



# Investigation of Asphaltene Precipitation using Response Surface Methodology Combined with Artificial Neural Network

Zeinab Hosseini-Dastgerdi<sup>a</sup>, Saeid Jafarzadeh-Ghoushchi<sup>b,\*</sup>

Faculty of Chemical Engineering, Urmia University of Technology, Urmia, Iran  
Faculty of Industrial Engineering, Urmia University of Technology, Urmia, Iran

Received: 4 July 2018, Revised: 14 October 2019, Accepted: 21 October 2019  
© University of Tehran 2019

## Abstract

The precipitation of asphaltene, one of the components of oil, in reservoirs, transfer lines, and equipment causes many problems. Accordingly, researchers are prompted to determine the factors affecting asphaltene precipitation and methods of avoiding its formation. Predicting precipitation and examining the simultaneous effect of operational variables on asphaltene precipitation are difficult because of the multiplicity, complexity, and nonlinearity of factors affecting asphaltene precipitation and the high cost of experiments. This study combined the use of response surface methodology and the artificial neural network to predict asphaltene precipitation under the mutual effects of various parameters. The values of such parameters were determined to reach the minimum amount of precipitation. We initially selected the appropriate algorithm for predicting asphaltene precipitation from the two neural network algorithms. The outputs of designed experiments in response surface methodology were determined using the optimum algorithm of the neural network. The effects of variables on asphaltene precipitation were then investigated by response surface methodology. According to the results, the minimum precipitation of asphaltene achieved at zero mole percent of injected nitrogen and methane, 10–20 mole percent of injected carbon dioxide, asphaltene content of 0.46, the resin content of 16.8 weight percent, the pressure of 333 psi, and temperature of 180 °F. Results showed that despite the complexities of asphaltene precipitation, the combination of artificial neural network with response surface methodology can be successfully used to investigate the mutual effect of different variables affecting asphaltene precipitation.

## Keywords:

Artificial Neural Network  
Asphaltene,  
Desirability,  
Precipitation,  
Response Surface  
Methodology

## Introduction

Oil is a compound comprising multiple hydrocarbons with different molecular weights. The identification of all these compounds is impossible, so oil hydrocarbons are instead classified into the four main groups of asphaltene, resin, aromatic, and saturate. Asphaltene is the heaviest and most polarized component of crude oil. This compound is soluble in aromatics such as benzene and toluene and insoluble in light n-alkanes such as n-heptane and n-pentane [1-4]. Studies show that asphaltene exists in oil in colloidal form and that the asphaltene solubility in oil is dependent on oil polarity and components [5,6]. Changes in thermodynamic properties such as pressure and temperature are also effective for asphaltene solubility. Any change in the oil thermodynamic equilibrium affects asphaltene solubility and leads to asphaltene precipitation [7].

\* Corresponding author:

Email: s.jafarzadeh@uut.ac.ir (S. Jafarzadeh-Ghoushchi)

Asphaltene precipitation in reservoir, transfer, and refinery equipment causes multiple problems such as a decrease in permeability, change of tank wettability, and clog of transfer lines. Multiple experimental studies were conducted to investigate the effect of different factors on asphaltene precipitation. The effects of pressure and temperature and the injection of nitrogen, methane, and Carbon dioxide gases on asphaltene precipitation are investigated [8-13]. Some scientists claim that the amount of asphaltene precipitation is directly proportional to its structure, whereas some others emphasize on the effect of resins on asphaltene precipitation [5,14,15].

The accurate determination of the effect of each effective parameter on the precipitation under reservoir conditions requires multiple experiments. The execution of asphaltene precipitation determination experiments in high temperature and pressure encounters various problems. These problems include excessive time consumption and high costs of experiments and equipment. The amount of problems increases when asphaltene precipitation in porous media is considered. Many studies used mathematical modeling to investigate the precipitation mechanism and to determine the effective parameters for asphaltene precipitation considering the restriction of performing these experiments. Various models are available to predict asphaltene precipitation. Some researchers used state equations such as Peng–Robinson (PR) and Soave–Redlich–Kwong (SRK) [16,17]. Examples of these models include polymeric solution theory, association theory, and molecular thermodynamics [18-20]. The common models in the prediction of asphaltene precipitation cause error and limitation because the calculations in these models possess multiple input parameters that require many experiments. Therefore, the lack of experimental data to predict input parameters leads to erroneous results. Despite all these drawbacks, these models have been used for many years to study asphaltene precipitation [21,22]. Thus, researchers aim to determine a model, which can predict the results of asphaltene precipitation with maximum accuracy. The prediction of precipitation is nonlinear and ambiguous because of the multiplicity and complexity of the effective parameters on the asphaltene precipitation phenomenon [23].

The artificial neural network is a useful method for solving nonlinear engineering problems [24]. The artificial neural network is a computer model that simulates complex engineering problems in a manner inspired by the biological central neural network. This model is composed of an input layer, one or more hidden layers, and an output layer with input and output elements that are connected to one another in a nonlinear form by some constants [25,26].

Central Composite Design (CCD) is a useful optimization method. In the traditional optimization methods, optimization is performed by changing a variable while the other variables remain constant. However, in CCD, the effect of variable interaction is considered in the optimization [27]. The system behavior can be predicted under various parameters and the most effective parameters can be determined by using the response surface methodology [25]. Thus, this method can investigate the behavior of the asphaltene precipitation under the simultaneous effects of different parameters and can determine the most effective parameters for asphaltene precipitation. Some researchers used the artificial neural network to model the asphaltene precipitation. Ahmadi et al. [28] studied the effect of temperature and pressure on asphaltene precipitation using the artificial neural network. Abedini et al. [29] investigated the effect of temperature on asphaltene precipitation in the artificial neural network. Khamechi et al. [30] studied the effect of carbon dioxide injection on the asphaltene precipitation onset and bulb pressure using neural network model. Ashoori et al. [31] compared the artificial neural network and empirical equations to predict asphaltene precipitation of dead oil under the effect of temperature and solvent ratio. Results showed that the neural network model can accurately predict asphaltene precipitation. The artificial neural network has been utilized for asphaltene precipitation modeling of dead oil in most of these studies. However, the effect of all parameters was not investigated due to the lack of input experimental data.

Experimental studies showed that in actual reservoir conditions, various variables such as temperature, pressure, amount of asphaltene and resin, saturation pressure, and American Petroleum Institute (API) gravity affect asphaltene precipitation [32-35]. Previous studies investigated the effect of limited parameters, and the simultaneous effect of operational variables on asphaltene precipitation was not determined [32-35]. In this study, the effect of temperature, pressure, saturation pressure at reservoir temperature, API gravity of stock tank oil, asphaltene, and resin composition, and the amount of injected nitrogen, methane, and Carbon dioxide was investigated, simultaneously. However, it is very difficult to investigate the simultaneous effect of variables on asphaltene precipitation by performing multiple experiments; it requires much time and cost. Though, ANN is a flexible mathematical structure which is capable to recognize complex nonlinear relationships between input and output data sets, the calibration model based on ANN is more unstable. On the other hand, the trained network model is not applicable to the optimization of a different number of experimental data. In order to overcome this problem, it seems that the combination of ANN and CCD is the appropriate method which can be considered to identify relationships between the inputs, operating condition, and outputs, asphaltene precipitation. Therefore, this article represents a new calculation approach for the estimation of asphaltene precipitation using combined ANN and CCD methods.

