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Asphaltene precipitation modeling through ACE reaping of scaling equations

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Precipitation and deposition of asphaltene have undesirable effects on the petroleum industry by increasing operational costs due to reduction of well productivity as well as catalyst poisoning. Herein we propose a reliable model for quantitative estimation of asphaltene precipitation. Scaling equation is the most powerful and popular model for accurate prediction of asphaltene precipitated out of solution in crudes without regard to complex nature of asphaltene. We employed a new mathematical-based approach known as alternating conditional expectation (ACE) technique for combining results of different scaling models in order to increase the accuracy of final estimation. Outputs of three well-known scaling equations, including Rassamdana (RE), Hu (HU), and Ashoori (AS), are input to ACE and the final output is produced through a nonlinear combination of scaling equations. The proposed methodology is capable of significantly increasing the precision of final estimation via a divide-and-conquer principle in which ACE functions as the combiner. Results indicate the superiority of the proposed method compared with other individual scaling equation models.

asphaltene precipitation, crude oil, alternating conditional expectation, scaling equation, prediction

1 Introduction

Asphaltene is recognized as the heaviest and the most polar component of petroleum fluid [1]. From the operational perspective, it is defined as the portion of petroleum mixture that is soluble in toluene but insoluble in n-heptane [2]. Elemental composition analysis of asphaltene includes heteroatoms (e.g., sulfur, oxygen, and nitrogen) as well as metal constituents (e.g., nickel, iron, and vanadium) in its molecular structure [3]. In the initial reservoir condition, asphaltene is kept dispersed in the petroleum medium through peptizing by resin [4]. Owing to the sensitivity of the phase stability of asphaltene to thermodynamic parameters, changes in pressure, temperature, and crude oil composition can cause desorption of resin from asphaltene. This phenomenon results in phase separation and deposition of asphaltene in the form of solid particles in different stages of the oil industry [5] and can have undesirable impacts on the petroleum industry both in downstream and upstream operations. In upstream operation, asphaltene precipitate mainly occurs due to pressure drop during natural depletion as well as composition alteration during enhanced oil recovery processes such as carbon dioxide, nitrogen, or methane injection [6–11]. In oil reservoirs, negative effects are observed on the efficiency of enhanced oil recovery processes, largely because of the mechanisms of wettability alteration and pore throat blockage [12, 13]. In downstream operations, precipitation and deposition of asphaltene cause clogging of transportation pipelines as well as loss of efficiency in production facilities, heat exchangers, and catalysts [13, 14].

Motivated by these numerous adverse impacts, which also affect reservoir characteristics and refining equipment,

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researchers have sought to shed light the mechanism of asphaltene precipitation and to develop powerful models to estimate where and when such problems might occur in different stages of the petroleum industry [15–22]. However, none of the predictive models developed so far are capable of achieving a comprehensive interpretation of this phenomenon, mainly because of the complex nature and diverse array of parameters that affect asphaltene precipitation [18, 21]. Mathematical approaches that facilitate estimations of both the precipitated asphaltene amount and its threshold value, in which asphaltene begins phase-separation from petroleum mixture, are divided into three distinct groups. The first is molecular thermodynamic models, in which asphaltene is dissolved in crude oil and crude oil forms real solution [23]. The second is colloidal approach, which is based on the assumption that asphaltene is stable in crude oil, as suspension, through peptizing by resin [24]. The third is scaling equations, in which a quantitative formulation between the amount of asphaltene precipitation and titration data is constructed regardless of the complex nature of asphaltene and its agglomeration [25]. Considering the costly problems associated with asphaltene precipitation and the limitations of previous models for highly accurate prediction of asphaltene precipitation, it is essential to propose a novel and potent model for quantitative estimation of asphaltene precipitation. In the literature, three different scaling equation models are proposed for making quantitative formulation between titration data and amount of asphaltene precipitation. In the current study, alternating conditional expectation is developed as an accurate, robust, and rapid mathematical model for combining the results of different scaling models to increase the accuracy of final predictions, using "a divide-and-conquer principle". For this purpose, outputs of individual models including Rassamdana *et al.* scaling (RE) [26], Hu and Guo scaling (HU) [27], and Ashoori *et al*. scaling (AS) [28] are employed as inputs of an alternating conditional expectation (ACE) model. ACE acts as a combiner in the proposed method in order to transform both real output and scaling equations independently such that they have higher correlation, followed by a regression between the transformed inputs and transform output [29]. The results of our model are compared with individual scaling equation models based on statistical criteria such as correlation coefficient, mean square error, average relative error, and absolute average relative error. It is observed that, compared to individual scaling models, an ACE model can predict the amount of asphaltene precipitation with higher satisfactory accuracy.

