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Modeling interfacial tension and minimum miscibility pressure in paraffin-nitrogen systems: Application to gas injection processes

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ABSTRACT

Nitrogen has emerged as an attractive gas for many petroleum and chemical engineering applications such as gas lift, gas recycling, pressure maintenance, and enhanced oil recovery, especially for some high-pressure reservoirs. Accurate determination of interfacial tension (IFT) in nitrogen/hydrocarbon systems is crucial and required for numerous applications in chemical and petroleum industries. The experimental measurements of IFT are costly, cumbersome and time consuming, especially at high pressure-high temperature conditions. In this study, the IFT of normal alkanes from n-C_5 to n-C_{16} (as the representatives of crude oil) and nitrogen is modeled at a wide range of pressure (from 0.1 to 69 MPa) and temperature (from 295 to 442 K) using group method of data handling (GMDH). Three inputs were used for modeling including pressure, temperature, and molecular weight of normal alkane. To develop the most efficient model, 60% of the dataset was used for model development and the remaining 40% was used to check the validity and accuracy of the developed model. The proposed model predicts the data satisfactorily with an average absolute relative error of 3.81% and 3.91% in training and testing subsets, respectively. Then, the proposed model was compared to the well-known IFT models, namely Linear Gradient Theory (LGT), Density Gradient Theory (DGT), and Parachor approaches combined with the Volume Translated Predictive Peng Robinson Equation of State (VT-PPR EOS). The results demonstrate that the proposed model not only is superior to the existing models in terms of accuracy, but also can predict the IFT with a simple mathematical expression with a quite low computational cost. Finally, a simple-to-use algorithm was proposed to calculate the minimum miscibility pressure (MMP) in normal alkane/nitrogen systems based on the measurements with vanishing interfacial tension (VIT) technique, which predicts the MMP with high accuracy.

1. Introduction

Enhanced oil recovery (EOR) techniques were originally proposed to reduce the residual oil saturation and extract the most possible amount of hydrocarbons over the lifetime of a reservoir prior to its abandonment [1–3]. Gas injection, whether in miscible or immiscible mode, is one of the most widely used EOR methods [1,4,5]. Various types of injection gas include carbon dioxide, natural gas, nitrogen, flue gas, etc. [6]. Carbon dioxide is a promising EOR agent which can increase oil recovery through mechanisms such as vaporization of oil, viscosity reduction and swelling [7–10], moreover, injecting carbon dioxide for carbon sequestration to reduce greenhouse gas emission makes it more favorable to the industry [11–21]. However, there are many technical and economic challenges involved in CO_2 injection. Asphaltene precipitation as a major drawback could result in failure of a CO_2 EOR process through oil relative permeability reduction and flow blockage in the reservoir and surface facilities; the gas source must be available in large amounts to make the process economically viable; the operation costs are pretty high and corrosion in some cases might cause financial loss to the company [6,22–24].

Nitrogen injection, on the other hand, causes neither asphaltene precipitation nor corrosion in the wellbore or facilities and the source is readily available through cryogenic separation of air or from natural underground reservoirs [25,26]. Nitrogen has a high minimum miscibility pressure (MMP) with crude oil and is usually utilized for immiscible displacement, pressure maintenance and gas lift, however, where the oil reservoir is relatively deep and the pressure is quite high, nitrogen could form a miscible bank

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with the crude [27]. Two miscible compounds form a single phase upon mixing and there is no interface in between, i.e., the interfacial tension (IFT) of the system is zero [4].

IFT is defined as the force exerted per unit length of the interface between two phases. It plays a key role in heat and mass transfer in chemical engineering calculations. IFT determines the distribution of phases in the porous medium of an oil reservoir and the residual oil saturation strongly depends on IFT [4,28]. MMP is the minimum pressure at which IFT is zero and accurate determination of MMP is of a high significance since increasing the injection pressure is costly [29]. Therefore, application of nitrogen in EOR processes basically requires the knowledge about the IFT of nitrogen-hydrocarbon systems.

IFT is a function of pressure, temperature and composition of the fluids present in the system [30]. Two methods for the determination of IFT include experimental measurements and theoretical models/correlations [31]. Capillary rise, drop weight, ring and Wilhelmy plate are classified as classic experimental methods [32] and pendent drop, sessile drop and spinning drop are known as the modern techniques of IFT measurement, in which IFT is modeled through drop shape analysis [33]. Pendent drop is well known to yield the most reliable results [34]. The theoretical approaches include Parachor, Linear Gradient Theory (LGT) and Density Gradient Theory (DGT). In the following, these theoretical models are reviewed briefly.

