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RIGOROUS MODEL DEVELOPMENT FOR ESTIMATING ASPHALTENE PRECIPITATION FROM CRUDE OIL BY N-ALKANE TITRATION

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Abstract

Precipitation of asphaltene, which is a complex hydrocarbon mixture, causes serious problems during petroleum production. Some problems associated with precipitation of asphaltene includes reduced oil recovery and relative permeabilities, blockage of rock pores, and reduction of flow rate. There is a number of issues (cost, time, and difficulty and accuracy of measurements) with regards to laboratory measurements and therefore it is important to develop reliable models to predict the phase behaviour of asphaltene precipitation. In this study, accurate models were developed using gene expression programming (GEP), decision tree (DT), and least squares support vector machine (LSSVM) methodologies for the determination of asphaltene precipitation from Iranian crude oil. To this end, molecular weight of precipitant (n-alkane), temperature, and ratio of precipitant (n-alkane) to oil dilution are considered as input variables for estimating the amount of precipitated asphaltene. The results obtained in this study reveal a satisfactory agreement between the values estimated by both the LSSVM and GEP models (compared to the developed DT model), to experimental data for asphaltene precipitation. Finally, the results obtained by the models developed in this study are compared with the results of a previously reported algorithm (*i.e.* artificial neural network) as well as the available empirically derived methods. The results show that models developed in this study are reliable, and superior to those comparative methods studied.

Keywords: *Asphaltene; Method; Temperature; Molecular weight; Precipitant.*

1. Introduction

Asphaltenes molecules are complex structures and heaviest part of crude oils that cannot be dissolved in some light hydrocarbons e.g. n-pentane, n-heptane, and n-decane [1]. The precipitation of asphaltenes from crude oil is directly associated with its stability conditions including changes in reservoir pressure and temperature as well as chemical composition of petroleum fractions [2-3]. As a result, precipitation of asphaltene is known as a problematic phenomenon in petroleum industry in particular during oil production from hydrocarbon reservoirs to the pipelines. In petroleum industry, the precipitation of asphaltene can cause serious problems including reduced oil recovery and relative permeabilities, blockage of rock pores, and reduction of flow rate. As a matter of fact, asphaltene precipitation causes the reduction of relative permeabilities with change of reservoir rock wettability from water-wet rock to oil-wet rock which can decrease the oil recovery factor [4-5]. Additionally, wellbore damage with blocking the rock pores, and the reduction of processing facilities capability with plugging of surface pipelines during petroleum production are two other serious problems associated with asphaltene precipitation [6]. Furthermore, asphaltene precipitation is an important issue in enhanced oil recovery processes in particular carbon dioxide flooding. During injection of carbon dioxide into wells, contacts between the oil and injected CO_2 can change the reservoir fluid properties and also its phase equilibrium conditions, and consequently cause precipitation of heavy and complex hydrocarbon mixtures or asphaltenes [7]. Here, it is worth mentioning

that the type and amount of asphaltene precipitated from the crude oils may differ from one geographical location to another because of nonuniformity of petroleum reservoirs in terms of oil gravity and density [8].

There is a significant number of attempts made by researchers to address the asphaltene precipitation during petroleum production. Loureiro *et al.* [9] studied effect of carbon dioxide and n-heptane on the behavior of asphaltene precipitated. To this end, they employed ultraviolet–visible (UV–vis) spectrometry to monitor phase behaviour of asphaltene precipitation. Hemmati-Sarapardeh *et al.* [10] developed a SARA fraction based model using least square support vector machine (LSSVM) algorithm for the estimation of asphaltene precipitation of Iranian oils. They indicated that the results obtained by their developed model are in satisfactory agreement with the corresponding experimental data. Zendehboudi *et al.* [11] performed some laboratory tests on asphaltene precipitation to observe the influence of mixture composition, temperature, pressure, pressure drop, and dilution ratio. They also compared the obtained results with an artificial neural network (ANN). They found that temperature and pressure drop have significant impacts on the precipitation of asphaltene. Ju *et al.* [12] developed a 3D multiphase model indicating the carbon dioxide transport into reservoir and precipitation of asphaltene. In the study, they observed the influence of asphaltene precipitation on the petroleum production trend during CO_2 injection. The results obtained in the study indicates that the permeability and production rate decrease with the precipitation of asphaltene. Lei *et al.* [13] conducted an experimental investigation as well as modeling approach to study asphaltene precipitation inducted with carbon dioxide flooding. Shahebrahimi and Zonnouri [14] developed a thermodynamics model for the determination of asphaltene precipitation. The model is composed of Flory–Huggins as well as None Random Two Liquid (NRTL) models. The obtained results demonstrate a satisfactory precision between the model values and experimental data.