2 Background theories

2.1 Scaling equation model

In 1996, Rassamdana and co-workers conducted a series of titration experiment to describe the behavior of asphaltene precipitation [25]. First, they applied a thermodynamic model based on the Flory-Huggins theory of polymer solution to devise a quantitative formulation between titration data and amount of asphaltene precipitation. They observed the traditional thermodynamic model produced results did not display acceptable agreement with corresponding experimental data. Therefore they developed a novel model, the so-called scaling equation, as an appropriate alternative to quantitative estimation of asphaltene precipitation. This model, which was premised on the assumption that the formation of asphaltene structure is to some extent similar to aggregation and gelation phenomena, achieved popularity because of its simplicity and excellent performance for predicting the amount of asphaltene under different conditions. These researchers combined three variables (the amount of precipitated asphaltene W_t , the solvent to oil dilution ratio R_v , and the molecular weight of solvent M_w) into to the following equation:

$$X = \frac{R_v}{M_w^Z} \tag{1}$$

$$Y = \frac{W_t}{R_v^{Z'}} \tag{2}$$

where Z and Z' are adjustable parameters. In order to achieve the best correlation between modeling results and corresponding experimental values, the values in the adjustable parameters must be selected with high precision.

These researchers also proposed the scaling equation as a three-order polynomial equation in term of X and Y:

$$Y = A_1 + A_2 X + A_3 X^2 + A_4 X^3,\ X > X_C \tag{3}$$

where (A_i, i = 1–4) is the scaling coefficient and X_C is the value of X at the threshold where asphaltene starts to phase separation from crude oil.

Hu *et al.* evaluated the accuracy and capability of the Rassamdana *et al*. scaling equation by applying it to experimental data collected from open literature sources [30]. They concluded that this scaling equation is an attractive tool that can predict the amount of asphaltene precipitation with high accuracy. Although the aforementioned scaling approaches were useful, they possessed some shortcomings. One of the defects of scaling approaches is that they can predict the amount of asphaltene precipitation in constant temperature. However, because temperature is variable within the scaling equation, it is not capable of predicting the amount of asphaltene precipitation in various temperatures. In attempts to overcome flaw, investigators such as Rassamdana *et al*. [26], Hu and Guo [27], and Ashoori *et al*. [28] tried to modify the previous scaling equation by inserting a temperature parameter into its formulation. For detailed studies of the formulation of previous scaling models, readers are referred to the original publications.

2.2 Alternating conditional expectations

The ACE is an advanced statistical technique that was proposed by Breiman and Friedman in 1985 [29]. This method is an appropriate alternative for solving the regression problems that contain unidentified relationships between predictor and response variables [31, 32]. This technique has garnered much attention for solving regression estimation problems of the petroleum industry, including permeability, minimum miscible pressure, and PVT properties estimation [33–35]. The popularity of this technique was attributed to its excellent performance for appropriately demonstrating a nonlinear relationship, if any, between variables in regression problems. Results produced by this technique achieve best fit with corresponding real values though approximating the optimal transformations for the dependent and independent variables [29]. Generally, a linear regression model for p independent variables, $X_1, X_2, \ldots, X_p$ and a response variable L are given as:

$$L = \beta_0 + \sum_{i=1}^{p} \beta_i X_i + \varepsilon \tag{4}$$

where (β_i, i = 0–p) are the regression coefficients to be estimated and ε is an error term. In ACE, in lieu of assessing the correlation between L and $X_1, X_2, \ldots, X_p$ which is conducted in conventional regression analysis, the relationship between $\theta(L)$ and $\phi_1(X_1)$, $\cdots$, $\phi_p(X_p)$, is computed. Based on Equation 8, the general form of non-parametric ACE algorithm is defined as [29]:

