

# An Investigation of Asphaltene Deposition Mechanisms During Natural Depletion Process by a Two Phase Modeling Using Genetic Algorithm Technique

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## ABSTRACT

In this work, the natural depletion process in sandstone and carbonate cores was modeled to investigate the asphaltene deposition mechanisms. A new permeability reduction correlation was proposed based on the Minssieux model that considers a combination of surface deposition, pore throat plugging, and filtration cake mechanisms. The results showed that the filtration cake is a dominant asphaltene deposition mechanism during natural depletion process in both core samples. Therefore, a modified model was proposed with adding formation of filtration cake mechanism due to pore filling to the Wang and Civan deposition model. The absolute average deviation (AAD (%)) for permeability reduction between the results of the three models (including new correlation, the modified model, and Wang and Civan model) and the experimental data were calculated and reported. These values for the three models were 3.28, 2.67, and 4.83% for sandstone core and 3.01, 2.58, and 4.69% for carbonate core respectively. The results showed that the modified model proposed in this study presented good performance for asphaltene deposition prediction

**Keywords:** Natural Depletion, Asphaltene Deposition, Modeling, Filtration Cake

## INTRODUCTION

Asphaltene is a mixture of heavy components in the oil reservoirs that can be precipitated out of oil by changing pressure, temperature, and oil composition. Natural depletion is one of the usual processes of asphaltene deposition. In this process, asphaltene particles set on the rock surface due to pressure changes. Asphaltene precipitation causes permeability reduction and formation damage in oil reservoirs, and it ultimately affects the oil production [1-3]. To predict the amount of precipitated asphaltene, several thermodynamic models

such as solubility model based on the Flory-Huggins polymer theory [4-7], solid model based on the solid-liquid equilibrium [8, 9], and thermodynamic models based on the various cubic and association equation of states [10-13] have been presented. On the other hand, several models have been proposed to simulate the permeability reduction as a result of asphaltene deposition in porous media. Gruesbeck and Collins proposed empirical correlations for calculating fluid velocities in pluggable and non-pluggable pathways.

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They found out that the permeability reduction is a function of the volumes of deposited asphaltene. They showed that the entrainment mechanism could decrease the production [14]. Wang and Civan developed a one-dimensional, three-phase asphaltene deposition model, including surface deposition, entrainment, and plugging terms [15]. Soulgani et al. investigated the effects of concentration, velocity, and temperature parameters on the asphaltene deposition and developed the surface deposition term based on these parameters [16]. Jafari Behbahani et al. developed a four-component, four-phase model based on the multilayer adsorption theory to describe formation damage during natural depletion and CO<sub>2</sub> injection processes. They showed that the oil and core types can affect the amount of asphaltene deposition during natural depletion process. They concluded that the live oil flooding caused more asphaltene deposition than dead oil flooding in porous media [17-19]. Khalifeh et al. investigated the asphaltene deposition on the carbonate and sandstone rock surfaces during dynamic flow tests. They simulated the permeability reduction by commercial software. They showed that the surface deposition and entrainment mechanisms were more important for asphaltene deposition process in carbonate rock than sandstone rock surfaces, and pore plugging mechanism had the same effect in both types of rocks [20]. Bagherzadeh et al. studied the permeability reduction mechanisms during simultaneous injection of CO<sub>2</sub> and recombined oil into sandstone and carbonate cores at reservoir conditions. They concluded that the surface deposition and pore plugging mechanisms were simultaneously effective on permeability reduction in initial injected pore volumes for both types of the rock. Thus, they presented a new model consisting of surface deposition and pore plugging mechanisms for permeability reduction predictions [21]. Recently Fallahnejad and Kharrat modeled the asphaltene deposition process, and they considered the asphaltene

particles in three conditions (precipitated, flocculated, and deposited). They used asphaltene precipitation model (solid model), kinetic model (first-order chemical), and deposition model consisting of adsorption, pore throat plugging, and re-entrainment terms to predict porosity and permeability reduction during the natural depletion process [22].

