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Experimental Investigation of the Effects of Different Parameters on the Rate of Asphaltene Deposition in Laminar Flow and Its Prediction Using Heat Transfer Approach

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1. INTRODUCTION

Flow assurance is referred to a technique to assess the ability of production facilities to transfer multi-phase fluids from the reservoir to the market. The fluid behavior is tested for any possible interruption during this fluids flow process. Flow assurance includes factors such as asphaltene and wax deposition, hydrate formation, scale, slugging, and corrosion.[1] Asphaltene deposition inside the oil reservoirs and production facilities is known as the main flow assurance problem in the oil industry.

In this study, asphaltene deposition from crude oil on the pipe surface has been studied experimentally using a novel designed test loop. Washing technique is used to quantitatively measure the rate of asphaltene deposition during laminar flow in the steel pipe. The effects of oil velocity, asphaltene content, and surface temperature on the thickness of asphaltene deposition are investigated. The results show that the asphaltene deposition rate increases with increasing surface temperature, results in asphaltene content reduction of the flowing crude oil. As the oil velocity increases, less deposition was noticed on the surface of the pipe. Besides, thermal approach was applied to the experimental procedure which shows good agreements between the predicted thickness and the measured value from the test loop.

Keywords Asphaltene, deposition, laminar flow, pipeline, washing method

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asphaltenes on steel surface would restrict oil flow in the transportation pipelines. The remediation of asphaltene is very costly, which limits the production design of many asphaltic crude oil reserves.

In many cases, the potential of organic solids deposition forces the field managers to rely mostly on the chemical and mechanical remediation methods. Therefore, better understanding of the mechanism of solid deposition is required to better design treatments, including the effect of pressure, temperature, composition, additives, and flow conditions. These are the most important parameters during flow assurance process.

Literature study on asphaltene deposition reveals that few works on asphaltene deposition at real pipe conditions have been reported. The aim of this work is a mechanistic study of asphaltene deposition in laminar flow to investigate the effects of oil velocity, temperature and asphaltene content on the rate of asphaltene deposition.

2. EXPERIMENTAL

2.1. Experimental Procedure

A test loop is used to investigate asphaltene deposition inside the pipe using an Iranian asphaltic crude oil. The fractions of saturates, aromatics, resins and asphaltene in the oil were obtained from the so-called SARA test depicted in Table 1.

In this work, the deposited asphaltene inside of tube was measured by “washing method.” For this purpose the tube was rinsed with heptane first to wash out non-asphaltene hydrocarbons, where asphaltenic components remained there attached to the wall. In the next step the deposited materials inside the tube were washed out using toluene and its weight was measured after evaporating of the toluene.

The following procedure was used to measure the asphaltene fraction of deposited materials inside the pipe. The collected asphaltenes from the previous step, “washing method,” was first precipitated with n-heptane at 40:1 volume ratio of n-heptane to sample solution. Then the mixture was left to equilibrate for 20–24 hours, and finally the sample was filtered by filter paper (Whatman Grade No. 42). The Filter with asphaltene was crumpled and placed in a Soxhlet apparatus and refluxed with n-heptane for 2 hours. The deposited material retained on the filter paper was considered as asphaltene after drying process.

2.2. Experimental Apparatus

Figure 1 shows schematic view of the novel designed flow assurance test loop which has been used to measure the thickness of asphaltene deposition as a function of time at different condition. The apparatus is made of a well-controlled temperature bath containing long stainless steel tube in coil shape. The temperature of the bath was maintained constant using heat source, controlling unit and stirrer. The long test tube was equipped with accurate pressure transducers and thermocouples at several intervals; transferring all the information into a data acquisition system.
system. The feed was prepared and transferred into feed storage and its temperature was maintained at pre-set temperature prior to flow through the pump into the flowing loop.

For this study, the pipe test section was made of 1 m length stainless steel tube (seamless, Fitok Company, China) with 3.74 mm inside diameter, which was coiled and placed inside the bath. The bulk temperature of the oil is measured with K-type thermocouples which are located in the tank and in mixing chambers before and after of the test section. The temperature of the bath was controlled within ±0.1°C, holding the stainless steel pipe. The absolute pressure at the outlet of the tube was controlled with back pressure control regulator (model of BP-66). The oil flow rate was controlled by the constant rate pump. A data acquisition system is used to monitor the temperature at various point of both bath and tube.