This work represents a novel way to approximate the asphaltene precipitation using the combination of CCD and ANN techniques. The primary use of this method is to reliably guarantee the dependence between the process factors and the outcome of the experiments, asphaltene precipitation. This methodology for predicting asphaltene precipitation has not been reported yet. Also, there is no study published in the literature, focuses on the prediction of asphaltene precipitation using this method to investigate the effect of methane, nitrogen, and carbon dioxide injection, and PVT properties, simultaneously. The last novelty of this work is using more than 100 experiments with an extensive range of changes that were introduced into ANN, to train the network and generate the model. Well training depends on the number of input experiments. More input experiments involved, the better the capability to approximate a real system.

In this research, firstly, more than 100 experimental data derived from previous studies were used to investigate the effect of different parameters on asphaltene precipitation, which was used for training the neural network model [32-40]. A complex nonlinear relation between nine operational parameters and the amount of asphaltene precipitation is established, and an optimum model with the least error is determined to predict asphaltene precipitation by using artificial neural network.

Finally, more than 500 precipitation static experiments were designed, considering the effect of nine effective operational parameters. Subsequently, the simultaneous effect of effective parameters on asphaltene precipitation was investigated by using ANN and CCD methods.

## Research Method

### Data Collection

The importance of asphaltene precipitation and its instability under different conditions led to the execution of many experimental studies under various operational conditions to identify the effective parameters of precipitation. The experimental data collection covers all the effective parameters and forms the input data bank. Data are analyzed in the data collection and summarized to reach the highest efficiency and the least cost. The data bank includes more than 100 PVT experiments obtained from previous studies [32-40]. The data bank is used as the input for the artificial neural network.

## Input Effective Parameters

The results of the studies showed that under static conditions, the asphaltene precipitation amount is dependent on the temperature, pressure, and composition [32-40]. The type of crude oil and injected gas into the oil during the enhanced oil recovery processes also affects the asphaltene precipitation amount [32-40]. Thus, the most important and effective parameters are temperature, pressure, saturation pressure at reservoir temperature, API gravity of stock tank oil, asphaltene and resin composition, and the amount of injected nitrogen, methane, and Carbon dioxide under static conditions. These parameters are the input data for the artificial neural network. Table 1 shows the range of the input parameters for the artificial neural network. Table 2 represents the experiments reported in the literature which were used as input data.

## Structure of The Artificial Neural Network

The artificial neural network is a technique used to create a nonlinear relation between plenty of input and output data. In mathematical modeling, the input parameters adapt to the specified output values using different algorithms. In the artificial neural network, this aim is accomplished by learning from some network-introduced input/output data. Then, the neural network uses the last data learned pattern to predict the desired output.

The most common model of the neural network is the multilayer feed-forward neural network [41,42]. This model has wide applications in computer science and oil engineering. In this type of neural network, neurons are located in input, hidden, and output layers. Each neuron in the hidden and output layers receives input data from neurons in its previous layer. The effect of each relation in the input, hidden, and output layers on the prediction of the neural network is determined by weights and biases, which are referred to as the neural network parameters [28,30,43]. The optimum predictive algorithm in the neural network depends on the algorithm, number of layers, number of neurons in each layer, and neural network parameters [25].

The data are entered into the neural network software, Neural Power version 2.5 (CPC-X software, USA). Different algorithms of the neural network were investigated to identify the best prediction of the test and train data. The artificial neural network model used was feed-forward with one hidden layer. Two algorithms, namely, quick propagation (QP) and batch backpropagation (BBP) were considered to determine the optimum algorithm for asphaltene precipitation prediction. The trial and error method was used to identify the optimal number of neurons in the hidden layer. The number of neurons varies in the range of 6 to 10.

The best algorithm and the optimal number of neurons are selected based on the least error between the predicted value of asphaltene precipitation and the experimental results. Root Mean Square Error (RMSE) and the coefficient of determination ( $R^2$ ) are the parameters used to determine the prediction capability of the neural network defined as follows [25]:

$$RMSE = \sqrt{\frac{\sum_{i=1}^N (y_p - y_e)^2}{N}} \quad (1)$$

$$R^2 = 1 - \frac{\sum_{i=1}^N (y_p - y_e)^2}{\sum_{i=1}^N (y_p - y_a)^2} \quad (2)$$

$N$  is the number of data,  $y_e$  is the amount of asphaltene precipitation under the experimental conditions,  $y_p$  is the predicted amount of precipitation by the neural network model, and  $y_a$  is the mean value of the asphaltene precipitation under experimental conditions. A neural network model is acceptable if the RMSE parameter is as small as possible and if  $R^2$  approaches unity.

## Response Surface Methodology Structure

Studies showed that the asphaltene precipitation mechanism is extremely complex, and many parameters such as pressure, temperature, and composition affect precipitation because crude oil comprises many components with unknown behavior. The changes in each parameter can influence the efficacy of other parameters on the asphaltene precipitation, according to the multiplicity of effective parameters on the asphaltene precipitation and the complexity of the mechanism. The simultaneous effect of all factors on the asphaltene precipitation cannot be investigated experimentally. The best solution under such conditions is the use of mathematical models. Artificial neural network cannot provide the model equation for the asphaltene precipitation behavior in terms of effective parameters despite its success in predicting the asphaltene precipitation. The artificial neural network acts similar to the human brain and represents the response based on the learning received from data.

The response surface method is a combination of statistical and mathematical models and is a branch of the experimental design. The response surface methodology can (i) investigate the effect of independent variables and the interaction between the variables on the response, (ii) predict the response and determine the important level of variables, (iii) create the quadratic regression model between the variables and response, and (iv) determine the optimal parameters to obtain the maximum or minimum response [25,27,44]. The response variable (asphaltene precipitation,  $Y$ ) is related to the effective parameters (temperature, pressure, the composition of asphaltene, and resin) by a second-order polynomial equation [25,27,44]. The model equation is as follows [45]:

$$Y = \beta_0 + \sum_{i=1}^k \beta_i X_i + \sum_{i=1}^k \beta_{ii} X_i^2 + \sum_{i < j=1}^k \beta_{ij} X_i X_j \quad (3)$$

In this relation,  $\beta_0$  is a constant,  $\beta_i$  is a linear constant,  $\beta_{ii}$  is a second-order constant, and  $\beta_{ij}$  is the cross product constant.  $X_i$  and  $X_j$  are the independent variables such as temperature ( $X_1$ ), pressure ( $X_2$ ), the composition of asphaltene ( $X_3$ ), and resin ( $X_4$ ). The effect of the variables (temperature, pressure, the composition of asphaltene, and resin) and the interaction between them on the response variable (asphaltene precipitation) was investigated using Expert Design (7.1.6 stat-Ease Inc., Minneapolis, MN, USA). In addition, the operational conditions that result in the minimum amount of precipitation were determined using the response surface method.