In this work, the natural depletion process in a sandstone and carbonate core samples from the Iranian oil reservoirs was modeled and simulated under reservoir conditions to investigate the asphaltene deposition mechanisms. A new correlation was proposed based on the Minssieux model to calculate the weights of different deposition mechanisms and find out the dominant mechanism in asphaltene deposition process. A new model was also proposed by the modification of the Wang and Civan deposition model. The dominant mechanism (internal cake filtration due to pore filling) was added to Wang and Civan model to predict the asphaltene deposition process as a two-phase system. Finally, the permeability reduction predictions were validated by experimental data and compared with that of Wang and Civan model.

## THEORETICAL CALCULATIONS

The live oil data from an oilfield in the southeast of Iran was used in this study. The oil compositions and properties of two different core flooding tests are given in Tables 1 and 2 respectively [17].

**Table 1: Studied live oil compositions**

Component	mol. %	Component	mol. %	Component	mol. %	Component	mol. %
H <sub>2</sub> S	0	C <sub>2</sub>	8.24	i-C <sub>5</sub>	1.38	C <sub>8</sub>	3.88
N <sub>2</sub>	0.3	C <sub>3</sub>	4.14	n-C <sub>5</sub>	1.59	C <sub>9</sub>	2.49
CO <sub>2</sub>	1.83	i-C <sub>4</sub>	1.19	C <sub>6</sub>	6.95	C <sub>10</sub>	4.03
C <sub>1</sub>	22.7	n-C <sub>4</sub>	3.61	C <sub>7</sub>	4.1	C <sub>11</sub>	2.85
$\rho_{oil} = 767.4 \text{ kg/m}^3$ , $\mu_{oil} = 4.35 \text{ cP}$ , MW C <sub>12+</sub> = 491 g/gmole, SG C <sub>12+</sub> = 0.9853						C <sub>12+</sub>	28.74
P <sub>reservoir</sub> = 326 bar, T <sub>reservoir</sub> = 96 °C, P <sub>sat</sub> = 97 bar, MW <sub>bottom hole live oil</sub> = 182 g/gmole							

**Table 2: Properties of the core flooding tests.**

Core type	Length (cm)	Diameter (cm)	Porosity (%)	Permeability (md)	PV	P <sub>backpressure</sub> (bar)	Q (cm <sup>3</sup> /hr.)
Sandstone	5.96	3.7	13.15	22.8	8.5	180	30
Carbonate	4.5	3.7	26	2.7	13.5	180	10

### Dominant Mechanism in Asphaltene Deposition

In this section the dominant mechanism in asphaltene deposition process during natural depletion was found by Minssieux model [23]. This model consists of three possible mechanisms, including formation of filtration cake due to pore filling, surface deposition, and pore throat plugging as respectively given in Equations 1-3:

$$\frac{k}{k_0} = \frac{1}{1 + A \cdot (PVI)} \quad (1)$$

$$\sqrt{\frac{k}{k_0}} = 1 - B \cdot (PVI) \quad (2)$$

$$\frac{k}{k_0} = 1 - C \cdot (PVI) \quad (3)$$

where,  $\frac{k}{k_0}$  is permeability reduction, and A, B, and C are filtration cake, surface deposition, and pore throat plugging mechanism's parameters respectively. PVI is the value for pore volume injection. The genetic algorithm method was used to find the model parameters (A, B, and C) and to investigate the dominant process during natural depletion.

A new permeability reduction correlation, which considers a combination of surface deposition, pore throat plugging, and filtration cake mechanisms, was also proposed to calculate the weights of different

asphaltene deposition mechanisms as follows:

$$\frac{k}{k_0} = d_1 \cdot \left( \frac{1}{1 + A \cdot (PVI)} \right) + d_2 \cdot (1 - B \cdot (PVI))^2 + d_3 \cdot (1 - C \cdot (PVI)) \quad (4)$$

$$d_1 + d_2 + d_3 = 1 \quad (5)$$

where,  $d_1$ ,  $d_2$ , and  $d_3$  are weight fractions of filtration cake, surface deposition, and pore throat plugging mechanisms respectively. These parameters are between 0 and 1, and they are obtained by optimization using a genetic algorithm technique.