3. RESULTS AND DISCUSSION

The results are provided in Table 2. These eight set of tests were designed to check the effects of oil velocity and surface temperature on the rate of asphaltene deposition. This procedure (washing method) could be used to find the deposition rate by carefully monitoring the temperature difference between the outlet and inlet and also bath temperature. The results for the asphaltene deposition rates are presented and discussed in the following sections. Also the analytical tests of the deposited materials show that the amount of asphaltene in the deposited layer is significant.

### 3.1. Concentration Measurement of Flocculated Asphaltene

The first step to study the mechanisms of asphaltene deposition is to determine the concentration of flocculated asphaltene particles in the oil at specified temperatures. One of the methods used to measure the amount of precipitated asphaltenes due to the solvent is 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. Their results showed that all asphaltene titration curves of the dead oil by solvents of normal alkane correlate into a single curve. This function is as follows:

\[ Y = A_0 + A_1X + A_2X^2 + A_3X^3. \]  \[ \text{[1]} \]

Parameters \( A_0 \) to \( A_3 \) are constant and indicate the scaling coefficients. The three main variables of titration of dead oil curves are the weight percentage of precipitated asphaltene \( W \), the solvent to oil dilution ratio \( R_m \) and the molecular weight of the solvent. Rassamdana et al. lumped these three parameters into two variables \( X \) and \( Y \) of the scaling equation:

\[ X = \frac{R_m}{M^2} \quad \text{and} \quad Y = \frac{W}{M^2}. \]  \[ \text{[2]} \]

Adjustable parameters are \( z \) and \( z' \) which must be carefully tuned to find the best fitting of the experimental data. They suggested \( Z' = -2 \) and \( Z = 0.25 \) in spite of oil and type of precipitant material used. In recent years, several investigators have verified the scaling model. Ashoori et al. modified the scaling equations and considered the scaling variables \( X \) and \( Y \) as follows:

\[ X = \frac{R_V}{(T^* \cdot M_w^Z)} \quad \text{and} \quad Y = \frac{W_t}{R_V^{Z'}}. \]  \[ \text{[3]} \]

The exponent \( n \) is a constant and its value is between 0.10 and 0.25. Two other constants, \( Z \) and \( Z' \), are the same as the first scaling equation, that is, \( Z = 0.25 \) and \( Z' = -2 \).

The measured mass of precipitated asphaltene as a function of the volumetric dilution ratio (n-heptane/oil) for several temperatures in this work has been shown in Figure 2. The mass of asphaltene deposition versus dilution

<table>
<thead>
<tr>
<th>Test number</th>
<th>( C_{\text{asph}} ) (gr/cm³)</th>
<th>( T_{\text{oil}} ) (°C)</th>
<th>( T_{\text{bath}} ) (°C)</th>
<th>Velocity (m/s)</th>
<th>Time (hr)</th>
<th>Mass of deposit (kg/m²) (Washing method)</th>
<th>% Asphaltene</th>
</tr>
</thead>
<tbody>
<tr>
<td>Test 1</td>
<td>1.5</td>
<td>51.5</td>
<td>70</td>
<td>0.23</td>
<td>70</td>
<td>0.4</td>
<td>63.3</td>
</tr>
<tr>
<td>Test 2</td>
<td>1.5</td>
<td>52.5</td>
<td>70</td>
<td>0.51</td>
<td>70</td>
<td>0.22</td>
<td>82</td>
</tr>
<tr>
<td>Test 3</td>
<td>1.5</td>
<td>50</td>
<td>70</td>
<td>0.67</td>
<td>70</td>
<td>0.118</td>
<td>87</td>
</tr>
<tr>
<td>Test 4</td>
<td>3</td>
<td>50</td>
<td>70</td>
<td>0.27</td>
<td>52</td>
<td>0.144</td>
<td>83.3</td>
</tr>
<tr>
<td>Test 5</td>
<td>5</td>
<td>50</td>
<td>70</td>
<td>0.17</td>
<td>47</td>
<td>0.208</td>
<td>85</td>
</tr>
<tr>
<td>Test 6</td>
<td>5</td>
<td>57</td>
<td>70</td>
<td>0.333</td>
<td>40</td>
<td>0.094</td>
<td>85</td>
</tr>
<tr>
<td>Test 7</td>
<td>1.5</td>
<td>57</td>
<td>80</td>
<td>0.333</td>
<td>24</td>
<td>0.103</td>
<td>84</td>
</tr>
<tr>
<td>Test 8</td>
<td>1.5</td>
<td>57</td>
<td>90</td>
<td>0.333</td>
<td>23</td>
<td>0.21</td>
<td>87</td>
</tr>
</tbody>
</table>