## Result and Discussion

### Optimal Neural Network Model

The applied neural network model includes nine inputs, one hidden layer, and an output. The input layer includes temperature, pressure, oil saturation pressure at reservoir temperature, oil API gravity, the weight percentage of asphaltene and resin in oil, the molar percentage of injected methane, carbon dioxide, and nitrogen. The output layer includes a weight percentage of asphaltene precipitation. The network input parameters were divided into three sections, namely, train, test, and validation data. If the rules of the neural network model are excessively adapted in the train data, then these rules may not fit the remaining data. To avoid this phenomenon, the model rules are evaluated and controlled in the test stage. In this stage, if the error exceeds a limit, then the data training is completed.

**Table 1.** Range of input parameters in the artificial neural network

	P [psi]	T [°F]	Asphaltene [wt (%)]	Resin [wt (%)]	Injected Nitrogen [mol (%)]	Injected Carbon dioxide [mol (%)]	Injected Methane [mol (%)]	API Gravity	Saturation Pressure [psi]	Precipitated Asphaltene [wt (%)]
Min	333	180	0.46	0.49	0.0	0.0	0.0	19.8	1090	0.25
Max	8561	255	13.8	16.8	30	54	45	6.33	3464	10.04

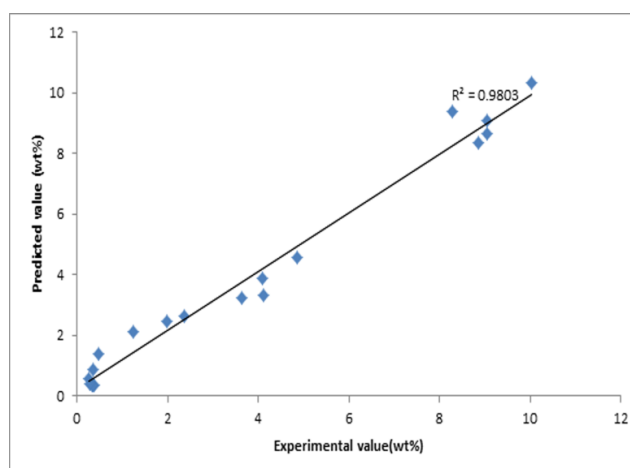
**Table 2.** The experiments reported in the references used as input data.

Reference	Reported Asphaltene Precipitation Experiments					
	Pressure Change	Temperature change	Crude oil type analysis	N <sub>2</sub> Injection	CO <sub>2</sub> Injection	CH <sub>4</sub> Injection
Reference	[32-40]	[32-40]	[34,36,37,40]	[36,39]	[32,34,38,39]	[33,36]

The two algorithms, namely, QP and BBP were investigated to determine the optimum model of the neural network. The number of nodes in the hidden layer is optimized as the effective parameter on the model algorithm. The variations in the number of nodes in the hidden layer are between 6 and 10. RMSE and  $R^2$  parameters are used as criteria to select the best algorithm with prediction capability. The capability of the algorithm improves when the RMSE parameter is smaller, and  $R^2$  parameter approaches unity. Table 3 shows a comparison between two BBP and QP algorithms. The number of nodes in the hidden layer in each algorithm shows that the BBP algorithm with seven nodes in the hidden layer at the testing and training stages has the least amount of RMSE compared with the other models. Furthermore, the amount of  $R^2$ , in this case, is also sufficiently close to one. The results showed that increasing the number of nodes in the hidden layer does not lead to error reduction. The optimized model is selected with 9-7-1 structure. The optimum model is used to predict the asphaltene precipitation experiments that are designed to be fed into the response surface software as inputs. The optimized model 9-7-1 has an output (asphaltene precipitation percentage) and nine inputs (temperature, pressure, saturation pressure, oil API gravity, the molar composition of the injected gas and weight percentage of asphaltene, and resin in the oil). The optimized model validation is performed using the data in the training and testing stages. Fig. 1 shows the predicted results of the asphaltene precipitation using the neural network model, according to the experimental results of asphaltene precipitation. In this figure, the accuracy of the neural network model in the prediction of asphaltene precipitation can be observed. The optimized model 9-7-1 predicted the precipitation amount of asphaltene under different operational conditions with extremely high accuracy ( $R^2 = 0.9803$ ).

**Table 3.** Comparison of algorithms in the prediction of asphaltene precipitation by artificial neural network

Algorithm	MLP	Training		Testing	
		RMSE	$R^2$	RMSE	$R^2$
QP	9-6-1	0.08000	0.9994	2.4402	0.4875
	9-7-1	0.09878	0.99908	1.5504	0.79311
	9-8-1	0.06084	0.99965	1.9787	0.66303
	9-9-1	0.04507	0.99981	1.7654	0.73177
	9-10-1	0.04637	0.9998	1.5465	0.79416
BBP	9-6-1	0.098284	0.99909	2.4173	0.49708
	9-7-1	0.033706	0.99989	1.2342	0.87298
	9-8-1	0.038160	0.99986	2.1957	0.58507
	9-9-1	0.044566	0.99981	1.808	0.71861
	9-10-1	0.091696	0.99921	2.208	0.68254



**Fig. 1.** Scatter plot for the predicted asphaltene precipitation using artificial neural network against the experimental results

### Response Surface Method and Model Fitting

The effective parameters on the amount of asphaltene precipitation include temperature, pressure, saturation pressure of oil under reservoir conditions, API gravity, the weight percentage of asphaltene and resin in oil, and molar composition of injected nitrogen, methane, and carbon dioxide. To investigate the simultaneous effects of these parameters on the asphaltene precipitation using the response surface method, some experiments were designed based on the effective parameters. Then, using the optimized model of the artificial neural network, the outputs of the designed experiments were determined. The variables have different units and ranges; thus, the importance level and their effectiveness on the asphaltene precipitation can be compared by expressing the variables in code forms. Therefore, a code is dedicated to each variable according to Table 4 to determine the statistical model. A total of 546 experiments were designed according to the coded variables. All analyses were performed using the design expert software. The mutual effects of variables on the amount of asphaltene precipitation were represented in the form of a second-order polynomial equation. The interacting coefficients are presented in Table 5. The first row of this table shows the coefficients of the first-order variables. P-values less than 0.05 indicate the significance of the terms. Therefore, the interacting coefficients which have a significant effect on the asphaltene precipitation ( $P\text{-value} < 0.05$ ) are specified in Table 5.

The results of the variance analysis are shown in Table 6. P-value indicates the significance of the model. If the p-value exceeds 0.1, then the model is insignificant. As shown in Table 6, the p-value is less than 0.0001. The F-value of 0.53 shows that the lack of fit is less important than the pure error [41]. The sum of square is the sum of square difference of the experimental data and the predicted data. Sum of square shows the level of model consistency on each point. High values of  $R^2$  and low values of the sum of squares result in a better model.

### Desirability analysis

Desirability analysis is performed in Design-Expert software by using the desirability function. Desirability function merges all responses into a single value, which varies in the range of 0 to 1. Zero value shows the minimum desirability function, and one value shows the maximum desirability function. The value of this function increases with the increase in the considered response and decreases with the decrease in the related response. Therefore, high desirability represents an effective process, and low desirability indicates an ineffective process. The effect of independent variables on the desirability of a system can be simultaneously investigated by using the desirability function.

