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A rigorous approach for determining interfacial tension and minimum miscibility pressure in paraffin-CO_2 systems: Application to gas injection processes

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ABSTRACT

Determination of interfacial tension (IFT) between the reservoir crude oil and the injecting gas as well as the minimum miscibility pressure (MMP) are the keys for successful gas injection process for enhanced oil recovery (EOR) in the matured oil fields. In this study, a novel supervised learning method called least square support vector machine (LSSVM) was developed to estimate IFT of paraffin-CO_2 system. Besides, the MMP of the same system is estimated using the same model by using the vanishing interfacial tension (VIT) technique. The IFT was assumed to be an explicit function of pressure, temperature and molecular weight of paraffin, which was considered as the basis of the proposed model. The results showed that the proposed model is able to predict the IFT values with an average absolute percentage relative error of 4.7%. The highest relative error for estimation of MMP was found to be only 6.79%. Also, relevancy factor showed that pressure has the largest impact on the IFT of paraffin-CO_2 systems. At the end, the Leverage approach demonstrated that the proposed model is statistically valid and acceptable and only 3.8% of the data points were out of the applicability domain of the model.

1. Introduction

Surface tension is a contractive attitude of the surface of a liquid that causes it to resist against external forces [1]. In a single phase system, net forces, *i.e.* attraction and repulsion forces between molecules are zero. In a two phase system, density of molecules in vapor phase is lower than their density in liquid phase and thus there are unequal number of molecules at the interface of two phases. Due to this fact molecules are pulled toward the liquid phase which is responsible for surface tension phenomena [1]. Interfacial tension (IFT) is an important criterion of similarity between two molecules. In other words, less difference between two molecules interfacial tension means more similarity between them [2].

Interfacial tension plays a key role in production from oil reservoirs such as determining the amount of remaining oil in reservoirs, performance prediction of fractured reservoirs, enhanced oil recovery (EOR) processes, etc. [3]. Interfacial tension also plays an important role in chemical engineering applications including heat transfer and phase changes. Production from oil reservoirs decreases after a while because of capillary and viscose forces. More than half of the crude oil remains in reservoirs without performing an enhanced oil recovery process. Gas injection is one of the most effective EOR methods and has been widely used for recovery enhancement purposes. Carbon dioxide, hydrocarbon gases, nitrogen and flue gas are some of the appropriate gases for EOR gas injection processes. Miscible and immiscible displacements are two methods for gas injection. Miscible gas injection is more considered to be utilized in industry than immiscible gas injection. Miscibility can be considered in terms of zero interfacial tension between two phases. Designing a miscible gas injection for EOR purposes requires accurate determination of minimum miscibility pressure (MMP). MMP is the key parameter for finding out if there is miscibility condition at a specific reservoir temperature. Different experimental methods have been recommended for MMP determination such as slim tube and rising bubble apparatus (RBA). Recently, new experimental approaches are used to

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determine MMP. Vanishing interfacial tension (VIT) technique measures IFT at different reservoir pressures to find the pressure at which IFT becomes zero. This pressure is called the MMP.

Interfacial tension varies with composition, pressure and temperature of the bulk fluid and is affected by composition more than other parameters over a normal range of changes [3]. There are two different methods to calculate interfacial tension between two immiscible fluid phases consisting of direct experimental measurements and empirical correlations [1]. Experimental measurements of IFT have been classified into classic and modern methods. Capillary rise, drop weight, ring, and Wilhelmy plate are known as the classic experimental methods which are discussed in detail in literature [4]. The modern methods include pendent drop, sessile drop, and spinning drop which measure the interfacial tension by analyzing the shape of drop. Manning [5] has discussed about these methods. Pendent drop method is the most reliable and accurate method for fluid/fluid systems. In this method, the shape of liquid droplet which is hanging from needle tube is specified and correlated to interfacial tension [6]. Rotenberg et al. [7] used Axisymmetric Drop Shape Analysis-Profile (ADSA-P) technique which is the best shape analyzing method for interfacial tension calculation. This method has been used in experimental works in the recent years for measuring interfacial tension between crude oil and different gases at reservoir condition. Ghasem et al. [8], Firoozabadi et al. [9] and Rao [10] have employed pendent drop technique to measure interfacial tension for oil/gas system at reservoir conditions. Few experimental data is available for two phase systems with multicomponent fluids. Thus, development of accurate empirical correlations for IFT prediction in multicomponent hydrocarbon systems is of great importance. Several methods have been developed for IFT estimation of single and multicomponent fluid systems. Parachor model [11,12], corresponding state theory [13], thermodynamic correlations [14] and Gradient Theory [15] are some of the most well-known models for IFT calculation. It is worth to mention that all of these methods have a similar background development. Parachor model [11,12] is the oldest and the most widely used method in the petroleum industry for IFT calculation between fluids. Macleud–Sudgen [11,12] developed a model that relates surface tension of pure compounds to molar density difference between phases. Parachor is the proportionality constant between IFT and density difference between phases. Exner [16] described Parachor as the molar volume at a temperature that surface tension has a unit value. Weinaug and Katz [17] developed Parachor model for hydrocarbon mixtures. This model has been widely used for calculating interfacial tension in binary mixtures. They used the following equation to determine IFT of hydrocarbon mixtures.