$$\theta(L) = \alpha + \sum_{i=1}^{p} \phi(X_i) + \varepsilon \tag{5}$$

where $\theta(L)$, $\phi_1(X_1)$, $\cdots$, $\phi_p(X_p)$ are the arbitrary measurable mean-zero functions of $L, X_1, X_2, \cdots, X_p$ respectively. The main purpose of ACE is to seek the optimal transformation $\phi_i^*(X_i)$, $i = 1, \ldots, p$ and $\theta^*(L)$, which lead to the maximum correlation between the transformed dependent variable and the sum of transformed predicted variables. This determination is equivalent to minimizing the value of the error variance (ε^2). In turn, the value of the error variance (ε^2) of a linear regression of the transformed dependent variable on the sum of the transformed independent variables (under the constraint, $E[\theta^2(L)]$=1) is given by the following equation [29]:

$$\varepsilon^2(\theta, \phi_i, \ \ldots, \ \phi_p) = E\left([\theta(L) - \sum_{i=1}^{p} \phi(X_i)] \right)^2 \Big/ E\theta^2(L) \tag{6}$$

By implementing minimization of the value of ε^2 with respect to $\theta(L)$ and $f_k(X_k)(i = 1,2,...,k)$ with a series of single-function minimizations, the following equations for response variables and predictor variables, respectively, are [26]:

$$\theta(L) = E\left[\sum_{i=1}^{p} \phi(X_i) \mid L \right] \Big/ \left\| E\left[\sum_{i=1}^{p} \phi(X_i) \mid L \right] \right\| \tag{7}$$

$$\phi_{j,1}(X_j) = E\left[\theta(L) - \sum_{i=1}^{p} \phi_i(X_i) \mid X_k \right] \tag{8}$$

Having implemented the iteration process of minimizing ε^2, the real-valued measurable zero-mean functions $\phi_i(X_i)$, $i = 1, \ldots, p$ and $\theta(L)$ are determined; here, these value are equal to the values of optimal transformation $\phi_i^*(X_i)$, $i = 1, \ldots, p$ and $\theta^*(L)$. In the transformed space, the response and predictor variables are related as [29]:

$$\theta^*(L) = \sum_{i=1}^{p} \phi_i^*(X_i) + e^* \tag{9}$$

where e^* is the error, not captured by the use of the ACE transformations and is assumed to have a normal distribution with zero mean [29].

3 Input/output data space

Data sets used in this study were collated from open-sources literature [28]. In the scaling equations, three variables (solvent-to-oil dilution ratio, temperature, and molecular weight of solvent) are used as inputs for the quantitative determination of asphaltene precipitation. Since 1990s, several experimental efforts have been implemented to investigate the temperature dependence of asphaltene precipitation [26]. It has reported that increasing the temperature induces two opposite effects in the amount of the precipitated asphaltene. In the data sets employed in our study, the amount of precipitated asphaltene reduced with temperature increase. Increasing the molecular weight of solvent causes less asphaltene precipitate from crude oil solution [27], and the amounts of asphaltene precipitation increase as the dilution ratio increases [28]. These data were input to three well-known scaling equations, including the models of Rassamdana *et al*. (RE) [26], Hu and Guo (HU) [27], and Ashoori *et al*. (AS) [28]. Outputs from these scaling equations were then used as inputs of alternating conditional expectations. The amount of asphaltene precipitation in the resultant data sets was measured with the gravity method. Table 1 includes statistics of the data used for modeling in the current study.

4 Results and discussion

Figure 1 depicts the general flowchart of current work. At

Table 1 Statistical description of data set used for developing predictive models

Parameter	Min	Max	Mean
Dilution ratio (mL/g)	0.670	20.000	7.617
Temperature (K)	303.150	343.150	323.150
Molecular weight of solvent (g/mol)	72.150	100.210	86.170
Amount of asphaltene precipitation (wt%)	0.500	10.400	4.785