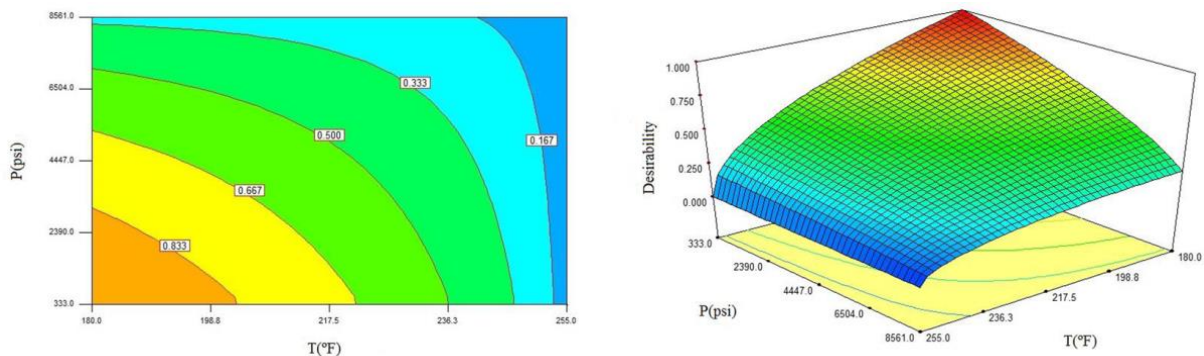
**Table 4.** Representative codes of the input variables in the response surface methodology

Variable	Code
Temperature [°F]	T
Pressure [psi]	P
Injected N <sub>2</sub> [mol%]	N
Injected CO <sub>2</sub> [mol %]	CO
Injected CH <sub>4</sub> [mol%]	CH
Saturation pressure [psi]	SP
Asphaltene [wt%]	AS
Resin [wt%]	RS
API gravity stock tank oil	API
Precipitated asphaltene [wt%]	Wt

The process desirability is the minimization of the amount of asphaltene precipitation with respect to the independent variables. Therefore, the minimum amount of asphaltene precipitation is determined by optimizing the independent parameters.

### Case 1

The first case aims to investigate the effect of pressure, temperature, API gravity of stock tank oil, saturation pressure at reservoir temperature, and weight percentage of resin and asphaltene in the oil on the desirability function. The desirability contour plots that change in the determined range of the experimental data are shown in Figs. 2 to 9. Other variables are held constant at the medium value of the variation interval. The number of effective independent variables is equal to nine; thus, the number of plots that simultaneously investigates the effect of parameters on desirability is equal to 29. The analysis of all plots is not possible in this article. Thus some graphs are investigated and the results of other graphs are discussed. Fig. 2, shows the simultaneous effect of temperature and pressure on desirability. At constant temperature and pressures lower than the saturation pressure, decreasing the pressure will increase oil density and decrease the amount of asphaltene precipitation. The amount of precipitation is decreased with decreased pressure and increased desirability. Increasing temperature increases the amount of precipitation and decreases desirability value. Resin is the stabilizer component of asphaltene in the oil. It avoids precipitation by its absorbance on the asphaltene surface and polar interactions. Thus, increasing the temperature decreases the interaction between asphaltene and resin and increases asphaltene precipitation. The decrease in oil density with an increase in temperature is another reason for this behavior. Thus, the maximum desirability is obtained at the minimum pressure, which is equal to 333 psi, and at the minimum temperature, which is equal to 180 °F.



**Fig. 2.** Surface plots and contour, representing the effects of temperature and pressure on desirability



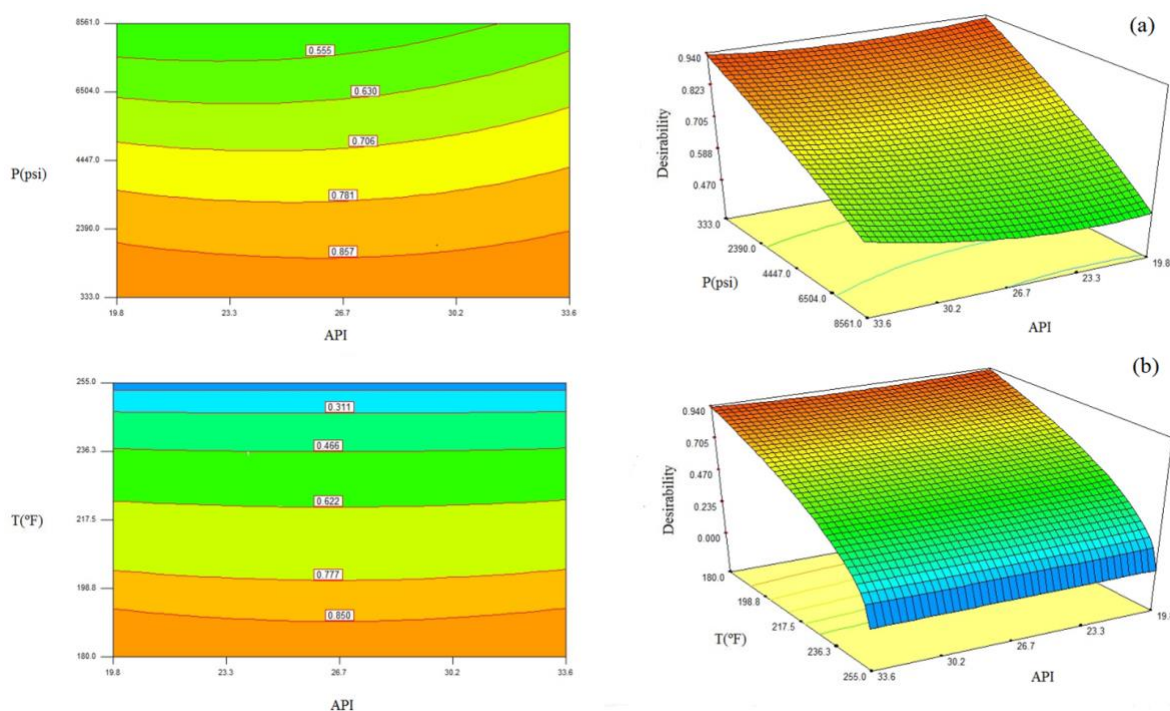
**Table 5.** Interacting coefficients of variables in the second-order polynomial

	T	P	N	CO	CH	SP	AS	RS	API
	-0.390*	3.240*	0.290	0.280*	0.860	0.052*	0.140	-0.320*	0.500*
T	-0.063	-0.170*	0.0770*	-0.045	0.130	-3.463E-3	4.383E-3	6.048E-4*	-0.074E-5
P	-	-0.170	0.400*	0.180	1.150*	-0.043*	0.056*	-0.047*	-9.740E-6
N	-	-	-0.540	4.602E-3	-0.400	-0.060	-0.220	-0.089	+8.185E-4*
CO	-	-	-	0.180	-0.110	9.040E-3	-2.013E-3	-0.025*	-9.638E-05
CH	-	-	-	-	-0.550	9.799E-4	-0.095	0.150*	3.817E-4
SP	-	-	-	-	-	0.120	0.018	2.604E-3	8.184E-8
AS	-	-	-	-	-	-	-0.12	-0.047	-1.385E-3
RS	-	-	-	-	-	-	-	-0.086	-4.535E-4
API	-	-	-	-	-	-	-	-	-9.274E-3