$$\sigma = \left[\sum_{i=1}^{n} P_i\left(\frac{x_i}{V_l} - \frac{y_i}{V_v}\right)\right]^4 \tag{1}$$

where P_i is the Parachor of component i, x_i and y_i are the mole fractions of component i in the liquid and vapor phase, respectively. Reno and Katz [18] used Eq. 1 to calculate IFT in multicomponent hydrocarbon mixtures containing dissolved nitrogen. Hough-Stegemeier [19] improved Weinaung and Katz correlation by making a change in the model parameters to achieve a more accurate IFT model for multicomponent mixtures. The only difference between their equation and Eq. 1 is that they used 3.67 instead of 4 for the scaling exponent. Lee and chain [20] developed a model for complex mixtures. In their equation, Parachor can be estimated according to the critical properties of components and scaling exponent is 3.91 instead of 4. Firrozabadi et al. [9] utilized an empirical correlation based on molecular weight to calculate Parachor. They suggested that scaling exponent of 4 is suitable for non-asphaltenic oil reservoirs. Franchi [21] developed another correlation for Parachor and used scaling exponent of 4 to calculate IFT. He developed a correlation for n-alkanes to calculate IFT which relates Parachor to their molecular weight. Fawcett [22] used a correlation for determining IFT in condensate systems. All of these modifications have been developed to match the experimental data.

Accurate calculation of interfacial tension between gas and crude oil is of vital importance as already mentioned. Unfortunately, there is not much experimental data for interfacial tension of gas/crude oil systems. Crude oil is a multicomponent hydrocarbon mixture and IFT prediction models have less accuracy in IFT determination of these systems compared with simple hydrocarbon systems. However, study on pure hydrocarbons could give us a good vision of analyzing complex mixtures and crude oil systems.

In this study, n-alkanes consisting of normal heptane, normal hexadecane, normal decane, normal tetradecane, normal butane and normal dodecane were considered as representative of crude oil. To obtain the best results, a wide range of pure hydrocarbons ($n-C_4$, $n-C_7$, $n-C_{10}$, $n-C_{12}$, $n-C_{14}$, $n-C_{16}$) were chosen. Carbon dioxide (CO_2) is known as the popular gas for gas injection processes in petroleum industry. A large number of experimental data points covering a wide range of IFT between CO_2 and the aforementioned normal alkanes from various literature sources [23–27] were covered. 500 experimental data sets of IFT information were collected which were measured from pendent drop method. Estimation of interfacial tension between normal alkanes and CO_2 gives a good vision of miscibility condition for crude oil-CO_2 systems.

2. Data collection

Validity and correctness of every model development depend on the comprehensiveness of the data set used for development of that model. It was found out from literature that the interfacial tension between alkanes and carbon dioxide is affected by temperature, pressure and composition of both liquid and gas phases [23–25,27,28]. In this study, an attempt was made to use acceptable range of experimental data sets containing interfacial tension information between six types of normal alkanes and carbon dioxide from various literatures sources [23–25,27,29]. These data sets include experimental values of temperature, pressure and composition of components in both gas and liquid phases. The ranges of temperature, pressure and experimental IFT used in this work are presented in Table 1.

3. Model development

Support Vector Machine (SVM) is known as a smart method developed from machine learning community, which recently has been widely used in various types of studies [30–32]. SVM is employed for data analyzing, patterns recognizing, and regression analysis. A principle of SVM algorithm states that any function f(x) can be expressed with two different parts [33].

$$f(x) = w^T\varphi(x) + b \tag{2}$$

The first part includes w^T and $\varphi(x)$ which are transposed output layer vector and kernel function, respectively. The second part of the function or b is called bias. The input of the function is a matrix which has a dimension of $N \times n$. N and n stand for the number of data points and number of input parameters, respectively. The following cost function was proposed by Vapnik [34] to calculate w and b.

$$Cost\ function = \frac{1}{2}w^T + c\sum_{k=1}^{N}\left(\xi_k - \xi_k^*\right) \tag{3}$$