In this study, reliable models were developed using gene expression programming, decision tree (DT), and least squares support vector machine methodologies for the determination of asphaltene precipitation from Iranian crude oil. Finally, the results obtained by models developed in this study are compared with the results of a previously reported algorithm (i.e. artificial neural network) as well as the available empirically derived methods.

2. Asphaltene scaling approach

As a result, existing thermodynamic models may have some short comings in phase behavior modeling of asphaltene precipitation. In other words, thermodynamics approaches need precise characterization of reservoir fluids and asphaltene, and sometimes modeling with thermodynamic methods is difficult because of complex nature of such techniques. Moreover, some of them may have convergence problems. Therefore, presenting alternative and easier methodologies such as asphaltene scaling approach may be of importance. To this end, Rassamdana *et al.* [15] proposed a two-variable scaling equation as a function of precipitant to oil dilution ratio (R), and precipitant molecular weight (M) for predicting the phase behaviour of asphaltene precipitation. Later, Rassamdana and Sahimi [16] modified the scaling equation proposed by Rassamdana *et al.* [15] by observing the influence of temperature (T). The asphaltene scaling equation proposed by Rassamdana *et al.* [15] is formulated as follows:

$$X = \frac{R_v}{M^z} \quad (1)$$

$$Y = \frac{W}{{R_v}^{z'}} \quad (2)$$

where Rv stands for the precipitant to oil dilution ratio; M expresses the precipitant molecular weight, and z and z′ denote the tuning parameters of the above equations. Regardless of oil and precipitant applied in the experiments, z and z′ should be considered 2 and −2 [15]. The scaling equations mentioned above can be expressed with a new form through a polynomial function as below:

$$Y = A_1 + A_2X + A_3X^2 + A_4X^3 \quad (X \geq X_c) \quad (3)$$

where Xc denotes the value of X at the onset point of asphaltene precipitation, and A1, A2, A3, and A4 are considered as the scaling coefficients. As mentioned earlier, Rassamdana and Sahimi [16] modified the scaling equation by considering the influence of temperature as follows:

$$x = \frac{X}{T^{C_1}} \quad (4)$$

$$y = \frac{Y}{X^{C_2}} \quad (5)$$

$$y = b_1 + b_2 x + b_3 x^2 + b_4 x^3 \quad (x \geq x_c) \quad (6)$$

In Eqs. (4-6), *X* and *Y* are same variables which are defined by Eqs. 1 and 2, C_1 and C_2 denote the adjustable parameters of the asphaltene precipitation scaling equation (a good fit of experimental data of asphaltene precipitation with predicted values by Eqs. (4-6) is obtained at C_1=0.25 and C_2=1.6), and b_1, b_2, b_3, and b_4 are considered as the coefficients of third-order polynomial scaling equation regarding effect of temperature.

3. Asphaltenic crude oil data

According to the asphaltene scaling equations proposed by Rassamdana *et al.* [15] and Rassamdana and Sahimi [16], the precipitant to oil dilution ratio, temperature, and precipitant molecular weight are the most important parameters for predicting the amount of precipitated asphaltene (Wt). To forecast phase behaviour of asphaltene precipitation, an asphaltenic crude [17] with oil density of 0.934 g/cc taken from one of the Southwestern reservoirs in Iran was used to pursue our modeling target in this study. The SARA analysis of asphaltenic crude oil is as follows: saturates=29.3 wt. %, aromatics=35.2 wt. %, resins=27.2 wt. %, and asphaltenes=8.3 wt. %. The temperatures tested for measuring the amount of precipitated asphaltene are 30, 50, and 70°C, which were undertaken at atmospheric pressure. Furthermore, at different dilution ratios, three asphaltene precipitants including n-pentane, n-hexane, and n-heptane were used. Table 1 summarizes the ranges of parameters applied for estimating the amount of precipitated asphaltene related to Iranian crude oil [17].