### Thermodynamic Model for Asphaltene Precipitation

The solid model was employed to model the precipitation of asphaltene. In this model, the oil was divided to precipitating asphaltene in the solid phase and non-precipitating asphaltene in the liquid phase. The asphaltene component in the liquid and solid phases is in thermodynamic equilibrium. The precipitating asphaltene was modeled by multiphase flash calculation using PR and SRK equations of states. The fugacity of a precipitating component in the solid phase was calculated using the following equation [24]:

$$\ln f_s = \ln f_s^* + \frac{v_s (p - p^*)}{RT} \quad (6)$$

where,  $p$  is the pressure, and  $f_s$  is the solid fugacity;  $v_s$  is the solid molar volume, and  $R$  stands for the universal gas constant;  $T$  represents the temperature, and \* superscript indicates the reference condition.

### Dynamic Modeling of Asphaltene Deposition

The oil, gas, and asphaltene mass balance equations, momentum equations, porosity, and permeability models were used as a two-phase system at a constant temperature for dynamic modeling the asphaltene deposition in porous media as follows respectively [15]:

$$\frac{\partial}{\partial t}(\varnothing S_l \rho_l w_{ol}) + \frac{\partial}{\partial x}(\rho_l u_l w_{ol}) = 0 \quad (7)$$

$$\frac{\partial}{\partial t}(\varnothing S_v \rho_g + \varnothing S_l \rho_l w_{gl}) + \frac{\partial}{\partial x}(\rho_l u_l w_{gl} + \rho_l u_g) = 0 \quad (8)$$

$$\frac{\partial}{\partial t}(\varnothing S_l \rho_A C_A + \varnothing S_l \rho_l w_{Al}) + \frac{\partial}{\partial x}(\rho_l u_l w_{SAI} + \rho_l u_l w_{Al}) = -\rho_A \frac{\partial E_A}{\partial t} \quad (9)$$

$$u_l = -\frac{k k_{rl}}{\mu} \frac{\partial P}{\partial x} \quad (10)$$

$$u_v = -\frac{k k_{rv}}{\mu} \frac{\partial P}{\partial x} \quad (11)$$

$$\varnothing = \varnothing_0 - E_A \quad (12)$$

$$k = k_0 \left( \frac{\varnothing}{\varnothing_0} \right)^3 \quad (13)$$

where,  $E_A$  is the volume fraction of deposited asphaltene in the bulk volume of the porous media.  $W_{Al}$  and  $W_{SAI}$  are the mass fractions of the dissolved and suspended asphaltene in the liquid phase respectively.  $W_{ol}$  and  $W_{gl}$  represent the mass fractions of the oil and gas in the liquid phase respectively.  $C_A$  stands for the volume fraction of the suspended asphaltene precipitated in the liquid phase.  $\rho_l$ ,  $\rho_g$ , and  $\rho_A$  represent the oil, gas, and asphaltene densities respectively.  $\varnothing$  is the porosity, and  $U_l$  and  $U_g$  stand for the liquid and gas velocity respectively.  $S_l$  and  $S_v$  are the liquid and vapor phase saturations respectively.  $\mu$  is the viscosity, and  $k$  is the permeability.  $K_{rl}$  and  $K_{rv}$  represent the relative permeabilities of the liquid and vapor phases respectively.