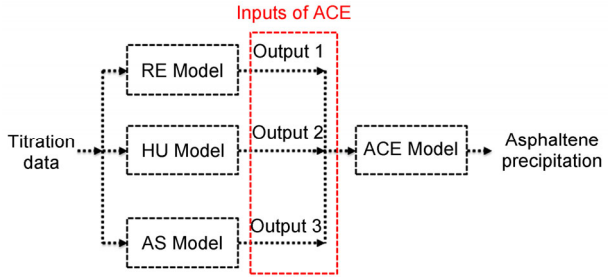

Figure 1 Schematic diagram of ACE-based model designed in this study. Results of three scaling approaches are employed as inputs of ACE models. ACE approximates the optimal transformations of input/output data so that correlation between the transformed dependent (asphaltene precipitation) and sum of the transformed independent variables (results of scaling models) is maximized. Indeed, ACE acts as nonlinear combiner to determining contribution of different scaling models in final prediction.

the first stage, three scaling models including Rassamdana *et al*. [26], Hu and Guo [27], and Ashoori *et al*. [28] are used to estimate the amount of asphaltene precipitation from titration data (i.e., temperature, dilution ratio, and molecular weight of solvent). Then, the ACE method is employed to improve the accuracy of final predictions by combining the results of each scaling equation. To achieve best fit between the modeling results and corresponding input values, ACE transforms the input and output data to a specific space. Figure 2(a–d) demonstrates the optimal transformations for the Rassamdana *et al*. (RE), Hu and Guo (HU), Ashoori *et al*. (AS) methods and the amount of asphaltene precipitation, all computed with the ACE algorithm. These figures demonstrate the qualitative impact of the input variables (RE, HU, and AS model results) on the amount of asphaltene precipitation. Figure 3 shows the optimal transformation of W_t versus the sum of optimal transformations of the RE, HU, and AS results. Figure 4 depicts a cross-plot between the measured and estimated asphaltene precipitation for the ACE-based model and the individual scaling models. Higher values of correlation coefficients between the estimated and experimental amounts prove the superiority of alternating conditional expectation methods in the quantitative estimation of asphaltene precipitation. In order to obtain a computational model for making quantitative formulations between the titration data and the amounts of asphaltene precipitation, the transformed data is related to the actual data through a simple polynomial. The polynomials for RE, HU, and AS models are as follows:

$$\phi^*(\mathrm{RE}) = a_1(\mathrm{RE}) + a_0 \tag{10}$$

where $a_1 = -0.471906$ and $a_0 = 2.234916$.

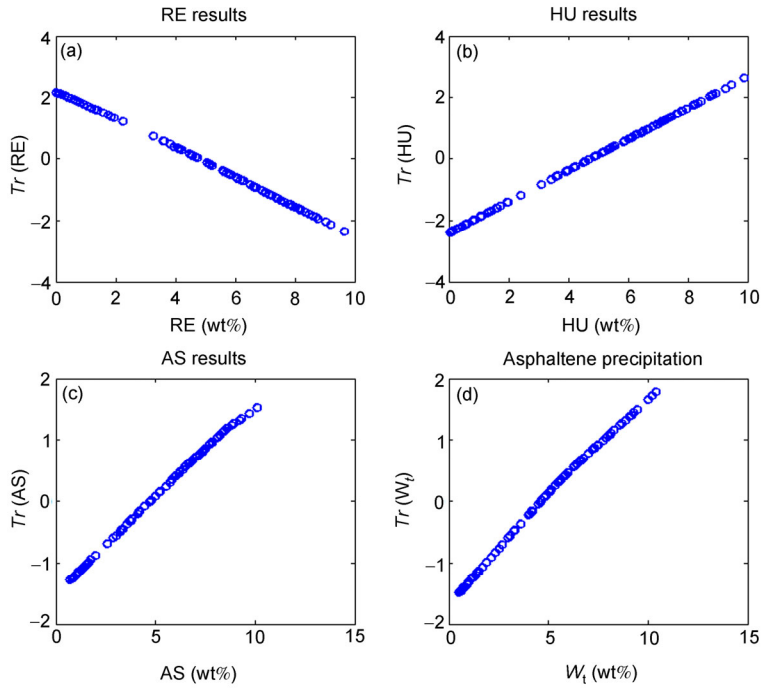

Figure 2 Optimal transformations of (a) RE model, (b) HU model, (c) AS model, and (d) amount of asphaltene precipitation which computed by ACE.