Table 1. Ranges of the parameters applied for asphaltene precipitation

Property	Unit	Minimum	Average	Maximum	Role
Asphaltene precipitation	wt %	0.5	4.78	10.4	Output
MW	–	72.15	68.18	100.21	Input
T	°C	30	50	70	Input
Rv	mL/g	0.67	7.61	20	Input

4. Model development

4.1. Least squares support vector machine

The support vector machine (SVM) algorithm analyzes data and identifies patterns applied for solving classification and regression problems which was developed on the basis of structural risk minimization and statistical learning theory [18-19]. Suykens and Vandewalle [18] presented LSSVM methodology, which is a modified version of classical SVM algorithm introduced by Vapnik [20]. Quadratic programming is applied to solve classical SVM form which is often convoyed by large memory requirement and time-consuming while LSSVM strategy implements equality constraints to replace the original convex quadratic programming problem [21].

In the presence of a dataset $\{(x_1, y_1), \ldots, (x_m, y_m)\} \subset \mathbb{R}^n \times \mathbb{R}$, where each output $y_i \in \mathbb{R}$ and the input $x_i \in \mathbb{R}^n$, LSSVM for regression problem is introduced as minimization of the following formula [22-23]:

$$\min J(w, \xi) = \frac{1}{2}\|w^2\| + \frac{1}{2\mu}\sum_{i=1}^{m}(\xi_i)^2 \quad (7)$$

$$s.t.: y_i = w^T \varphi(x_i) + b + \xi_i, i = 1, 2, \ldots, m$$

where $\varphi(x_i)$ is a nonlinear function which maps the input space into a higher dimensional space. By introducing Lagrange multipliers and exploiting the optimality constraints, the decision function takes the following form:

$$f(x) = \sum_{k=1}^{N} \alpha_i K(x, x_j) + b \quad (8)$$

where α_i stands for the introduced Lagrange multiplier and $K(x,x_j)$ denotes the Kernel function as follows:

$$K(x, x_k) = \Phi(x)^T . \Phi(x_k) \quad (9)$$

As a result, the radial basis function (RBF) Kernel was used in this study as formulated below [18,24-25]:

$$K(x_i, x_j) = \exp\left(-\|x_i - x_j\|^2 / \sigma^2\right) \quad (10)$$

where σ is an adjustable parameter called kernel bandwidth.

4.2. Decision tree

The decision tree algorithm is one of the known artificial intelligence algorithm which is able to develop mathematical models to solve regression or classification problems with the shape of a tree structure. Decision trees can process both categorical and numerical data [26-28]. As a result, the DT algorithm splits a main databank into smaller ones while a related decision tree is incrementally proposed simultaneously. The outcome of this process is a main tree with decision and leaf nodes so that each decision node has two or more divisions. Furthermore, the leaf node illustrates a decision on the target. As a matter of fact, the top decision node in a tree is related to the best predictor so-called root node. As a consequence, classification and regression tree (CRT), chi-square automatic interaction detector (CHAID), Exhaustive CHAID, and quick-unbiased-efficient statistical tree (QUEST) are three kinds of the DT algorithm. The CRT is a recursive subdividing technique, which is used both for regression and classification [29-31]. The CHAID technique was established based on the X^2-test of association [30,32]. The Exhaustive CHAID procedure tries to solve the problem of optimum division by continuing to combine groups, regardless of significance level, until only two groups stay for each predictor [30,33]. Finally, the QUEST method is a binary split decision tree process for classification and data analysis.

4.3. Gene expression programming

Recently, Ferreira [34] developed an intelligent evolutionary algorithm called gene expression programing which is able to construct symbolic models mathematically. In GEP approach, control parameters, function set, fitness function, terminal set, and termination condition are recognized as the key components [35]. Those parse trees are known as expression trees (ETs) for the GEP algorithm [36]. Hence, the nature of gene expression programming authorities the evolution of more complex programs composed of various substructures or subprograms so-called GEP genes. For illustrating the mathematical performance of the GEP methodology in developing symbolic models, a simple GEP-based equation counting a chromosome composed of two genes connected together by a multiplication fitness function is expressed as follows:

$$(u * v) + \left(\frac{f}{l}\right) \quad (11)$$

where u, v, f and l express the input variables for estimating the target variable (asphaltene precipitation), and /, * and + stand for the fitness functions.