\* Interacting parameters with a significant effect on asphaltene precipitation

**Table 6.** Analysis of variance table for the asphaltene precipitation prediction

Source	Sum of squares	Mean square	Degree of freedom	F-Value	P-Value	Status	R-Squared
Model	7040.87	160.02	44	67.2	<0.0001	Significant	0.8551
Residual	1193	2.38	501	-	-	-	-
Lack of fit	365	1.6	228	-	-	-	-
Pure error	827	3.03	273	0.53	1.00	Insignificant	-
Total	8233.93	-	545	-	-	-	-

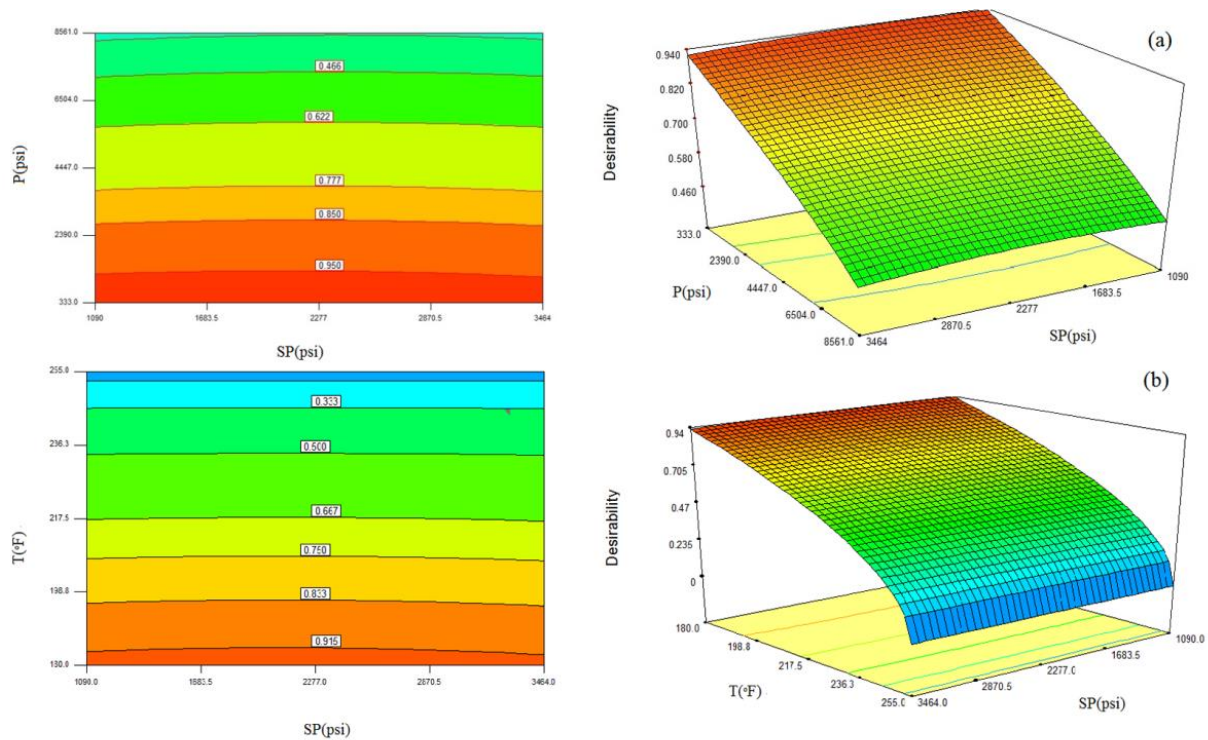


**Fig. 3.** Surface plots and contour, representing the effects of (a) pressure and API gravity and (b) temperature and API gravity on desirability

All plots, which include pressure and another independent variable, have similar behavior to that in Fig. 2. In all these plots, desirability occurs at the minimum pressure. In Fig. 3a, the plot of desirability variations is shown against the API gravity of stock tank oil and pressure. The maximum desirability occurs at the minimum pressure. When API gravity increases, the desirability value decreases at first and then increases. Thus, the maximum desirability is

achieved at API, 33.6 and 19.8. As shown in Fig. 3b, the maximum desirability occurs at API grades of 19.8 and 33.6 and at a temperature of 180 °F.

Fig. 4a depicts the desirability variation according to the oil saturation pressure at reservoir conditions and pressure. Fig. 4b shows the effect of saturation pressure at reservoir conditions and temperature on desirability. Fig. 4 indicates that desirability is much more sensitive to the pressure and temperature than saturation pressure at reservoir conditions.



**Fig. 4.** Surface and contour plots, representing the effects of (a) pressure and saturation pressure and (b) temperature and saturation pressure on desirability

Fig. 5 shows the effect of temperature and resin weight percent in the oil on desirability. As expected, the maximum desirability is achieved at the maximum amount of resin, i.e., 16.8 weight percent. Resin, which is absorbed on the surface of asphaltene, is the stabilizer for asphaltene. Thus, increasing the resin concentration improves the stability of asphaltene and decreases the amount of precipitation. To reach the maximum desirability, the operating pressure should be 333 psi. Fig. 5b indicates that the minimum amount of asphaltene precipitation is obtained at the minimum temperature. The investigation on the effect of asphaltene weight percent and pressure on desirability shows that the asphaltene content of the oil insignificantly affects the precipitation of asphaltene.

Fig. 6 shows the effect of resin and asphaltene weight percent on the desirability. The maximum desirability is achieved at asphaltene content, 0.46 and resin content, 16.8 weight percent. For a specified amount of asphaltene content, the asphaltene stability increases with an increase in the resin content of the oil. For less amount of asphaltene, the available amount of resin for asphaltene stability is increased. Thus, the maximum desirability is obtained at the highest level of resin content to asphaltene content ratio.

## Case 2

In this section, the effect of methane, carbon dioxide, and nitrogen injection on the desirability is investigated. In all plots, other variables are considered constant at the central level of value in their change interval.

Fig. 7 shows the desirability variation as a function of the molar percent of the injected nitrogen and methane gases. The maximum desirability is obtained at zero percent of injected nitrogen and methane. The addition of gas increases its solubility in oil, decreases the solubility parameter of asphaltene, and increases asphaltene precipitation. At high molar percentages of injected gas, the gas is not dissolved in oil; light compounds are transferred from liquid to gas phase. Thus, the liquid enriches with heavy components, its density increases, and asphaltene precipitation reduces. The minimum point in the plot is related to the critical concentration in the p-x phase diagram in the oil and gas injection system.

Fig. 8 shows the effect of injected nitrogen and carbon dioxide to the oil on the desirability. Results indicate that by increasing the mole percent of injected carbon dioxide, desirability initially increases and then decreases. This trend is related to the high density of carbon dioxide at the operating pressure and temperature. With increased mole percent of injected gas, the oil density and oil solubility parameter increase and asphaltene precipitation decreases. Maximum desirability depends on the gas critical composition in the p-x phase diagram. Thus, the maximum desirability is achieved at zero mole percent of injected nitrogen and 10–20 mole percent of injected carbon dioxide.

