Modeling of CO_2 vapor-liquid equilibrium in Colombian heavy oil using SARA analysis

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Abstract
The solubility of CO_2 in Colombian heavy oil was calculated using the Peng-Robinson cubic equation of state and the Lee-Kesler correlations. The crude was represented as a mixture of pseudo-components and for each one of them, the thermodynamic and critical properties were estimated. The results obtained in representing the oil with four, five and six pseudo-components show that all these representations produce similar results and therefore the use of four pseudo-components is sufficient and has a lower computational cost. Excellent results were obtained by comparing the experimental and calculated data. For this system, it is enough to have a complete characterization of the SARA analysis and to use four pseudo-components to adequately model the vapor-liquid equilibrium of CO_2 -heavy oil.

Keywords: Equation of state; pseudo-component; SARA analysis; crude; solvent; phase equilibrium.

Modelado del equilibrio líquido-vapor CO_2-crudo pesado colombiano con análisis SARA

Resumen
La solubilidad del CO_2 en un crudo pesado colombiano fue calculada usando la ecuación cúbica de estado de Peng-Robinson y la correlación de Lee-Kesler. El crudo fue representado como una mezcla de pseudo-componentes y para cada uno de ellos se calcularon las propiedades termodinámicas y críticas. Los resultados obtenidos en la representación del crudo con cuatro, cinco y seis pseudo-componentes muestran que todas las representaciones producen resultados similares y por lo tanto el uso de cuatro pseudo-componentes es suficiente y tiene un costo computacional más bajo. Fueron obtenidos excelentes resultados al comparar los datos experimentales y calculados. Para este sistema es suficiente tener una completa caracterización del análisis SARA y usar cuatro pseudo-componentes para modelar adecuadamente el equilibrio líquido-vapor de CO_2-crudo pesado.

Palabras clave: Ecuación de estado; pseudo-componente; análisis SARA; crudo; solvente; equilibrio de fases.

1. Introduction

Oil is a complex mixture of hydrocarbons produced from sedimentary rocks in gas form (natural gas), liquid (crude oil), semisolid (bitumen) or solids (wax or asphaltite) [1].

Most of the world's oil resources correspond to viscous and heavy hydrocarbons, which are difficult and expensive to produce and refine. Heavy oil, extra heavy oil and bitumen make up 70% of the world's total oil resources oscillating between 9 and13 trillion barrels [2].

A significant number of laboratory studies have been undertaken to identify technologies that can be applied as a solution to the phenomena that negatively affect the performance of implemented secondary recovery processes. These methods focus mainly on increasing oil mobility and thus increase the production thereof. The goal is to reduce its viscosity so that oil can flow easily. Enhanced Oil Recovery (EOR) can be achieved through technologies such as gas

injection, chemical injection, microbial injection, or thermal recovery [3].

The implementation of EOR methods plays a key role as a technology to increase the recovery factor of Colombian fields, and to refurbish them in the implementation of other technologies designed to increase well productivity [4].

The main advantage of gas injection is its microscopic sweep. The gases used to EOR are mostly methane, butane, propane, nitrogen and carbon dioxide. Many of the aforementioned gases have the advantage of being produced in-situ, i.e. large volumes are available in the wellbore for injection. The choice of a particular gas is strongly linked to its availability and to the increased recovery generated. Thus, CO_2 appears as one of the most promising gases for use in EOR.

It is therefore important to have a thermodynamic model to adequately characterize the phase behavior of CO_2 - heavy oil mixtures. Such models can provide judgment elements for decision-making regarding production, transportation, refining and upgrading oil.

The experimental data reported in the literature to characterize the phase equilibrium for CO_2 - heavy oil systems are generally few and so the difficulties in establishing appropriate thermodynamic models are high. Therefore, tools allowing phase equilibrium calculations of such systems with little experimental data are required.

The aim of this work is to develop a model for the calculation of vapor-liquid equilibrium for CO_2 - Colombian heavy oil (called here UnalMed), using the representation of pseudo-components and to estimate the critical and thermodynamic properties of those components. Additionally, we seek here to establish the right number of components to be used for a good representation of the system, with little experimental information and at low computational cost. This information may be useful for developing appropriate thermodynamic models for the Colombian oil industry. To our knowledge, data on thermodynamic properties of pseudo-components to represent Colombian heavy oils have not been previously published.