5. Results and discussion

In this study, three reliable models were developed to estimate the amount of asphaltene precipitated from a crude oils extracted from Iranian reservoirs. To this end, asphaltene precipitation models are considered as a function of precipitant to oil dilution ratio, temperature, and precipitant molecular weight. As mentioned earlier, the LSSVM modeling approach has two adjustable parameters including γ and σ^2 which should be adjusted through an external optimization methodology. In this study, coupled simulated annealing (CSA) [37-39] was employed for obtaining the optimum values of the LSSVM parameters. As a result, the values adjusted by the CSA approach for the LSSVM developed in this study to estimate asphaltene precipitation are 1.0465 and 927850.8749 for σ^2 and γ, respectively. To develop a DT model,

we used the available MATLAB codes related to the regression DT approach. In regression DT approach, the Y variable takes ordered values and a regression model is fitted to each node to give the predicted values of Y [40].

To propose the GEP-based model, we applied one gene with 30 chromosome, and also average absolute relative deviation was utilized as accuracy function. The head size equals to 7 and a function set including *, /, – and + is selected during applying the GEP methodology. To achieve a high accurate and more capable model, the stop condition of the algorithm was set on maximum generation with a best number of 72 thousands. The final model obtained by the GEP algorithm developed in this study is a simple-to-use with lowest possible coefficients as follows:

$$W_t = \frac{-0.9048\, R_v\, (M - 188.76)}{6.1211\, R_v + T + 33.302} \qquad (12)$$

where W_t denotes the amount of precipitated asphaltene (wt. %), M stands for the precipitant molecular weight, T expresses the temperature (°C), and finally, R_v is the precipitant to oil dilution ratio (mL/g).

In this study, an important error parameter called average absolute relative deviation (AARD) was used for measuring the accuracy and prediction capability of the developed models as follows:

$$AARD\% = \frac{1}{n}\sum_{i=1}^{n}\left|\frac{X_{\exp} - X_{\text{rep./pred}}}{X_{\exp}}\right| \times 100 \qquad (13)$$

Furthermore, AARD error parameter was applied to compare the results obtained by the models developed in this study with the results obtained by the artificial neural network model proposed by Ashoori *et al.* [17] and their scaling equation as well as asphaltene scaling equations presented by Hu and Guo [41] and Rassamdana *et al.* [15].

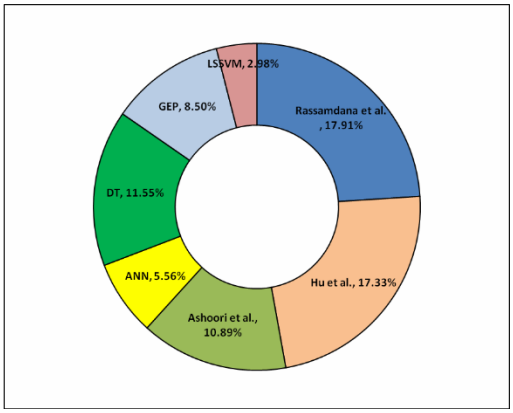

Fig. 1. AARD calculated for the developed models as well as the comparative methods investigated in this study

Fig.1 illustrates the graphical comparison of AARD calculated for all models developed in this study (*e.i.* the LSSVM, DT and GEP-based models) and ANN modeling approach, as well as three asphaltene scaling equations proposed by Ashoori *et al.* [17], Hu and Guo [41], and Rassamdana *et al.* [15]. To compare all models investigated in this study, two panels including smart techniques and symbolic equations should be considered. In the smart techniques panel, the LSSVM model developed in this study has the highest accuracy in comparison with the DT and ANN models. The AARD reported for the LSSVM model is 3 %, while AARDs calculated for the ANN and DT models are 5 and 11%, respectively.

In another panel, the equation developed based on the GEP approach is more accurate than the scaling equations proposed by Ashoori *et al.* [17], Hu and Guo [41], and Rassamdana *et al.* [15]. The AARD calculated for the methods mentioned earlier are 8.5, 10.9, 17.3, and 17.9, respectively.

To show the capability performance of the models investigated in this study, a graphical analysis in terms of parity diagram or crossplot and relative error distribution plot was performed. To this end, four most accurate methods investigated (*e.i.* the LSSVM, ANN, DT, and GEP-based models) for estimating asphaltene precipitation were considered. Fig. 2 demonstrates the crossplots sketched for the four methods. As is clear from the figure, the distribution of data points obtained by LSSVM model around unit slope line is lower than the other methods. In other words, R-squared error obtained by the LSSVM model is higher than the ANN, DT,

and GEP-based models. Fig. 3 illustrates the relative error distribution plots for the LSSVM, ANN, DT, and GEP-based models. The figure clearly shows that the distribution of relative error around Y=0 (zero error) is lower than the other methods.