The Wang and Civan model was used as the asphaltene deposition model [15]:

$$\frac{\partial E_A}{\partial t} = \alpha \varnothing S_l w_{SAI} \frac{\rho_l}{\rho_A} - \beta E_A (v - v_{cr}) + \gamma S_l u_l w_{SAI} \frac{\rho_l}{\rho_A} \quad (14)$$

$$v = \frac{u_l}{\phi} \quad (15)$$

$$\beta = \beta_i \quad \text{if } v > v_{cr}, \quad \text{else } \beta = 0 \quad (16)$$

$$\gamma = \gamma_i (1 + \sigma E_A) \quad \text{if } D_{pt} \leq D_{ptcr}, \quad \text{else } \gamma = 0 \quad (17)$$

$v$  and  $v_{cr}$  are the interstitial and critical interstitial velocity of liquid phase respectively, and  $\sigma$  represents the snowball-effect deposition constant.  $D_{pt}$  and  $D_{ptcr}$  stand for the pore throat diameter and critical pore throat diameter respectively. The first, second, and third terms in the asphaltene deposition model (the right hand side of Equation 14) are the surface deposition rate, entrainment rate, and pore throat plugging rate respectively, the parameters of which are correspondingly  $\alpha$ ,  $\beta$ , and  $\gamma$ .

An auxiliary equation was required to explain the relation between vapor and liquid phase saturations (Equation 18). Saturation is a measure of the fluid content of the porous rock [25]. The liquid and vapor saturations were calculated by flash calculation at different pressures during natural depletion process.

$$S_v + S_l = 1 \quad (18)$$

The boundary and initial conditions can be considered as follows:

$$\varnothing = \varnothing_0, \quad k = k_0, \quad E_A = 0 \quad : 0 \leq x \leq L, \quad t = 0 \quad (19)$$

$$u_l = \frac{Q}{A} \quad : x = 0, \quad t > 0 \quad (20)$$

$$p = p_{\text{back pressure}} \quad : x = L, \quad t > 0 \quad (21)$$

## Modification of Wang and Civan Deposition Model

A modified model was proposed with adding internal cake filtration mechanism due to pore filling to the Wang and Civan model for predicting the permeability reduction during natural depletion process (Equation 22). The pore filling, following the pore throat plugging, leads to an internal cake formation. This mechanism was presented by Chang and Civan [26], but it has not been used simultaneously with surface deposition, entrainment, and pore throat plugging mechanisms for predicting asphaltene deposition up to now.

$$\frac{\partial E_A}{\partial t} = \alpha \varnothing S_l w_{SAI} \frac{\rho_l}{\rho_A} - \beta E_A (v - v_{cr}) + \gamma S_l u_l w_{SAI} \frac{\rho_l}{\rho_A} + \lambda \varnothing S_l u_l w_{SAI} \frac{\rho_l}{\rho_A} \quad (22)$$

$$\lambda \neq 0 \quad \text{if } t > t_{cr} \text{ or } b < b_{cr}, \text{ else } \lambda = 0 \quad (23)$$

$$b = \frac{D_{pt}}{D_{asp}} \quad (24)$$

The fourth term in the modified asphaltene deposition model (the right hand side of Equation 22), is the pore filling (or internal filtration cake) rate ( $\lambda$ ).  $t_{cr}$  represents the critical time when the pore throat is first jammed by asphaltene particles.  $D_{asp}$  is an asphaltene particle diameter, and  $b_{cr}$  is the critical value below which ( $b_{cr} = 7$ ) the filtration cake is occurred [27].

Values of asphaltene particle size depend on the method of measurement and also on the solvent type. Leontaritis et al. concluded that the asphaltene size distributions before asphaltene flocculation are about 20 to 350 Å, while after flocculation, asphaltene particles larger than 1000 Å are formed [28]. Leontaritis presented asphaltene particle size distribution versus asphaltene fraction in the oil reservoir at different pressures [29]. Therefore, based on the literature studies [28, 29] and asphaltene weight fraction value of using oil (16.3 wt.%), asphaltene particle diameter is considered 1000 Å.