2. Model

Based on the nature of the mixture of oil, there are several ways to express its composition. Some of the most important types of composition [1,5,6] are PONA analysis (paraffins, olefins, aromatics and Naphthenes), PNA (paraffins, Naphthenes and Aromatics), PIONA (paraffins, Isoparaffins, Olefins, Naphthenes and Aromatics), elemental (C, H, S, N, O) and SARA analysis (Saturates, Aromatics, Resins and Asphaltenes) [7].

PNA and PINA analyses are useful for petroleum products in a range of low boiling temperatures, such as distillates of atmospheric distillation units for crude. However, the SARA analysis is useful for heavy oil fractions, residues and fossil fuels (i.e., coal liquids) that have a high content of aromatics, resins and asphaltenes.

The above analyses are ways to represent oil as a mixture of pseudo components (several components represented in a single fraction), given the complexity of the composition of this. With SARA analysis, for example, oil can be represented in four pseudo components, which are the saturates, aromatics, resins and asphaltenes, and each of them has its properties such as the boiling point, specific gravity, density, molecular weight, among others. In addition, each pseudo component can be modeled as a pure substance.

The critical properties, molecular weight and acentric factor of each pseudo component can be calculated by correlations reported in the literature for heavy oil fractions.

In this work, the critical temperature, critical pressure and acentric factor of each pseudo component were calculated using the Lee-Kesler correlations [8], as shown in (eq.1-6); the molecular weight was calculated with the Daubert-Riazi correlation (eq. 7) and the critical volume with the Hall-Yarborough correlation (eq. 8). These correlations were selected by considering the good results that have been obtained with respect to experimental data along its applications [9,10]. The input parameters of these correlations are the boiling point (T_b) and the specific gravity (SG).

$$
\begin{aligned}
T_c &= 189.8 + 450.6SG + (0.4244 + 0.1174SG)T_b \\
&+ \left(\frac{(0.1441 - 1.0069SG)10^5}{T_b} \right)
\end{aligned}
\tag{1}
$$

$$
\begin{aligned}
lnP_c = 5.689 &- \frac{0.0566}{SG} \\
&- \left(0.43639 + \frac{4.1216}{SG} + \frac{0.21343}{SG^2} \right) 10^{-3}T_b \\
&+ \left(0.47579 + \frac{1.182}{SG} + \frac{0.15302}{SG^2} \right) 10^{-6}T_b^2 \\
&- \left(2.4505 + \frac{9.9099}{SG^2} \right) 10^{-10}T_b^3
\end{aligned}
\tag{2}
$$

For $T_{br} > 0.8$

$$
\begin{aligned}
\omega &= -7.904 + 0.1352K_w - 0.007465K_w^2 \\
&+ 8.359T_{br} + \frac{1.408 - 0.01063K_w}{T_{br}}
\end{aligned}
\tag{3}
$$

Kesler and Lee [11] proposed the following correlation for $T_{br} < 0.8$

$$
\omega = \frac{\frac{-lnP_c}{1.01325} - 5.92714 + \frac{6.09648}{T_{br}} + 1.28862lnT_{br} - 0.169347T_{br}^6}{15.2518 - \frac{15.6875}{T_{br}} - 13.4721lnT_{br} + 0.43577T_{br}^6}
\tag{4}
$$

Where T_c y T_b are in K and P_c in bar and T_{br} is the reduced boiling point defined as,

$$T_{br} = \frac{T_b}{T_c} \tag{5}$$

K_w is the Watson characterization factor [1], given by

$$K_w = \frac{(1.8T_{br})^{1/3}}{SG} \tag{6}$$

$$M = 42.965[exp(2.097 * 10^{-4}T_b - 7.78712SG + 2.08476 * 10^{-3}T_bSG)]T_b^{1.26007}SG^{4.98308} \tag{7}$$

Where T_b is in Kelvin and the molecular weight, M is in g/mol.

$$V_c = 1.56M^{1.15}SG^{-0.7935} \tag{8}$$

In 1976, Peng and Robinson proposed a modification to the Soave-Redlich-Kwong equation of state (EOS) that has been well received internationally [26]. This equation of state is well accepted in technical publications, research and simulators to predict the behavior of natural hydrocarbon systems. PVT variables are related to predict the thermodynamic behavior of a system [12-14].

$$P = \frac{RT}{v - b} - \frac{a(T)}{v(v + b) + b(v - b)} \tag{9}$$

Where P is the system pressure, T is the system temperature, $a(T)$ is a parameter of attraction, b is a parameter of repulsion, v is the molar volume and R is the ideal gas constant.