The Parachor model [35,36] provides a satisfactory description of hydrocarbon mixtures at high pressure and temperature and it is the most commonly used approach in the industry. Weinaug and Katz [37] extended the model developed by Macleod [35] and Sugden [36], which was developed for pure substances, to mixtures using molar averaging as expressed in Eq. (1).

$$IFT = \left[\sum_{i=1}^{N} P_{ch,i}(x_i\rho^l - y_i\rho^v)\right]^E, \tag{1}$$

where N is the number of components, $P_{ch,i}$ is the Parachor value of component i, x_i and y_i are the equilibrium mole fractions of component i in the liquid and vapor phases, respectively, ρ^l and ρ^v represent the liquid and vapor phase molar density, respectively and E is the scaling exponent. Values ranging between 3.45 and 4 for the scaling exponent and correlations for the Parachor values as functions of molecular weight, specific gravity and/or critical properties have been proposed in literature [38,39].

The DGT is a more mathematically involved approach which has shown high capability at predicting the IFT of mixtures. The density profiles of components in the system are computed across the interface using the phase equilibrium properties of bulk phases [40,41]. This approach found popularity after the work of Chan and Hilliard [42]. The following equations are used to calculate the IFT between two phases.

$$IFT = \int_{-\infty}^{+\infty} \sum_i \sum_j c_{ij} \frac{d\rho_i}{dz}\frac{d\rho_j}{dz} dz = \int_{-\infty}^{+\infty} 2\Delta\Omega(\rho) dz, \tag{2}$$

$$\Delta\Omega(\rho) = f_0(\rho) - \sum_i \rho_i\mu_i + p, \tag{3}$$

where c_{ij} is the cross influence parameter, $\Delta\Omega$ represents the variation in the grand thermodynamic potential, f_0 is Helmholtz free energy at the local density ρ, p is the equilibrium pressure and μ_i stands for the chemical potential of pure component i at equilibrium conditions.

A mixing rule is applied to the influence parameters of pure components, c_i and c_j, to compute the cross influence parameter, c_{ij}, as follows:

$$c_{ij} = (1 - \beta_{ij})\sqrt{c_i c_j}, \tag{4}$$

where β_{ij} is the binary interaction coefficient.

The LGT approach was developed by Zuo and Stenby [43,44], where the density profiles are assumed to change linearly across the system interface of thickness L, as shown in Eq. (5). This assumption reduces the computational load to a large extent.

$$\frac{d\rho_i}{dz} = \frac{\rho_i^l - \rho_i^v}{L}, \tag{5}$$

Zuo and Stenby [44] proposed the following equation for the computation of the influence parameter of the mixture c from the cross influence parameters.

$$c = \sum_i^N \sum_j^N c_{ij} x_i x_j, \tag{6}$$

Then, the IFT of the mixture can be calculated using Eq. (7), where ρ_1 is the reference component density (usually the density of the less volatile component which changes monotonically with z) and ρ_2 is the density of the second component expressed as a function of ρ_1.

$$IFT = \int_{\rho_1^v}^{\rho_1^l} \sqrt{2c\Delta\Omega(\rho_1, \rho_2)}\, d\rho_1, \tag{7}$$

As already mentioned, accurate determination of IFT in gas/crude oil systems is of vital significance and is required for numerous applications in chemical and petroleum industries. The experimental measurements are costly, cumbersome and time consuming. The empirical correlations do not usually provide the adequate accuracy and theoretical approaches such as DGT involve a large load of computational and numerical effort, for example, using an equation of state and flash calculations are inevitable. The purpose of this study is to develop a simple model for the prediction of IFT, which is less computationally demanding and is able to yield accurate results quickly and efficiently. For this aim, a large data bank of experimental IFT values between normal alkanes of n-C_5, C_6, C_7, C_8, C_{10} and C_{16} (as the representatives of crude oil) and nitrogen [45–50], covering a wide range of temperature and pressure measured from pendent drop method, was gathered from different literature sources. The experimental data points on the IFT between injection gases such as nitrogen and crude oil are not sufficient for developing a precise model and approximating the crude with normal alkanes would provide a good insight into the analysis of IFT of gas/oil systems. The only variables in the proposed model are temperature, pressure and molecular weight of the normal alkane and the procedure of computing IFT is explicit and quite simple. In addition, an easy-to-follow algorithm with an approximate error of 3% is proposed for predicting MMP of nitrogen-normal alkane mixtures.

2. Data collection

The validity of a model greatly depends on the accuracy and the range of operating conditions covered by the dataset used in the model development. In this study, a large dataset covering a wide range of pressure (from 0.1 to 69 MPa) and temperature (from 295 to 442 K) including 368 experimental IFT measurements between six normal alkanes and nitrogen, as presented in Table 1, was collected from various open literature sources [45–50]. The statistical description of the selected input variables and the output of the model developed in this study are shown in Table S1. Temperature, pressure and the molecular weight of the n-alkane were chosen as the input variables. The small values of correlations between the mentioned variables, as shown in Table S2, confirms the independence of the chosen variables as the inputs of the proposed model. A random division of the dataset into subsets of training and test-