\( a \) Average oil temperature of the input and output in the pipe.
volume ratio found here is based on work of Ashoori et al. [26]

3.2. Mechanism of Asphaltene Deposition

The published works show that no comprehensive model to describe the effect of operating conditions on the mechanism of asphaltene deposition in the pipes is available. [27] Broseta et al. [28] and Wang et al. [19] investigated asphaltene deposition in a capillary tube. Jamialahmadi et al. [21] investigated the mechanisms of deposition of flocculated asphaltene under forced convective conditions and turbulence condition. Results showed that the rate of asphaltene deposition increases with increasing flocculated asphaltene concentration and temperature while it decreases with increasing oil velocity.

There are likely several steps in deposition process for asphaltenes: precipitation, flocculation, surface contact and adhesion. [18] Asphaltenes could be directly absorbed on the solid surfaces if no precipitation has been occurred however, it is expected that for this case the amount of absorption is negligible. [29] To predict asphaltene deposition accurately one needs to understand the fundamental mechanism for each step. Fouling in the heat transfer cases can be used to make an analogy for the asphaltene deposition/release mechanism that was presented by Kern and Seaton. [30] The net rate of growth of asphaltene deposit is the difference between the rate of deposited of material $m_d$ and the rate of its removal $m_r$ expressed as follows:

$$\frac{dm}{dt} = m_d - m_r.$$  \hspace{1cm} [4]

3.3. Effect of Asphaltene Concentration

One of the main reasons of asphaltene deposition is the concentration of flocculated asphaltene in the flowing oil. So long as the removal rate can be ignored and all particles arriving at the heat transfer surface are deposited, the rate of deposition may be generally expressed as:

$$m_d = k(C_{AS})^n,$$  \hspace{1cm} [5]

where $C_{AS}$ is the flocculated asphaltene concentration at the surface conditions. For mass transfer and surface deposition controlled processes $n$ generally varies between 1 and 2. Equation (5) shows that the concentration has a strong effect on deposition, regardless of the mechanism of deposition. The effect of asphaltene concentration on rate of deposition at a Reynolds number about 1000 and constant bulk and bath temperature is shown in Figure 3. The results indicate that the increase of asphaltene concentration lead to enhance the rate of asphaltene deposition. The results indicate that there are a linear relation between rate of asphaltene deposition and asphaltene concentration and the best $n$ for fitting $m_d$ versus concentration of asphaltene is approximately equal to 1.2.

3.4. Effect of Wall Temperature

To study the effect of wall temperature on asphaltene deposition, three different tests at different wall temperatures were performed at constant oil velocity, bulk temperature, and asphaltene concentration. The rate of asphaltene deposition is plotted at different wall temperature in Figure 4, where shows that the deposition thickness increases at higher wall temperature.

3.5. Effect of Oil Velocity

In this section, the effect of velocity on the deposition thickness in the range of 0.23 to 0.67 m/s is discussed. Figure 5 shows the rate of asphaltene deposition for different oil velocities which were measured using the “washing method.” This clearly indicates that the deposition rate is decreased significantly when the oil velocity is increased.
FIG. 3. Variation of asphaltene deposition rate with flocculated asphaltene concentration. (Re = 1000, T_{bath} = 70°C, T_{bulk} = 50°C).