In terms of the compressibility factor,

$$Z^3 - (1 - B)Z^2 + (A - 3B^2 - 2B)Z - (AB - B^2 - B^3) = 0 \tag{10}$$

Where

$$A = aP/R^2T^2 \tag{11}$$

$$B = bP/RT \tag{12}$$

$$Z = Pv/RT \tag{13}$$

The constants a and b are related to the critical pressure, critical temperature and acentric factor by:

$$a_i(T) = a_i(T_c) * \alpha_i(Tr_i, \omega_i) \tag{14}$$

$$b_i(T) = 0.07780\frac{RT_{c_{i_i}}}{P_{c_i}} \tag{15}$$

Where,

$$a_i(T_c) = 0.45724\frac{R^2T_{ci}^2}{P_{c_i}} \tag{16}$$

$$a_i(Tr_i, \omega_i) = \left(1 + m_i\left(1 - Tr_i^{0.5}\right)\right)^2 \tag{17}$$

$$m_i = 0.37464 + 1.54226\omega_i - 0.26992\omega_i^2 \tag{18}$$

$$Tr_i = \frac{T}{Tc_i} \tag{19}$$

$$a = \sum_i \sum_j x_i x_j a_{ij} \tag{20}$$

$$b = \sum_i x_i b_i \tag{21}$$

$$a_{ij} = (1 - k_{ij})a_i^{0.5}a_j^{0.5} \tag{22}$$

$$k_{ij} = 1 - \frac{8(V_{ci}V_{cj})^{1/2}}{\left(V_{ci}^{1/3} + V_{cj}^{1/3}\right)^3} \tag{23}$$

k_{ij} is the interaction coefficient between components i and j. These parameters have no theoretical basis but are empirical, and their role is to help overcome the deficiencies of the theorem of corresponding states [15].

The fugacity coefficient can be calculated from EOS (eq. 9) [13],

$$ln\frac{f}{P} = \int_0^P \left(\frac{v}{RT} - \frac{1}{P}\right)dP \tag{24}$$

In this manner, the expression for the fugacity coefficient of a pure component is:

$$ln\left(\frac{f}{P}\right) = Z - 1 - \ln(Z - B) - \frac{A}{2\sqrt{2}B}\ln\left(\frac{Z + 2.141B}{Z - 0.414B}\right) \tag{25}$$

The functional form of $\alpha_i(Tr_i, \omega_i)$ was determined by using the values of vapor pressure in the literature and the Newton method to find the values of α to be used in (eq. 10, 25) in a way that the equilibrium condition

$$f^L = f^V \tag{26}$$

is satisfied along the curve of vapor pressure [13]. Where f^L y f^V are the fugacity in the liquid and vapor phase, respectively.

The fugacity coefficient of a k component in a mixture can be calculated by:

$$\ln\left(\frac{f_k}{x_k P}\right) = \frac{b_k}{b}(Z-1) - \ln(Z-B) - \frac{A}{2\sqrt{2}B} * \left(\frac{2\sum_i x_i a_{ik}}{a} - \frac{b_k}{b}\right)\ln\left(\frac{Z+2.141B}{Z-0.414B}\right) \qquad (27)$$

Calculating fugacity coefficients for each component in both, liquid and gas phase with (eq. 27), the vapor-liquid distribution ratio K_i can be calculated by (eq. 28):

$$K_i = \frac{\hat{\Phi}_i^L}{\hat{\Phi}_i^V} = \frac{f_k/x_k P}{f_k/y_k P} \qquad (28)$$

The relationships described by (eq. 28) (one for each pseudo component) provide the main equations for VLE calculations using EOS.

For the development of this work, the heavy oil (UnalMed) was modeled with three different fractions groups: four, five and six pseudo components and CO_2 solubility with UnalMed oil was calculated, in order to determine the amount of pseudo components that should be used to obtain an acceptable thermodynamic model.

3. Results

To model the CO_2 - UnalMed oil system, the crude was initially characterized with four pseudo components, using the SARA analysis. The initial parameters for the calculation of the solubility of CO_2 in oil at different conditions, such as boiling temperature (T_b) and specific gravity (SG) were assumed following some tips outlined in the work of Kariznovi *et al.*, [9]:

- Boiling temperature of the distillable fraction (saturates and aromatics) is less than 600 °C.
- Boiling temperature of the non-distillable fraction (resins) is greater than 600 °C.
- Boiling temperature of the asphaltenes is greater than the boiling temperature of the non-distillable fraction.

Table 1 shows the final parameters for each input of pseudo components that compose the UnalMed crude. These parameters were calculated iteratively within the developed algorithm.