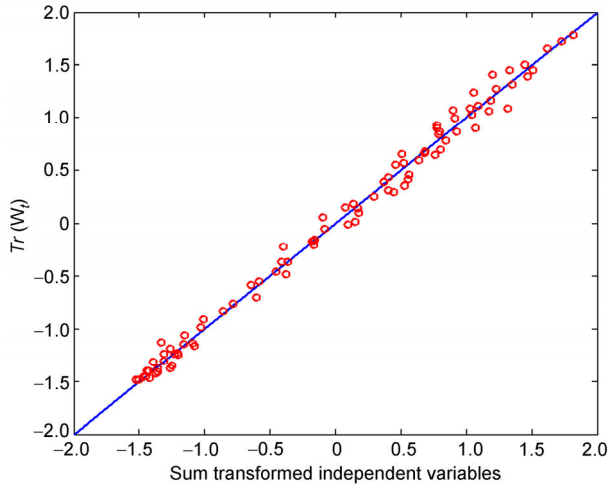

Figure 3 Optimal transformation of W_t vs. the sum of the optimal transformations of RE, HU, and AS models.

$$\phi^*(\mathrm{HU}) = a_1(\mathrm{HU}) + a_0 \tag{11}$$

where $a_1 = -0.508423$ and $a_0 = 2.414638$.

$$\phi^*(\mathrm{AS}) = a_4(\mathrm{AS})^4 + a_3(\mathrm{AS})^3 + a_2(\mathrm{AS})^2 + a_1(\mathrm{AS}) + a_0 \tag{12}$$

where $a_4 = -0.000150$, $a_3 = 0.002422$, $a_2 = -0.013705$, $a_1 = 0.350145$, and $a_1 = -1.534589$.

The linear regression between these dependent and independent variables is given as:

$$\theta^*(W_t) = \phi^*(\mathrm{RE}) + \phi^*(\mathrm{HU}) + \phi^*(\mathrm{AS}) \tag{13}$$

The value of the amount of asphaltene precipitation is determined by employing the following polynomial correlation:

$$W_t = a_2(\theta^*(W_t))^2 + a_1\ (\theta^*(W_t)) + a_0 \tag{14}$$

where $a_2 = 0.166994$, $a_1 = 2.969975$, and $a_0 = 4.620931$.

Table 2 illustrates correlation coefficient (R^2), mean square error (MSE), average relative error (ARE), and absolute average relative error (AARE) values for ACE model. Figure 5 contains a comparison between measured and predicted asphaltene amount versus different samples. This figure shows that there is good agreement between the measured and predicted values using the proposed methodology. According to Figures 4 and 5 and Table 2, ACE has a satisfying performance in predicting amount of asphaltene precipitation.

5 Conclusions

Precipitation and deposition of asphaltene are among the most drastic issues in the oil industry; as such, they can have negative impacts on downstream and upstream opera

Table 2 Comparison of ACE-base model and different scaling models versus R^2, MSE, ARE and AARE

Parameter	RE model	HU model	AS model	ACE model
R^2	0.9572	0.9632	0.9837	0.9912
MSE	0.3778	0.3237	0.1543	0.0768
ARE	0.0722	0.0622	–0.0544	–0.0044
AARE	0.1791	0.1733	0.1089	0.0676