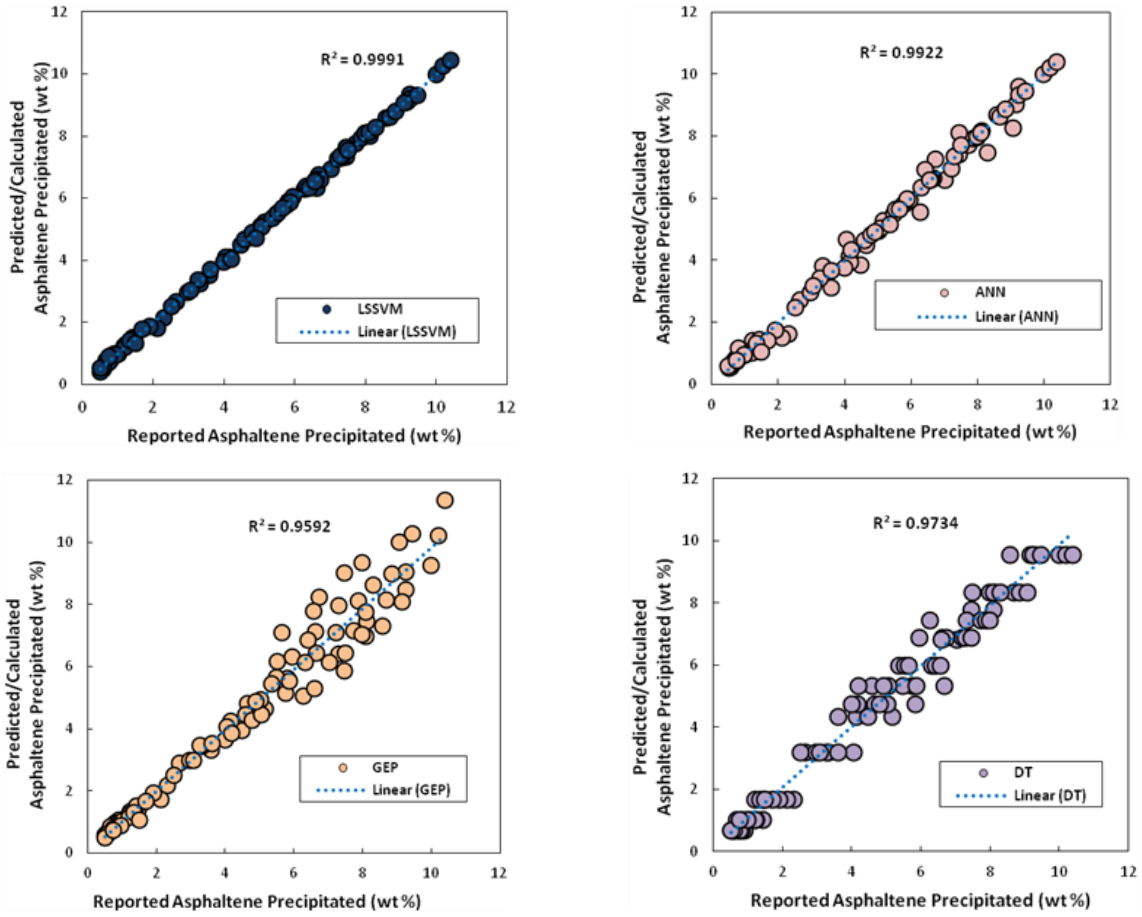

Fig. 2. Crossplots for the different methods investigated in this study with respect to R-squared error

To show smoothness performance of the models mentioned earlier, a trend analysis of the asphaltene precipitation versus precipitant to oil dilution ratio data was done at various temperatures for *n*-pentane, *n*-hexane, and *n*-heptane. Figs. 4-6 indicate the trend plot of asphaltene precipitation changes versus dilution ratio at temperature of 30°C for *n*-pentane, *n*-hexane, and *n*-heptane, respectively. Furthermore, Figs. 7-9 illustrate the changes of asphaltene precipitation versus precipitant to oil dilution ratio at temperature of 50°C for *n*-pentane, *n*-hexane, and *n*-heptane, respectively. Finally, Figs. 10-12 show the changes of asphaltene precipitation at 70°C. As is clear from the figures, the data points related to the LSSVM model are matched with the experimental values better than the other model.