### Numerical Solution

A computer program was written and used to solve the above equations simultaneously by backward implicit finite difference method. Numerical simulation was carried out in a Cartesian system with 200 grid blocks and 50 time steps with a  $\Delta t$  of 100 seconds. The genetic algorithm technique was used as the optimization tool for the determination of the model parameters ( $\alpha$ ,  $\beta$ ,  $v_{cr}$ ,  $v_p$ ,  $\sigma$ , and  $\lambda$ ). In the genetic algorithm method, the numbers of generation, population size, and elite count were taken 1000, 100, and 2 respectively. Population size determines the number of individuals in each generation. The selection of a large population size in the calculation causes the genetic algorithm to search the solution region completely in order to find the global minimum. Elite count is the number of individuals with the best fitness values in the current

generation insured to survive to the next generation. The rank method was used for fitness scaling, while stochastic uniform was used as a method to specify the next generation. In this study, the objective function is a root mean squared error (RMSE):

$$RMSE = \sqrt{\frac{SSE}{m}} \quad (25)$$

$$SSE = \sum_{i=1}^m \left( \left( \frac{k}{k_0} \right)_{exp,i} - \left( \frac{k}{k_0} \right)_{model,i} \right)^2 \quad (26)$$

where, SSE is the sum of squared error.  $\left( \frac{k}{k_0} \right)_{model,i}$  and  $\left( \frac{k}{k_0} \right)_{exp,i}$  represents the model and the experimental values of permeability reduction respectively, and  $m$  is the number of experimental points.

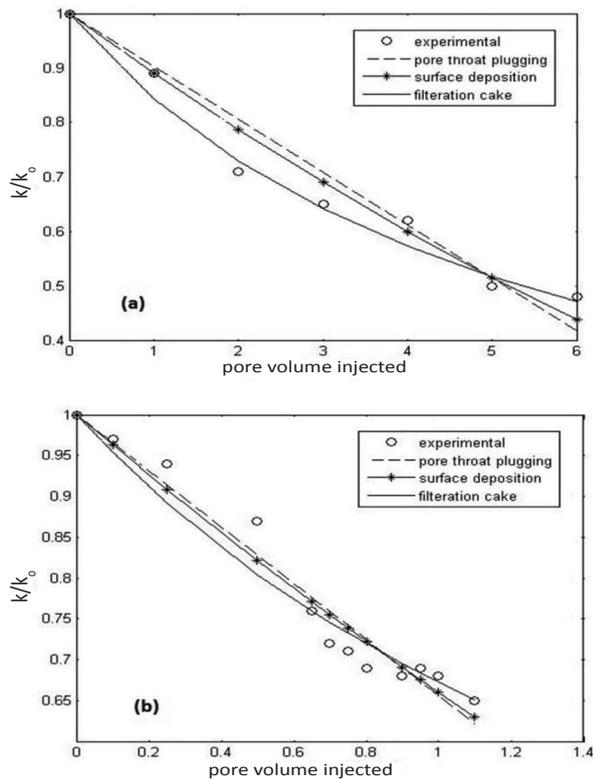
### RESULTS AND DISCUSSION

In this study, the permeability reduction for different asphaltene deposition mechanisms was calculated by Minssieux model. Table 3 shows the optimization values of Minssieux model parameters.

**Table 3: Results of Minssieux model parameters by genetic algorithm.**

Parameter (1/s)	Sand stone core	Fitness value (R <sup>2</sup> )	Carbonate core	Fitness value (R <sup>2</sup> )
Filtration cake parameter (A)	0.18667	0.992	0.48675	0.996
Surface deposition parameter (B)	0.05642	0.973	0.18751	0.987
Pore throat plugging parameter (C)	0.09708	0.984	0.34375	0.989

Figures 1a and 1b show the predictions of the permeability reduction by each mechanism correlation in the sandstone and carbonate cores respectively.



