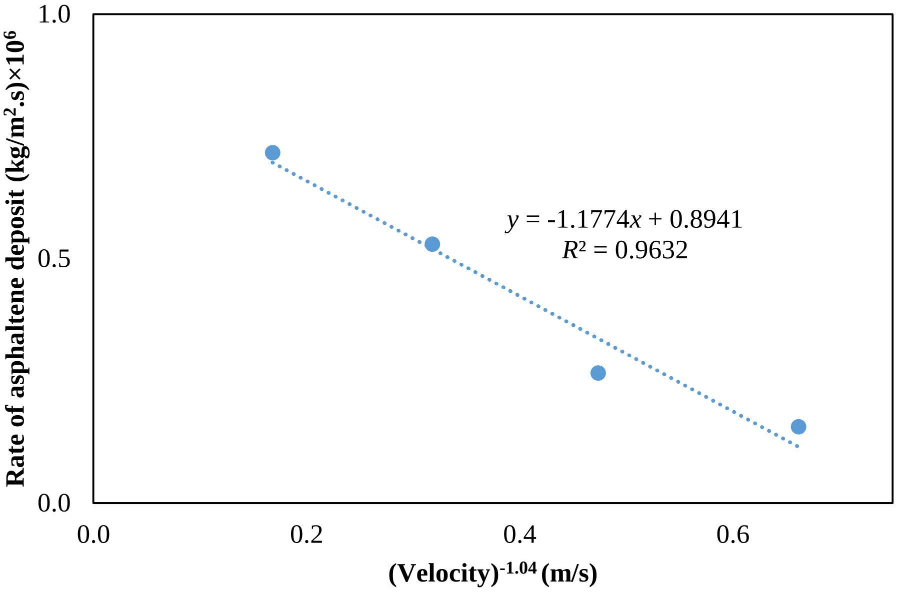

Figure 10
Asphaltene deposition rate at different velocities compared with Equation 20. (T_{bath}=70 °C).

Finally, Equation 22, by considering asphaltene concentration, will be obtained:

$$\dot{m} = k_d \frac{\exp\left(\frac{E_a}{RT}\right)}{v^{1.04}} (C_{Ab})^{1.2} \tag{22}$$

The results of our previous work indicated that concentration power, n, is equal to 1.2 (Salimi et al., 2013). Equation 22 shows that the rate of the attachment of asphaltene particles flocculated in oil to the surface is proportional to the concentration and inversely proportional to the oil velocity, which is in good agreement with the present experimental observations. Here, by the curve-fitting of the experimental data, E_a/R and k_d were respectively found equal to 10285 K and $2.1\times10^5 \frac{m^{2.04}}{s^{2.04}\left(\frac{kg}{m^3}\right)^{0.2}}$.

5. Conclusions

In the present study, after carefully applying the thermal approach for the measurements of asphaltene deposition thickness, the effects of operating parameters such as oil velocity and pipe surface temperature were investigated on the deposition process in an asphaltenic crude oil flow in a tube. The results show that the deposition rate is increased as the surface temperature elevated. The experimental results also indicate that the deposition rate is inversely proportional to the oil velocity, and the thickness of the deposited components decreases as the oil velocity increases. The experimental results have been used to develop a mechanistic model for the prediction of the rate of asphaltene deposition. Considering the complexity of the deposition process, the quantitative agreement between the measured and the predicted values is good. Finally, the proposed model is applicable to modeling the asphaltene deposition of crude oil in pipelines.

Nomenclature

C_p : Specific heat [kcal/(kg.°C)]
ΔT_{ln} : The logarithmic mean temperature difference [°C]
μ : Viscosity [Pa.s]
μ_w : Viscosity of the fluid near the wall [Pa.s]
A_i : Cross-sectional area [m^2]
C : Concentration [kg/m^3]
C_D : Drag coefficient
D : Dynamic diameter[m]
d_p : Particle diameter[m]
E : Attachment activation energy [J/mole]
f : Darcy friction factor
F_{adh} : Adhesion forces [N]
F_{drag} : Drag force [N]
h_g : Convection coefficient of the bath [kcal/(hr.m^2. °C)]
h_o : Convection coefficient of oil [kcal/(hr.m^2. °C)]
k_B : Boltzman constant [1.38×10^{-23}J/K]
k_p : Pipe's thermal conductivity [kcal/(hr.m. °C)]
k_t : Transport coefficient [m/s]
k_w : Deposit's thermal conductivity [kcal/(hr.m. °C)]
L : Pipe length [m]
m : Mass of deposit per unit area[kg/m^2]
$\dot{m}$: Rate of deposition [kg/m^2.s]
N_{Pr} : Prandtl number
Q : The total heat-transfer rate [kcal]
r : Radius [m]
Re : Reynolds number
Sc : Schmidt number

Sh : Sherwood number
S_p : Sticking probability
T : Temperature [°C]
U_b : The overall heat-transfer coefficient [kcal/(hr.m^2. °C)]
v : Velocity [m/s]
β : Mass transfer coefficient [m^2/s]
δ : Thickness of deposit[m]
ρ : Density [kg/m^3]

Subscripts–superscripts

a : asphaltene
b : bulk
d : deposit
w : wall
r : removal
o : oil or outside

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