This clearly shows that the LSSVM model is more capable for the estimation of asphaltene precipitation as a function of temperature, precipitant to oil dilution ratio and precipitant molecular weight. Additionally, the model proposed by LSSVM approach has only two adjustable parameter, while the other methods require more parameters. As a result, the high number of adjustable parameters can increase the error of a model considerably. The results obtained in this study confirm this fact that they are appropriate for the prediction targets in petroleum industry, and also simulating the heavy organics precipitation such as asphaltene.

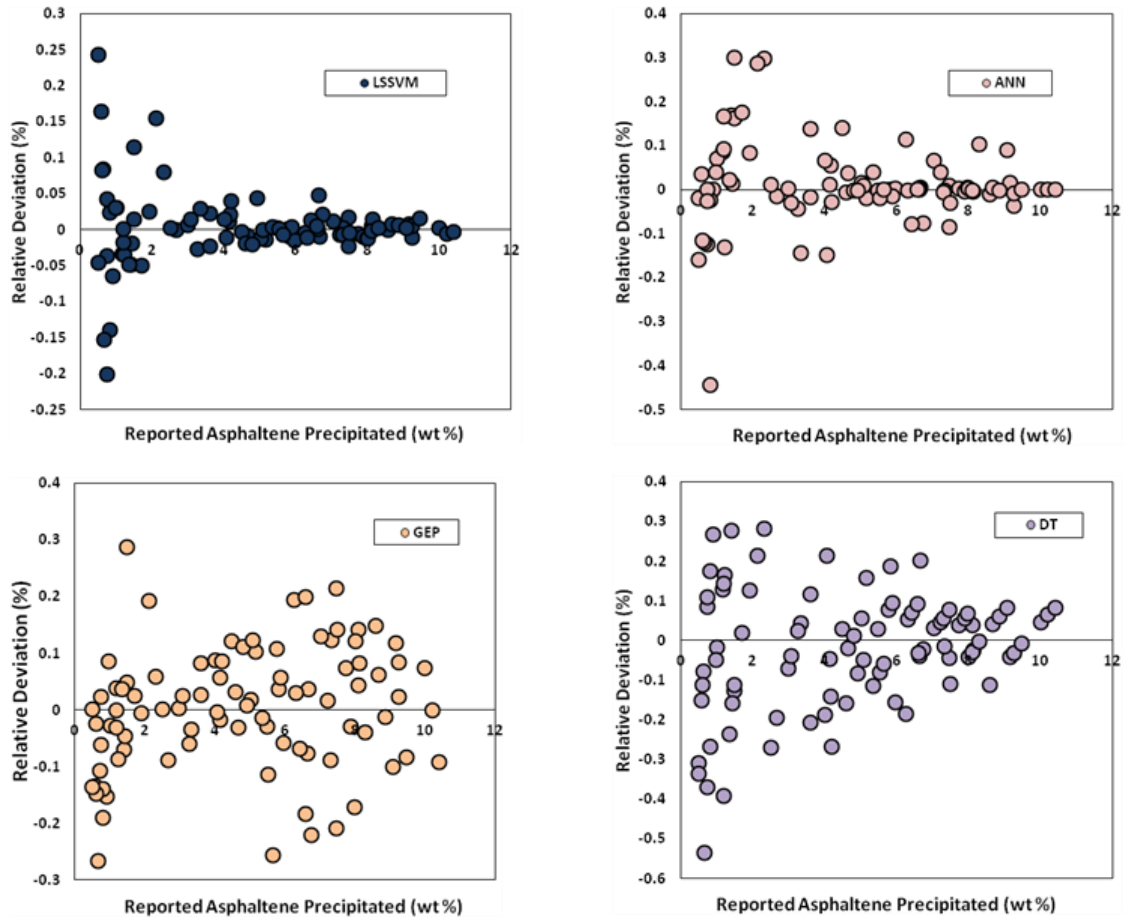

Fig. 3. Graphical relative error distribution analysis for the different methods investigated in this study