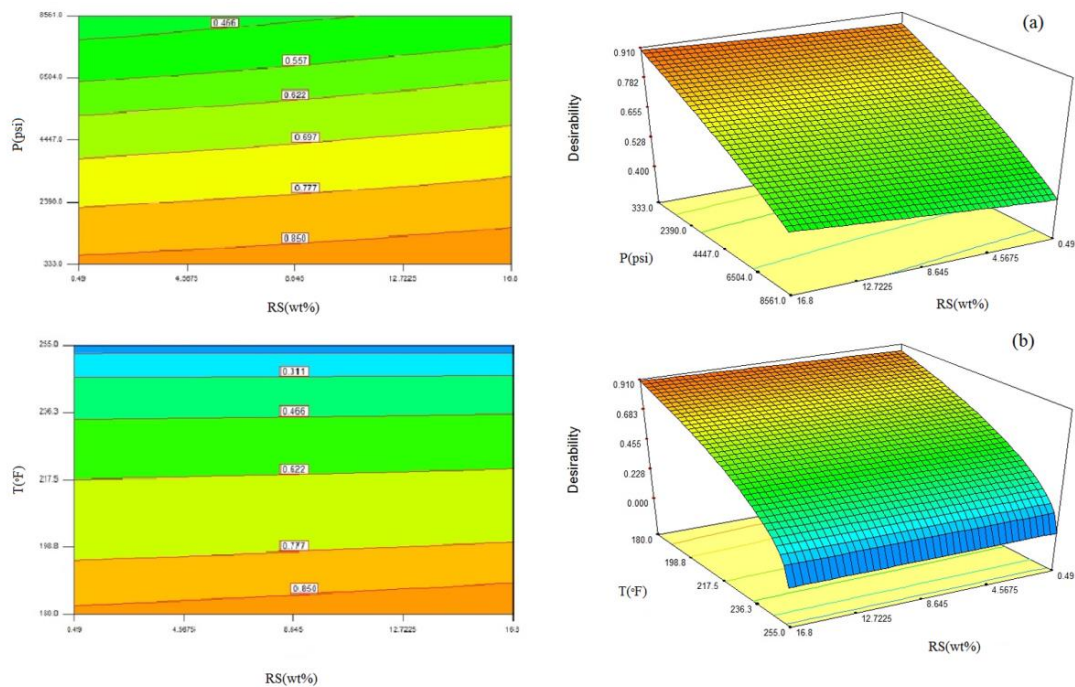


Fig. 5. Three dimensional and contour plots of the effect of (a) pressure and (b) temperature and resin weight percent in the oil on the desirability

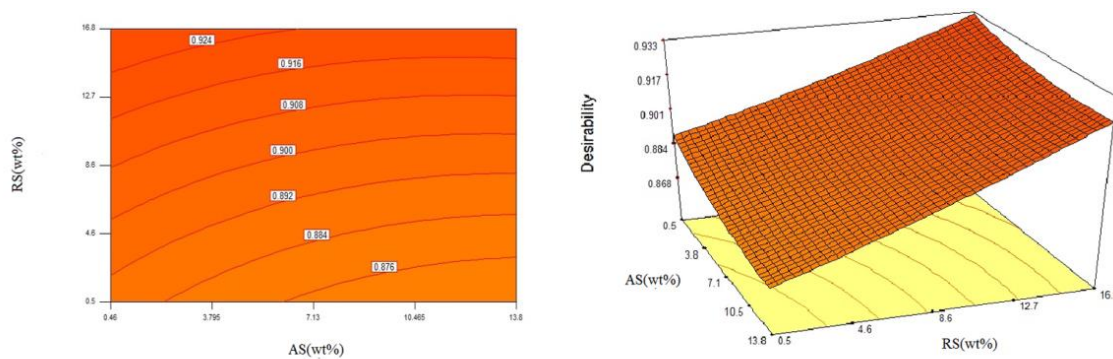


Fig. 6. Three dimensional and contour plots of asphaltene and resin effect on the desirability

Fig. 9 shows the desirability variation as a function of molar percent of the injected carbon dioxide and methane. The desirability change in Fig. 9 is confirmed by the results obtained from Figs. 7 and 8.

Fig. 9 indicates that the maximum desirability is obtained at 10–20 mole percent of injected carbon dioxide without nitrogen injection.

The results of this study indicated that oil characteristics and operating parameters especially, pressure, temperature, and injected gas composition can affect the asphaltene desirability. To determine the effect of live oil composition on the asphaltene desirability, further investigations are required.

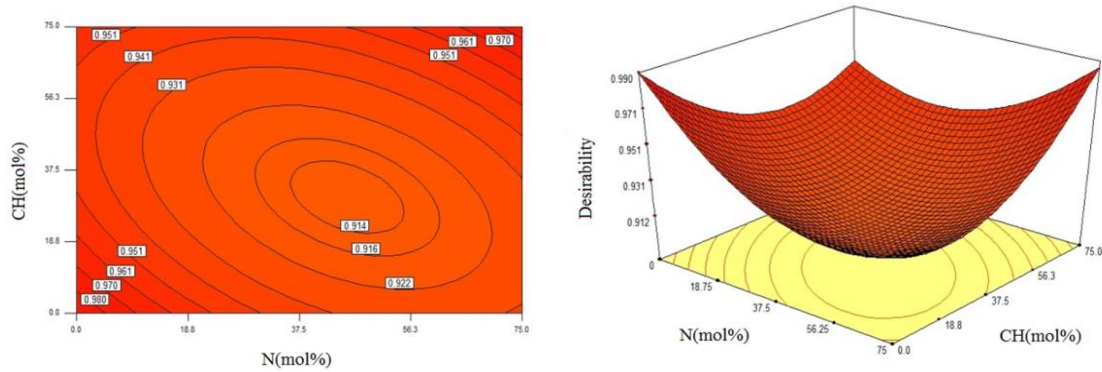


Fig. 7. Three dimensional and contour plots of the effect of nitrogen and methane molar percentage on the desirability

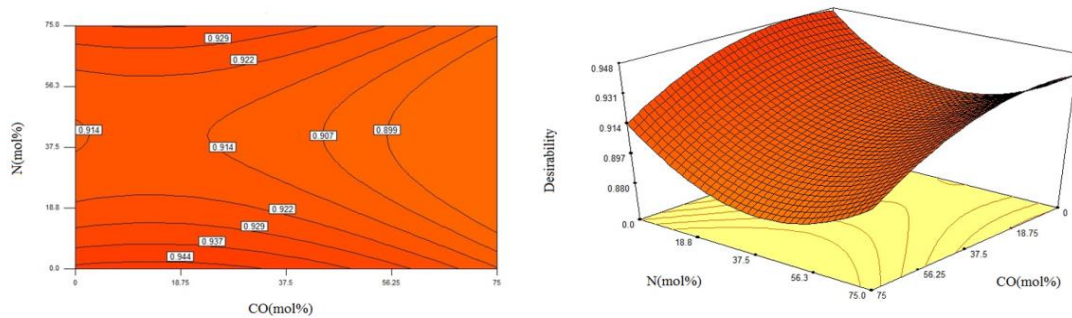


Fig. 8. Three dimensional and contour plots of the effect of nitrogen and carbon dioxide on the desirability