FIG. 4. Variation of asphaltene deposition rate with wall temperature (v = 0.33 m/s, T_{bath} = 57°C).

FIG. 5. Variation of asphaltene deposition rate with velocity (T_{bath} = 70°C, T_{bulk} = 50°C).
4. THE MODEL

Fouling method was used to predict the rate of asphaltene deposition on pipe surface, and an equation was developed. The models describing fouling usually are based on the well-known concept of Kern and Seaton’s\(^\text{[30]}\) approach, where the net fouling rate is the difference between the rates of deposition and removal: Fouling

\[ \text{Rate} = \text{Rate of deposition} - \text{Rate of removed}. \]

According to Equation (5), it can be put the fouling model in \(K_t\) coefficient (overall transfer coefficient) and used it for predicting of deposition. So it can be done as follows:

\[ K_t = \text{Rate of deposition} - \text{Rate of removed}. \]

The type of the description of the deposition and removal terms is the basic differences between various models reported in literature. The rate of deposition is described by either a transport-reaction model or reaction alone model while the rate of removal is described either by shear-related or mass-transfer related expressions. The first term of the right-hand side of this expression is dependent on both the surface temperature and also rate of transport the particles from the fluid bulk toward the wall.

The rate of fouling increases exponentially with increasing surface temperature for almost all fouling mechanisms,\(^\text{[31–34]}\) which is generally expressed by an Arrhenius-type equation.

However, the rate of particle transport from bulk toward wall depends on the type of flow regime. In this work, regime of flow is laminar and the Sieder–Tate correlation\(^\text{[35]}\) is used. Rate of deposition is obtained by substituting of terms includes wall temperature and rate of particle transport as follows:

\[ \text{Rate of deposition} \propto (Re.Pr(D/L))^{1/3} \]
\[ (\mu/\mu_w)^{0.14} \exp(E_a/RT_w). \]

Both length and diameter are constant and do not change during experiments, so the equation can be simplified as follows:

\[ \text{Rate of deposition} = K_d (Re.Pr)^{1/3} \exp(E_a/RT_w), \]

where \(K_d\) is constant and can be determined from the experimental data. As it was already described, there are two proposed mechanisms for the removal rate which are expressed as follows:

1. The rate of removal depends on shear-related or velocity flow which is expressed as follows:

\[ \text{Rate of removed} = K_{\text{removal}} \tau_w. \]

2. The rate of removal instead of being dependent on the shear stress is affected by the rate of mass-transfer which was already proposed by Polley et al.\(^\text{[36]}\)

\[ \text{Rate of removal} = K_{\text{removal}} \tau_w, \]

where \(K_{\text{removal}}\) and \(n\) constants have been obtained by curve-fitting experimental data in final model. After substituting equations in equations, following equations are obtained:

\[ m = (k_d(Re.Pr)^{1/3} \exp(E_a/RT_w) - K_{\text{removal}} \tau_w)C_{Ab}^{1.2} \quad \text{[6]} \]
\[ m = (k_d(Re.Pr)^{1/3} \exp(E_a/RT_w) - K_{\text{removal}} \tau_w)C_{Ab}^{1.2} \quad \text{[7]} \]

Results of curve fitting will tabulate in Table 3. Results in Table 3 indicate that Equation (7) is better than Equation (6) for fitting of data. It seems that curve fitting is better when the rate of removal is dependent on rate of the rate of mass-transfer.

5. CONCLUSIONS

In the present study, after carefully verifying of the use of the “washing method” for the measurements of asphaltene deposition thickness, the effects of operating parameters such as oil velocity and pipe surface temperature on the deposition process in asphaltene crude oil in a tube was investigated. The results showed that the deposition rate was increased as the surface temperature and asphaltene concentration were increased. The experimental results also indicate that the deposition rate is inversely proportional to the oil velocity and the thickness of deposited components was decreased as the oil velocity was...
increased. Besides, fouling model was employed to predict value of asphaltene deposition from the test loop and good agreement between the predicted thickness and the measured value was noticed.

REFERENCES