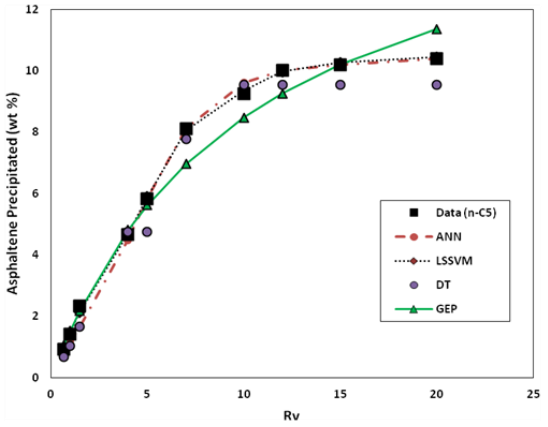

Fig. 4. Trend plot of asphaltene precipitation changes versus dilution ratio at temperature of 30°C for n-C_5

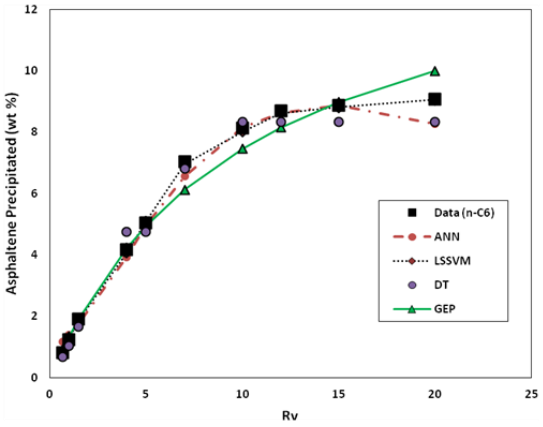

Fig. 5. Trend plot of asphaltene precipitation changes versus dilution ratio at temperature of 30°C for n-C_6

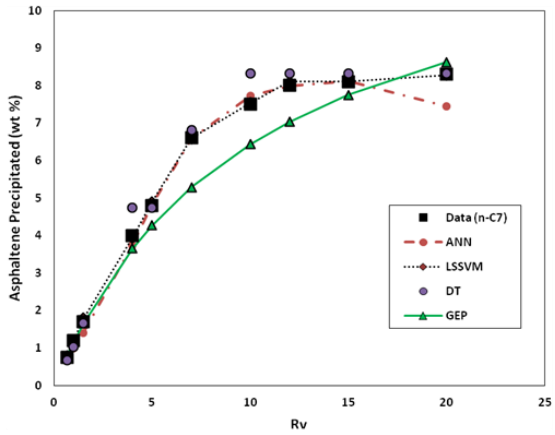

Fig. 6. Trend plot of asphaltene precipitation changes versus dilution ratio at temperature of 30°C for n-C7

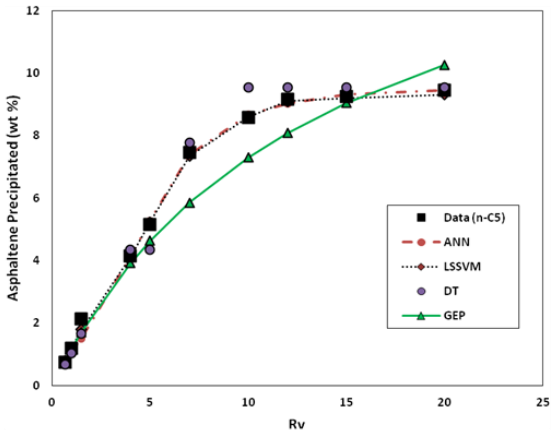

Fig. 7. Trend plot of asphaltene precipitation changes versus dilution ratio at temperature of 50°C for n-C5

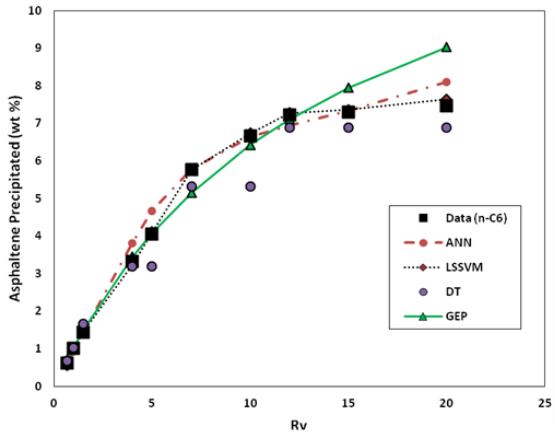

Fig. 8. Trend plot of asphaltene precipitation changes versus dilution ratio at temperature of 50°C for n-C6