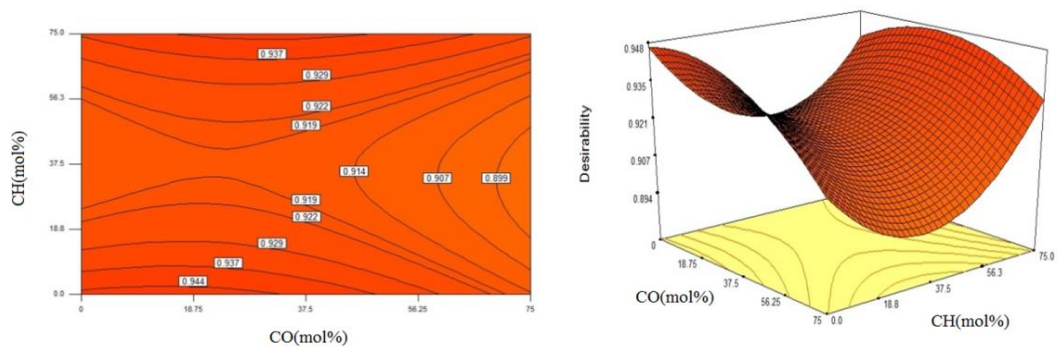


Fig. 9. Three-dimensional and contour plots of the effect of carbon dioxide and methane molar percentage on the desirability

## Conclusion

The simultaneous effects of operative parameters on the asphaltene precipitation are investigated using the combination of artificial neural network and response surface methodology. Batch backpropagation with the structure of 9-7-1 was selected as the optimized algorithm with reasonable prediction accuracy for asphaltene precipitation. The response surface methodology was applied to investigate the simultaneous effects of variables on asphaltene desirability. The maximum desirability was obtained at temperature 180 °F, pressure 333 psi, asphaltene content, 0.46 weight percent, resin content, 16.8 weight percent, and API grades of 19.8 and 33.6. A comparison of variables showed the significant effect of pressure and temperature on asphaltene precipitation. The minimum asphaltene precipitation was achieved at the highest level of resin content to asphaltene content ratio. Results indicated that the maximum desirability depends on the gas critical composition in the p-x phase diagram. Accordingly, the maximum desirability occurred at zero mole percent of injected nitrogen and methane and 10–20 mole percent of injected carbon dioxide. Results showed that the combination of artificial neural network and response surface methodology can appropriately investigate the simultaneous effects of different parameters on asphaltene precipitation and predict the optimum condition to reach minimum asphaltene precipitation.

## References

- [1] Hammami A, Ratulowski J. Precipitation and deposition of asphaltenes in production systems: a flow assurance overview. Mullins OC, Sheu EY, Hammami A, Marshall AG, (eds). *Asphaltenes, Heavy Oils, and Petroleomics*. New York, NY: Springer; 2007.
- [2] Mullins OC, Sabbah H, Eyssautier J, Pomerantz AE, Barré L, Andrews AB, Ruiz-Morales Y, Mostowfi F, McFarlane R, Goual L, Lepkowicz R. Advances in asphaltene science and the Yen–Mullins model. *Energy & Fuels*. 2012 May 8;26(7):3986-4003.
- [3] Hosseini-Dastgerdi Z, Meshkat SS. An experimental and modeling study of asphaltene adsorption by carbon nanotubes from model oil solution. *Journal of Petroleum Science and Engineering*. 2019 Mar 1;174:1053-61.
- [4] Dastgerdi ZH, Meshkat SS, Hosseinzadeh S, Esrafil MD. Application of Novel Fe<sub>3</sub>O<sub>4</sub>–Polyaniline Nanocomposites in Asphaltene Adsorptive Removal: Equilibrium, Kinetic Study and DFT Calculations. *Journal of Inorganic and Organometallic Polymers and Materials*. 2019 Jul 1;29(4):1160-70.
- [5] Hosseini-Dastgerdi Z, Tabatabaei-Nejad SA, Khodapanah E, Sahraei E. A comprehensive study on mechanism of formation and techniques to diagnose asphaltene structure; molecular and aggregates: a review. *Asia-Pacific Journal of Chemical Engineering*. 2015 Jan;10(1):1-14.
- [6] Hosseini-Dastgerdi Z, Tabatabaei-Nejad SA, Sahraei E, Nowroozi H. Morphology and size distribution characterization of precipitated asphaltene from live oil during pressure depletion. *Journal of Dispersion Science and Technology*. 2015 Mar 4;36(3):363-8.
- [7] Akbarzadeh K, Hammami A, Kharrat A, Zhang D, Allenson S, Creek J, Kabir S, Jamaluddin A, Marshall AG, Rodgers RP, Mullins OC. Asphaltenes—problematic but rich in potential. *Oilfield Review*. 2007 Jul;19(2):22-43.
- [8] Tavakkoli M, Panuganti SR, Taghikhani V, Pishvaie MR, Chapman WG. Precipitated asphaltene amount at high-pressure and high-temperature conditions. *Energy & Fuels*. 2013 Nov 20;28(3):1596-610.
- [9] Zanganeh P, Ayatollahi S, Alamdari A, Zolghadr A, Dashti H, Kord S. Asphaltene deposition during CO<sub>2</sub> injection and pressure depletion: a visual study. *Energy & Fuels*. 2012 Jan 13;26(2):1412-9.
- [10] Syunyaev RZ, Likhatsky VV. Effects of temperature and pressure on the phase state of oils and asphaltene solutions observed using dielectric spectroscopy. *Energy & Fuels*. 2010 Jan 20;24(4):2233-9.