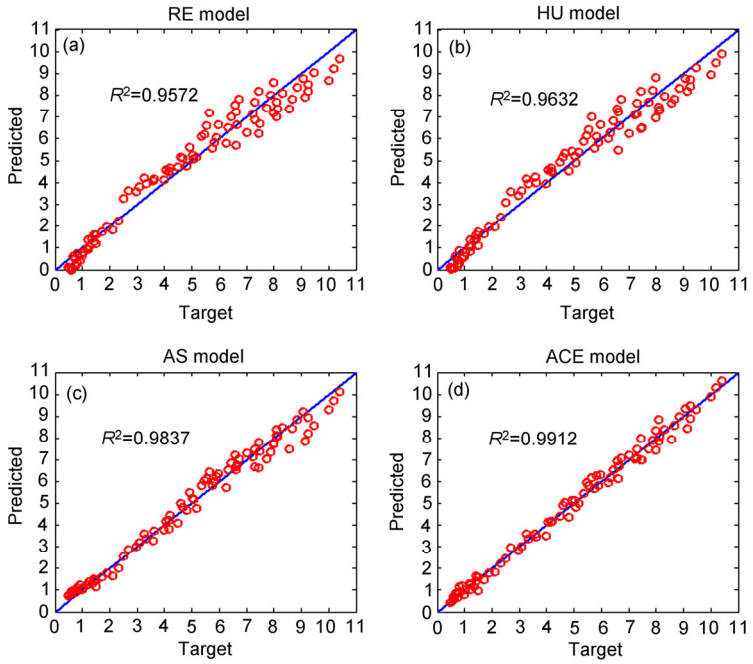

Figure 4 Cross-plots illustrating the correlation coefficient between measured and predicted asphaltene precipitation amounts using (a) RE model, (b) HU model, (c) AS model, and (d) ACE based model.

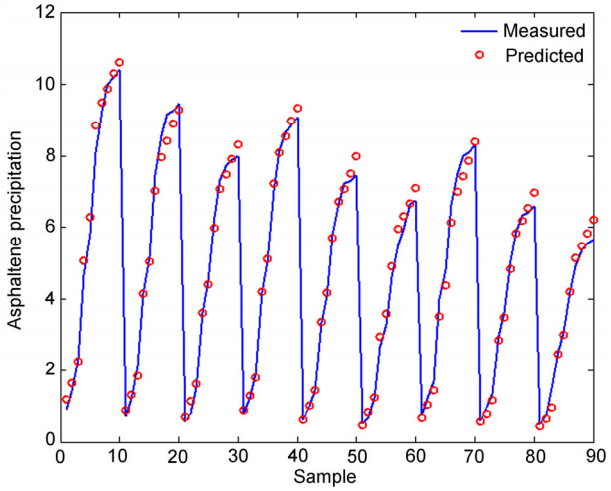

Figure 5 A comparison between measured asphaltene amount and estimated values by ACE based model.

tions. In the current study, a novel method- the so-called alternating conditional method or ACE-is employed as a mathematical approach for combining the results of different scaling models and thereby achieving final results with more accuracy. Outputs of different scaling models are used as inputs for the ACE-based model. ACE produced satisfactory results by performing optimal transformation to inputs and outputs independently, such that the transformed input data would have higher correlations with the transformed output data. Results of this study show that with little additional computation, it is possible to increase the accuracy of final asphaltene modeling through ACE combinations of scaling equations. Evaluation of efficacy of the ACE-based model compared with individual scaling equations versus correlation coefficients and statistical errors proved the superiority of our proposed methodology. Finally, the proposed model provides a reliable alternative for the quantitative estimation of asphaltene precipitation amounts in an accurate, cost-effective, and rapid way. If the mathematics in the results and discussion sections are utilized, accurate estimations can be made. We believe that if more sophisticated combiner would be found, it would be possible to enhance the accuracy of final predictions even further.

Nomenclature

AARE	Absolute average relative error (%)
a_1, a_2, a_3, a_4	ACE polynomial correlation
Z, Z',C_1, C_2	Adjustable parameters for scaling equation
ACE	Alternating conditional expectation
W_t	Amount of asphaltene precipitation (weight percent)
AS	Ashoori *et al*. scaling equation
ARE	Average relative error (%)
R^2	Correlation coefficient
E	Error term in Eq.(5)
e^*	Error not captured with ACE transformation
X	Function defined by Eq.(1)
Y	Function defined by Eq.(2)
HU	Hu and Guo scaling equation
X_i, $i = 1–p$	Independent variable
MSE	Mean square error
M_w	Molecular weight of solvent (g/mol)
ϕ^*	Optimal transformation of independent variable
θ^*	Optimal transformation of response variable
RE	Rassamdana . scaling equation
β_i, $i = 0–p$	Regression coefficient in Eq. (4)
L	Response variable
A_1, A_2, A_3, A_4	Scaling equation coefficient in Eq. (3)
R_v	Solvent-to-oil dilution ratio (mL/g)
T	Temperature (K)
Φ	Transformation of independent variable
θ	Transformation of response variable
X_C	Value of X on onset of precipitation

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