UnalMed crude characterization shown in Table 1 is described by the following order: the first pseudo component corresponds to saturate fraction, aromatics is represented by

PSC 2, the third and fourth pseudo components correspond to resins and asphaltenes, respectively. UnalMed oil composition and CO_2 solubility curve derived from experimental measurements performed by the Colombian Petroleum Institute (ICP).

Table 2 shows the properties of the four pseudo components calculated using correlations presented in Section 2. The binary interaction coefficients between CO_2

Table 1.
Characterization of four pseudo components for UnalMed crude oil.

| Pseudo-component	T_b (ºC)	SG	% Wt
PSC 1	350.00	0.5500	47.529
PSC 2	516.67	0.6833	36.787
PSC 3	673.34	0.8167	11.415
PSC 4	850.00	0.9500	4.269

Source: Prepared.
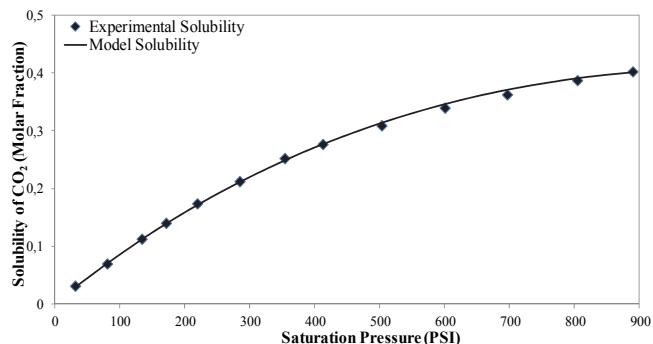
Figure 1. CO_2 solubility in UnalMed crude represented by four pseudo components at 80°F.
Source: Prepared.

and UnalMed crude were tuned to give a closer approximation to the experimental data.

Fig. 1 shows the results obtained by applying four pseudo components to the model. This figure shows the solubility of CO_2 in the crude at 80°F and different pressures, the *x*-axis corresponds to the saturation pressure and the *y*-axis corresponds to the molar composition of the solvent in terms of mole fraction of CO_2. It can be seen that having a relatively low operating temperature, and increasing the saturation pressure, CO_2 is more highly compressed, increasing its solubility in UnalMed crude.

Table 2.
Physical properties of four pseudo components and CO_2 - UnalMed crude interaction

Pseudo-component	M (g/mol)	T_c (ºK)	P_c (MPa)	ω	V_c (cm³/mol)	k_{ij}
PSC 1	233.19	676.59	0.17655	0.99553	1324.4	0.0374
PSC 2	512.58	827.39	0.17336	1.5705	2758	0.0553
PSC 3	931.32	978.56	0.18003	1.8565	4757.4	0.0685
PSC 4	1665.0	1147.5	0.16186	2.0489	8230.4	0.0812

Source: Prepared.

For oil represented by five pseudo components, the same methodology as for the crude representation with four pseudo components was used. The additional pseudo component results from separation of the non-distillable oil fraction, namely resins, as these resins were separated into the asphaltene-free and resins containing asphaltenes. With this as mentioned, the order of

pseudo components is as follows: the first two pseudo components represent UnalMed crude distillable fraction, third and fourth represent the non-distillable fraction of resins (resins 1 and 2), and the fifth pseudo component represents asphaltenes.

Table 3 shows the final input parameters of UnalMed crude represented by five pseudo components.

Table 3.
Characterization of five UnalMed crude pseudo components

Pseudo-component	T_b (ºC)	SG	% Wt
PSC 1	350.00	0.5500	47.529
PSC 2	480.00	0.6500	36.787
PSC 3	590.00	0.7500	7.991
PSC 4	670.00	0.8500	3.425
PSC 5	850.00	0.9500	4.269

Source: Prepared.

Table 4 presents the properties of the five pseudo components calculated by the model, as well as the binary interaction coefficient between CO_2 and UnalMed crude, which were tuned to provide a closer approximation to the experimental data.

Figure 2. CO_2 solubility in UnalMed crude represented by five pseudo components at 80°F.
Source: Prepared.

Table 4.
Physical properties of five pseudo components and CO_2- UnalMed crude interaction

Pseudo-component	M (g/mol)	T_c (ºK)	P_c (MPa)	ω	V_c (cm^3/mol)	k_{ij}
PSC 1	233.19	676.6	0.17655	0.9955	1324.4	0.0388
PSC 2	436.02	792	0.16773	1.4728	2382.4	0.0536
PSC 3	689.46	899.3	0.18344	1.7227	3602.1	0.0641
PSC 4	926.19	991.7	0.23425	1.7963	4579.7	0.0701
PSC 5	1665	1148	0.16186	2.0489	8230.4	0.0843

Source: Prepared.