- [11] AlHammadi AA, Chen Y, Yen A, Wang J, Creek JL, Vargas FM, Chapman WG. Effect of the gas composition and gas/oil ratio on asphaltene deposition. *Energy & Fuels*. 2017 Mar 10;31(4):3610-9.
- [12] Zanganeh P, Dashti H, Ayatollahi S. Visual investigation and modeling of asphaltene precipitation and deposition during CO<sub>2</sub> miscible injection into oil reservoirs. *Fuel*. 2015 Nov 15;160:132-9.
- [13] Wang P, Zhao F, Hou J, Lu G, Zhang M, Wang Z. Comparative Analysis of CO<sub>2</sub>, N<sub>2</sub>, and Gas Mixture Injection on Asphaltene Deposition Pressure in Reservoir Conditions. *Energies*. 2018 Sep;11(9):2483-97
- [14] Mullins OC. The modified Yen model. *Energy & Fuels*. 2010 Jan 19;24(4):2179-207.
- [15] Anisimov MA, Ganeeva YM, Gorodetskii EE, Deshabo VA, Kosov VI, Kuryakov VN, Yudin DI, Yudin IK. Effects of resins on aggregation and stability of asphaltenes. *Energy & Fuels*. 2014 Sep 24;28(10):6200-9.
- [16] Li Z, Firoozabadi A. Cubic-plus-association equation of state for asphaltene precipitation in live oils. *Energy & fuels*. 2010 Apr 5;24(5):2956-63.
- [17] Dehaghani YH, Assareh M, Feyzi F. Asphaltene precipitation modeling with PR and PC-SAFT equations of state based on normal alkanes titration data in a Multisolid approach. *Fluid Phase Equilibria*. 2018 Aug 25;470:212-20.
- [18] Merino-Garcia D, Corraera S. A shortcut application of a Flory-like model to Asphaltene precipitation. *Journal of Dispersion Science and Technology*. 2007 Mar 1;28(3):339-47.
- [19] Manshad AK, Edalat M. Application of continuous polydisperse molecular thermodynamics for modeling asphaltene precipitation in crude oil systems. *Energy & Fuels*. 2008 May 22;22(4):2678-86.
- [20] Arya A, Liang X, Von Solms N, Kontogeorgis GM. Modeling of asphaltene onset precipitation conditions with cubic plus association (CPA) and perturbed chain statistical associating fluid theory (PC-SAFT) equations of state. *Energy & Fuels*. 2016 Jul 26;30(8):6835-52.
- [21] Gharagheizi F, Eslamimanesh A, Sattari M, Mohammadi AH, Richon D. Corresponding states method for evaluation of the solubility parameters of chemical compounds. *Industrial & Engineering Chemistry Research*. 2012 Feb 22;51(9):3826-31.
- [22] Pazuki GR, Nikookar M. A modified Flory-Huggins model for prediction of asphaltenes precipitation in crude oil. *Fuel*. 2006 May 1;85(7-8):1083-6.
- [23] Yazdizadeh M, Nourbakhsh H, Jafari Nasr MR. A Solution Model for Predicting Asphaltene Precipitation. *Iranian Journal of Chemistry and Chemical Engineering (IJCCE)*. 2014 Mar 1;33(1):93-102.
- [24] Ahmadi M, Jafarzadeh-Ghoushchi S, Taghizadeh R, Sharifi A. Presentation of a new hybrid approach for forecasting economic growth using artificial intelligence approaches. *Neural Computing and Applications*.:1-20.
- [25] Salem S, Jafarzadeh-Ghoushchi S. Estimation of optimal physico-chemical characteristics of nano-sized inorganic blue pigment by combined artificial neural network and response surface methodology. *Chemometrics and Intelligent Laboratory Systems*. 2016 Dec 15;159:80-8.
- [26] Jafarzadeh-Ghoushchi S, Rahman MN. Performance study of artificial neural network modelling to predict carried weight in the transportation system. *International Journal of Logistics Systems and Management*. 2016;24(2):200-12.
- [27] Al Ameen A, Mondal S, Pudi SM, Pandhare NN, Biswas P. Liquid phase hydrogenolysis of glycerol over highly active 50% Cu-Zn (8: 2)/MgO catalyst: reaction parameter optimization by using response surface methodology. *Energy & Fuels*. 2017;31(8):8521-33.
- [28] Ahmadi MA, Golshadi M. Neural network based swarm concept for prediction asphaltene precipitation due to natural depletion. *Journal of Petroleum Science and Engineering*. 2012 Nov 1;98:40-9.
- [29] Abedini A, Ashoori S, Torabi F, Saki Y, Dinarvand N. Mechanism of the reversibility of asphaltene precipitation in crude oil. *Journal of Petroleum Science and Engineering*. 2011 Aug 1;78(2):316-20.
- [30] Khomehchi E, Behvandi R. Prediction of Bubble Point Pressure & Asphaltene Onset Pressure During CO<sub>2</sub> Injection Using ANN & ANFIS Models. *Journal of Petroleum Science and Technology*. 2011 Aug 10;1(2):35-45.

- [31] Ashoori S, Abedini A, Abedini R, Nasheghi KQ. Comparison of scaling equation with neural network model for prediction of asphaltene precipitation. *Journal of Petroleum Science and Engineering*. 2010 May 1;72(1-2):186-94.
- [32] Tavakkoli M, Kharrat R, Masihi M, Ghazanfari MH, Fadaei S. Phase behavior modeling of asphaltene precipitation for heavy crudes: a promising tool along with experimental data. *International Journal of Thermophysics*. 2012 Dec 1;33(12):2251-66.
- [33] Moradi S, Rashtchian D, Ganjeh Ghazvini M, Emadi MA, Dabir B. Experimental investigation and modeling of asphaltene precipitation due to gas injection. *Iranian Journal of Chemistry and Chemical Engineering (IJCCE)*. 2012 Mar 1;31(1):89-98.
- [34] Nakhli H, Alizadeh A, Moqadam MS, Afshari S, Kharrat R, Ghazanfari MH. Monitoring of asphaltene precipitation: Experimental and modeling study. *Journal of Petroleum Science and Engineering*. 2011 Aug 1;78(2):384-95.
- [35] Afshari S, Kharrat R, Ghazanfari MH. Asphaltene precipitation study during natural depletion at reservoir conditions. In *International Oil and Gas Conference and Exhibition in China 2010* Jan 1. Society of Petroleum Engineers.
- [36] Moradi S, Dabiri M, Dabir B, Rashtchian D, Emadi MA. Investigation of asphaltene precipitation in miscible gas injection processes: experimental study and modeling. *Brazilian Journal of Chemical Engineering*. 2012 Sep;29(3):665-76.
- [37] Kord S, Ayatollahi S. Asphaltene precipitation in live crude oil during natural depletion: experimental investigation and modeling. *Fluid Phase Equilibria*. 2012 Dec 25;336:63-70.
- [38] Alizadeh A, Nakhli H, Kharrat R, Ghazanfari MH, Aghajani M. Experimental study of asphaltene precipitation behavior during miscible carbon dioxide injection. *Energy Sources, Part A: Recovery, Utilization, and Environmental Effects*. 2014 Jul 18;36(14):1523-30.
- [39] Bahrami P, Kharrat R, Mahdavi S, Firoozinia H. Prediction of the gas injection effect on the asphaltene phase envelope. *Oil & Gas Science and Technology—Revue d'IFP Energies nouvelles*. 2015 Nov 1;70(6):1075-86.
- [40] Alizadeh A, Nakhli H, Kharrat R, Ghazanfari MH. An experimental investigation of asphaltene precipitation during natural production of heavy and light oil reservoirs: The role of pressure and temperature. *Petroleum Science and Technology*. 2011 Mar 16;29(10):1054-65.
- [41] Jafarzadeh SG, Rahman MN, Wahab DA. Forecasting capabilities of spare part production with artificial neural networks model in a supply chain. *World Applied Sciences Journal*. 2012;20(5):674-8.
- [42] Jafarzadeh SG, Rahman MN, Wahab DA. Optimization of supply chain management based on response surface methodology: A case study of iran khodro. *World Applied Sciences Journal*. 2012;20(4):620-7.
- [43] Ahmadi MA. Prediction of asphaltene precipitation using artificial neural network optimized by imperialist competitive algorithm. *Journal of Petroleum Exploration and Production Technology*. 2011 Dec 1;1(2-4):99-106.
- [44] Khayet M, Cojocar C, García-Payo C. Application of response surface methodology and experimental design in direct contact membrane distillation. *Industrial & engineering chemistry research*. 2007 Aug 15;46(17):5673-85.
- [45] Mohammadi M, Dadvar M, Dabir B. Application of response surface methodology for optimization of the stability of asphaltene particles in crude oil by TiO<sub>2</sub>/SiO<sub>2</sub> nanofluids under static and dynamic conditions. *Journal of Dispersion Science and Technology*. 2018 Mar 4;39(3):431-42.



This article is an open-access article distributed under the terms and conditions of the Creative Commons Attribution (CC-BY) license.