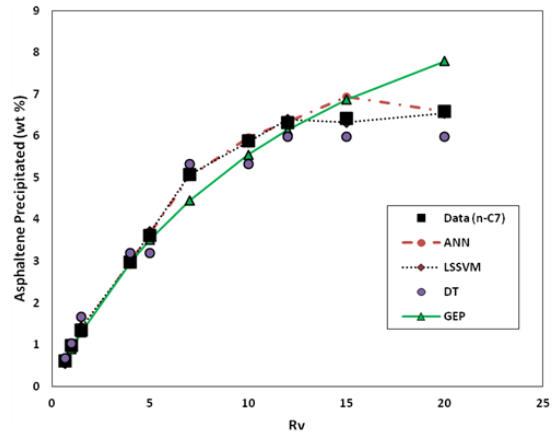

Fig. 9. Trend plot of asphaltene precipitation changes versus dilution ratio at temperature of 50°C for n-C7

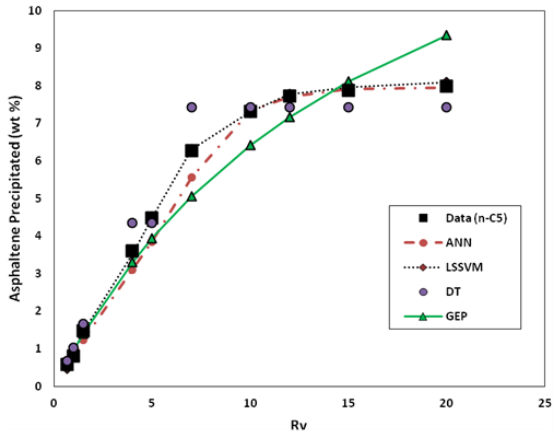

Fig. 10. Trend plot of asphaltene precipitation changes versus dilution ratio at temperature of 70°C for n-C5

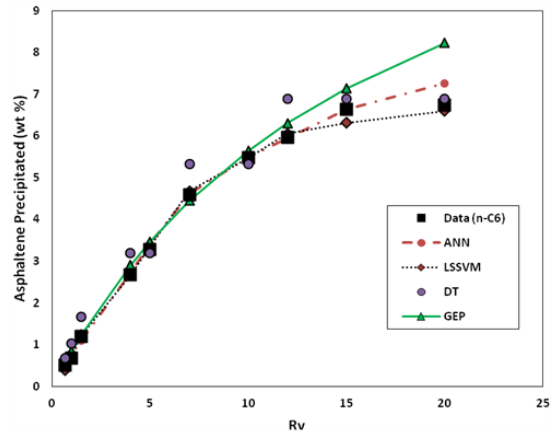

Fig. 11. Trend plot of asphaltene precipitation changes versus dilution ratio at temperature of 70°C for n-C6

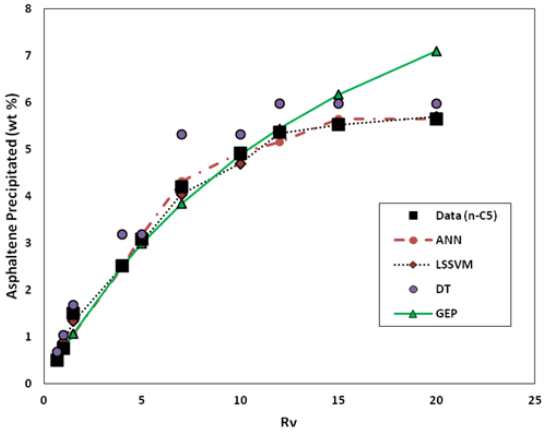

Fig. 12. Trend plot of asphaltene precipitation changes versus dilution ratio at temperature of 70°C for n-C_7

6. Conclusions

An Iranian asphaltenic crude oil was used to study the changes of amount of precipitated asphaltene versus precipitant to oil dilution ratio at different temperatures. Applying the data, it was found that the previously reported scaling equations are not fully accurate and satisfactory. Therefore, most reliable modeling approaches including least squares support vector machine, decision tree and gene expression programming were developed in this study. To compare all methods investigated in this study, two panels were considered. In the smart based panel, the LSSVM approach has the highest precision so that could estimate the asphaltene with an AARD of 3%. In symbolic equations panel, the method proposed on the basis of GEP approach gave an acceptable AARD of 8.5 %. The methods developed in this study are applicable for the simulation of asphaltene precipitation softwares. Moreover, simple symbolic method presented in this study can be a reliable alternative for existing scaling equations and complex thermodynamic methods.

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