Fig. 2 shows the results obtained by applying the model of VLE of CO_2 - UnalMed crude represented with five pseudo components. This figure shows the solubility of CO_2 in the oil at 80 ° F and different pressures.

The results are practically the same to those shown in Fig. 1, which is an indication that modelling the CO_2 - UnalMed crude VLE with four pseudo components is appropriate.

Now, for oil represented by six pseudo components, the same methodology as for the four and five pseudo components was used. In this case, the additional pseudo component represents a portion of the non-distillable fraction with a portion of the asphaltenes fraction, i.e. the resins and asphaltenes. This pseudo component is called transition pseudo component. The pseudo components order is as follows: The first two represent the distillable fraction, the third and fourth pseudo components represent the non-distillable fraction (resins 1 and 2), the fifth represents the transition between the resins and asphaltenes and the sixth pseudo component corresponds to asphaltenes.

Table 5 shows the final input parameters of UnalMed crude represented by six pseudo components, and Table 6 shows the properties calculated for the six pseudo components as well as the binary interaction coefficients between CO_2 and UnalMed crude.

Table 5.
Characterization of six UnalMed crude pseudo components.

Pseudo-component	T_b (ºC)	SG	% Wt
PSC 1	350.00	0.5500	38.023
PSC 2	450.00	0.6300	29.430
PSC 3	550.00	0.7100	19.146
PSC 4	650.00	0.7900	6.392
PSC 5	750.00	0.8700	2.740
PSC 6	850.00	0.9500	4.269

Source: Prepared.
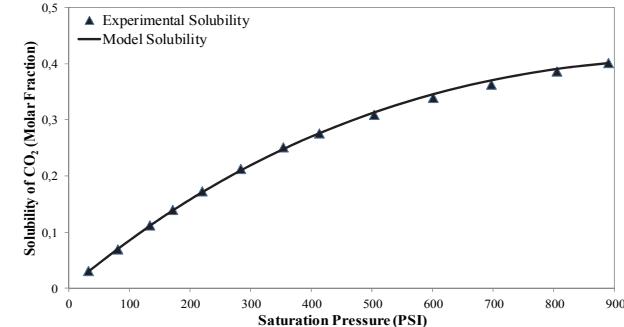
Figure 3. CO_2 solubility in UnalMed crude represented by six pseudo components at 80°F.
Source: Prepared.

Table 6.
Physical properties of six pseudo components and CO_2 - UnalMed crude interaction

Pseudo-component	M (g/mol)	T_c (ºK)	P_c (MPa)	ω	V_c (cm^3/mol)	k_{ij}
PSC 1	233.19	676.6	0.17655	0.9955	1324.4	0.0365
PSC 2	383.52	766.3	0.17479	1.3822	2107.2	0.0475
PSC 3	586.94	858.3	0.17265	1.6488	3126.3	0.057
PSC 4	855.26	952.6	0.1699	1.8339	4428.7	0.0653
PSC 5	1205.9	1049	0.16636	1.962	6090.3	0.0727
PSC 6	1665	1148	0.16186	2.0489	8230.4	0.0794

Source: Prepared

Fig. 3 shows again the same behavior as shown in Fig. 1, 2. In general, results show a high level of agreement with the UnalMed crude experimental data, which indicates that it is possible to model the CO_2 - UnalMed crude VLE through four pseudo components, which can be commonly obtained with SARA analysis.

4. Conclusions

For the calculation of CO_2 - UnalMed crude VLE, the crude representation was tested with different amounts of pseudo components to compare the model results with experimental data.

All crude representations showed very similar results and it is important to note that it is sufficient to have a complete characterization of the SARA analysis and use these four pseudo components to model the VLE.

Having specific data, such as boiling point and specific gravity of each pseudo component, is useful for further reducing the uncertainty of the results.

Binary interaction coefficients, required in the parameters of the cubic EOS are a key element in the study of the solubility of CO_2 in the oil, because the saturation pressure proved to be highly sensitive to the values of binary interaction parameters, thus had to be tuned to obtain a better approximation of the experimental data.

We observed that with a relatively low operating temperature, the solubility of CO_2 in the oil is favored when the system is at high pressures, which is an important factor in the recovery of oil, because this could decrease the viscosity thereof without needing too much energy to heat the contents of the fields.

Crude representation using four pseudo components and the model used were suitable for the analysis of phase behavior in these kinds of systems.

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