**Figure 1: The experimental and calculated permeability reduction by Minssieux model in (a) sandstone core (b) carbonate core.**

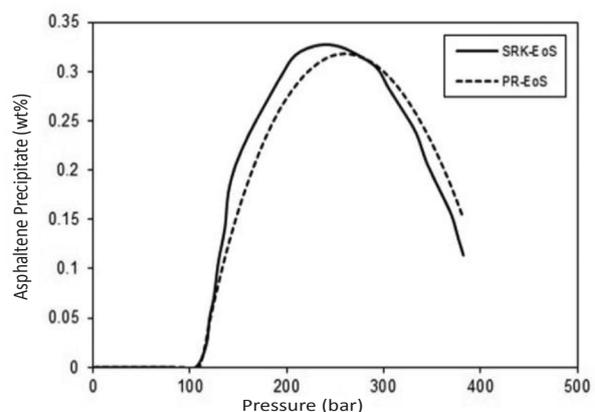
The results show that the formation of filtration cake due to pore filling is dominant in the deposition of asphaltene with the highest fitting values to the experimental data ( $R^2 > 0.99$ ) in both sandstone and carbonate core samples. The weights of asphaltene deposition mechanisms were estimated by the proposed correlation, which considers a combination of surface deposition, pore throat plugging, and filtration cake mechanisms. The proposed correlation parameters ( $d_1, d_2$ , and  $d_3$ ) were optimized by the genetic algorithm technique. Table 4 shows the optimum values of the parameters in the proposed correlation.

**Table 4: Results of the proposed correlation parameters by genetic algorithm.**

Parameter	Sandstone core	Carbonate core
$d_1$	0.82	0.59
$d_2$	0.04	0.19
$d_3$	0.14	0.22

The results demonstrate that the controlling mechanism during asphaltene deposition followed the order of filtration cake  $\gg$  pore throat plugging  $>$  surface deposition for both sandstone and carbonate core samples. Also, it can be concluded that the surface deposition and pore throat plugging mechanisms have higher effects on asphaltene deposition in carbonate core than in the sandstone core. This may be due to the fact that the carbonate core surface contains polar groups and asphaltene is a polar component. Thus, the  $\pi$ -interaction and hydrogen bonding can form between the asphaltene and core surface polar groups, and they increase the amount of asphaltene deposition on the surface of the carbonate rock samples [30]. The obtained results are in good agreement with literature studies [19-21].

After finding the dominant mechanism in asphaltene deposition process, the precipitation of asphaltene under different pressures was calculated by using solid model and using PR and SRK equations of state. Figure 2 shows the weight fraction of precipitated asphaltene versus pressure. The results show that the same results are obtained by using PR and SRK equations of state. The results of thermodynamic modeling were applied to asphaltene dynamic modeling so as to calculate the precipitated asphaltene.

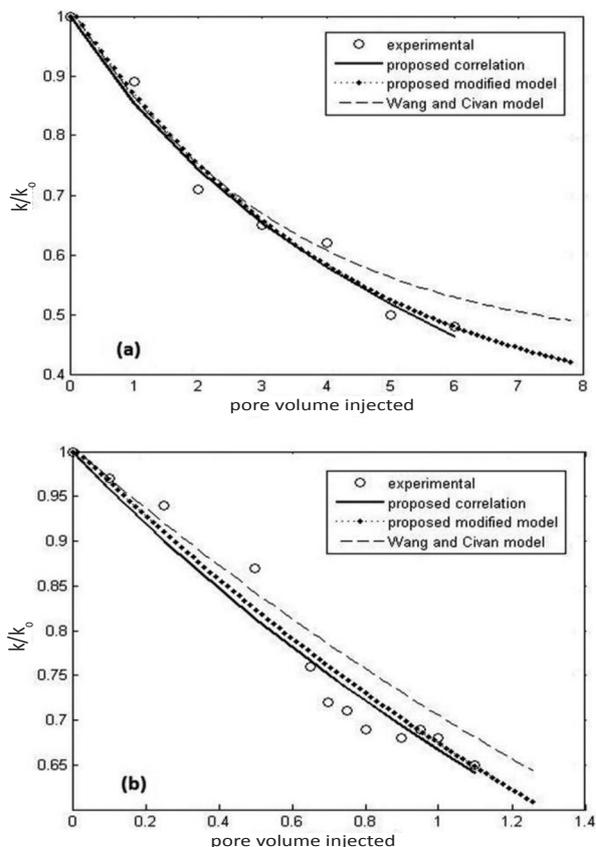


**Figure 2: Asphaltene thermodynamic modeling by solid model using PR and SRK equations of state.**

For dynamic modeling, a modified model was presented with adding dominant mechanism (internal cake filtration due to pore filling) to Wang and Civan model for predicting asphaltene deposition process. The genetic algorithm technique was used for determining model parameters. Table 5 shows the optimum values of tuning parameters in Wang and Civan and our proposed modified models. Figures 3a and 3b show the predictions of permeability reduction by the proposed correlation, Wang and Civan model, and the proposed modified model during natural depletion process in sandstone and carbonate cores respectively.

**Table 5: Asphaltene deposition model parameters obtained by genetic algorithm.**

Model para.	Sandstone core		Carbonate core	
	Wang and Civan model	Proposed modified model	Wang and Civan model	Proposed modified model
$\alpha$ (1/s)	0.00085	0.00073	0.00041	0.00049
$\beta$ (1/cm)	0.04772	0.04485	0.00179	0.03252
$U_{cr}$ ( $\frac{cm}{s}$ )	0.00194	0.00446	0.00740	0.101494
$\gamma_i$ (1/cm)	0.00284	0.00217	0.04773	0.06087
$\sigma$	0.03994	0.02226	0.01116	0.07060
$\lambda$	-	0.08961	-	0.0989



**Figure 3: Experimental and calculated permeability reduction by the proposed correlation, Wang and Civan model, and the proposed modified model in (a) sandstone core (b) carbonate core.**

Average absolute deviations (AAD (%)) of the predicted permeability reduction from the experimental data using three different models are presented in Table 6. These results confirm that the proposed modified model based on the filtration cake mechanism is capable of correlating the permeability reduction experimental data with an AAD of 2.67% and 2.58% in sandstone and carbonate cores respectively. The proposed correlation based on the combination of three mechanisms (surface deposition, pore throat plugging, and filtration cake) correlated asphaltene deposition experimental data with an AAD of 3.28% and 3.01% in sandstone and carbonate cores respectively. However, the Wang and Civan model has an AAD of 4.83% in sandstone core and an AAD of 4.69% in carbonate core.

**Table 6: Average absolute deviation of the correlated permeability reduction from the experimental results by the proposed correlation, proposed modified model, and Wang and Civan model.**

Core type	Proposed correlation (AAD (%))	Proposed modified model (AAD (%))	Wang and Civan model (AAD (%))
Sandstone	3.28	2.67	4.83
Carbonate	3.01	2.58	4.69

Therefore, the modified model proposed in this study showed good accuracy for asphaltene deposition prediction in comparison with the other models. This suggests that the filtration cake mechanism has an important role in asphaltene deposition process. Moreover, the obtained results indicate that the proposed modified model has similar effects on asphaltene deposition prediction in carbonate core samples with an AAD of 2.58% in comparison to sandstone core samples with an AAD of 2.67%.

## CONCLUSIONS

A new correlation based on the Minssuix model was proposed to investigate the permeability reduction during natural depletion process. The results showed that the filtration cake formation is a dominant mechanism in formation damage. Furthermore, the surface deposition and pore throat plugging mechanisms indicated more effects on asphaltene deposition process in carbonate core comparing to sandstone core sample. Therefore, the new modified model was proposed with adding filtration cake mechanism, due to pore filling, to Wang and Civan deposition model. It was found that the proposed modified model was more accurate than Wang and Civan model. It can be concluded that the formation of filtration cake mechanism has the highest effect on asphaltene deposition in both sandstone and carbonate core samples.

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