Modeling of Transport Properties Using the SAFT-VR Mie Equation of State

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Abstract

Carbon capture and storage (CCS) has been presented as one of the most promising methods to counterbalance the CO_2 emissions from the combustion of fossil fuels. Density, viscosity and interfacial tension (IFT) are, among others properties, crucial for the safe and optimum transport and storage of CO_2-rich steams and they play a key role in enhanced oil recovery (EOR) operations. Therefore, in the present work the capability of a new molecular based equation of state (EoS) to describe these properties was evaluated by comparing the model predictions against literature experimental data.

The EoS considered herein is based on an accurate statistical associating fluid theory with variable range interaction through Mie potentials (SAFT-VR Mie EoS). The EoS was used to describe the vapor-liquid equilibria (VLE) and the densities of selected mixtures. Phase equilibrium calculations are then used to estimate viscosity and interfacial tension values. The viscosity model considered is the TRAPP method using the single phase densities, calculated from the EoS. The IFT was evaluated by coupling this EoS with the density gradient theory of fluids interfaces (DGT). The DGT uses bulk phase properties from the mixture to readily estimate the density distribution of each component across the interface and predict interfacial tension values.

To assess the adequacy of the selected models, the modeling results were compared against experimental data of several CO_2-rich systems in a wide range of conditions from the literature. The evaluated systems include five binaries (CO_2/O_2, CO_2/N_2, CO_2/Ar, CO_2/n-C_4 and CO_2/n-C_{10}) and two multicomponent mixtures (90%CO_2 / 5%O_2 / 2%Ar / 3%N_2 and 90%CO_2 / 6%n-C_4 / 4%n-C_{10}).

The modeling results showed low absolute average deviations to the experimental viscosity and IFT data from the literature, supporting the capabilities of the adopted models for describing thermophysical properties of CO_2-rich systems.

Introduction

Nowadays Carbon capture and storage (CCS) is being studied as one of the most promising process to reduce the CO_2 emissions from burning fossil fuels. Several research projects have been focused in improving the safety and reducing both the costs and the environmental risks associated with CCS operations (Li *et al*. 2011). From a thermodynamic point of view, an accurate description of thermophysical properties is crucial to the design of capture, transport and storage of CO_2. Therefore, the research and the assessment of new equation of state (EoS) models are important for the deployment of more accurate models.

In the oil and gas industry, with regard to the upstream, there is no interest in looking for more accurate EoSs, because in reservoir simulations the cubic EoSs provide enough accuracy due to uncertainties in the geology and the flow in porous media. Likewise, considering the computational time, thanks to their simplicity, cubic EoSs are scarcely replaced by more complex PVT models in reservoir simulations. However in the downstream industry and CCS, more complex thermodynamic models can be applied in order to increase the accuracy of PVT modeling: for example non-cubic EoSs can provide a better density description at high pressures and high temperatures (HPHT) (Yan et al. 2015).

This paper focuses on one of the latest versions of the family of SAFT EoS, the SAFT-VR Mie. The adequacy of this EoS to describe transport properties like viscosity and interfacial tension (IFT), when coupled with appropriated models, has been investigated with results showing a very good agreement predictions and experimental data. The investigated systems include five binaries (CO_2/O_2, CO_2/N_2, CO_2/Ar, CO_2/*n*-C_4 and CO_2/*n*-C_{10}) and two multicomponent mixtures (90%CO_2 / 5%O_2 / 2%Ar / 3%N_2 and 90%CO_2 / 6%*n*-C_4 / 4%*n*-C_{10}) in a wide range of pressure and temperature conditions.

Model Description

Equation of state

The SAFT-VR Mie EoS proposed by Lafitte *et al*. 2013 is one of the latest versions of the SAFT equations of state. The SAFT-VR Mie is featured by the use of the Mie potential to describe the attraction-repulsion interactions between the segments which build the molecules. The Mie potential is defined as:

$$u^{Mie}(r) = \frac{\lambda_r}{\lambda_r - \lambda_a}\left(\frac{\lambda_r}{\lambda_a}\right)^{\frac{\lambda_r}{\lambda_r-\lambda_a}} \varepsilon \left(\left(\frac{\sigma}{r}\right)^{\lambda_r} - \left(\frac{\sigma}{r}\right)^{\lambda_a}\right) \quad \text{(Eq. 1)}$$

where r is radial distance, σ the temperature-independent segment diameter, ε the potential depth; λ_r and λ_a are the repulsive and attractive ranges, respectively. The Mie parameters for the substances studied in this work are reported in the **Table 1**.

Table 1: Molecular parameters of the compounds used in this work (Lafitte et al. 2013)

Substance	m_s	σ/Å	(ε/k)/K	λ_r	λ_a
Carbon dioxide	1.5000	3.1916	231.88	27.557	5.1646
n-butane	1.8514	4.0887	273.64	13.650	6
n-decane	2.9976	4.5890	400.79	18.885	6
Nitrogen	1.4214	3.1760	72.438	9.8749	6
Oxygen	1.4283	2.9671	81.476	8.9218	6
Argon	1.0000	3.4038	117.84	12.085	6

The SAFT-VR Mie EoS can be expressed in terms of the reduced Helmholtz energy as the summation of several contributions:

$$a = \frac{A}{NkT} = a^{IDEAL} + a^{MONO} + a^{CHAIN} + a^{ASSOC} \quad \text{(Eq. 2)}$$

where a^{IDEAL} is the ideal gas contribution, a^{MONO} the monomer contribution, a^{CHAIN} the contribution due to chain formation and a^{ASSOC} the association contribution. The expressions of each contribution can be found in the studies of Lafitte et al. 2013.

Regarding to the phase equilibria, the thermodynamic criterion for phase equilibrium is the equality of chemical potentials of each component in all co-existing phases. Using the SAFT EoS, the chemical potential can be derived as $\mu=(\partial A/\partial N)_{V,T}$ and then used to calculate the fugacity of each component (Chapoy *et al.* 2013). In addition to the pure molecular parameters, temperature independent binary interaction parameters k_{ij} have been adjusted against solubility data (database Knapp et al. 1982) to improve the description of vapor-liquid equilibria. The optimum k_{ij} parameters used in **Eq. 3** are reported in **Table 2**. The VLE predictions using SAFT-VR Mie with the adjusted k_{ij} are presented in the **Fig. 1**. The percentage absolute average deviation (%AAD) between the predicted saturation pressure

and vapor composition and experimental data of Nagarajan are 2.8%*Psat* and 0.9%*y*.

$$\varepsilon_{ij} = \left(1 - k_{ij}\right) \frac{\sqrt{\sigma_i^3 \sigma_j^3}}{\sigma_{ij}^3} \sqrt{\varepsilon_i \varepsilon_j} \qquad \text{(Eq. 3)}$$

Table 2: Binary interaction parameters between O_2, Ar, N_2 and CO_2, and between CO_2, *n*-butane and *n*-decane for the SAFT-VR Mie used in this work.

	N_2	O_2	Ar	CO_2	*n*-C_4	*n*-C_{10}
N_2	0	-0.009	-0.017	-0.127	-	-
O_2	-0.009	0	-0.023	-0.033	-	-
Ar	-0.017	-0.023	0	-0.014	-	-
CO_2	-0.127	-0.033	-0.014	0	0.053	0.057
n-C_4	-	-	-	0.053	0	-0.004
n-C_{10}	-	-	-	0.057	-0.004	0

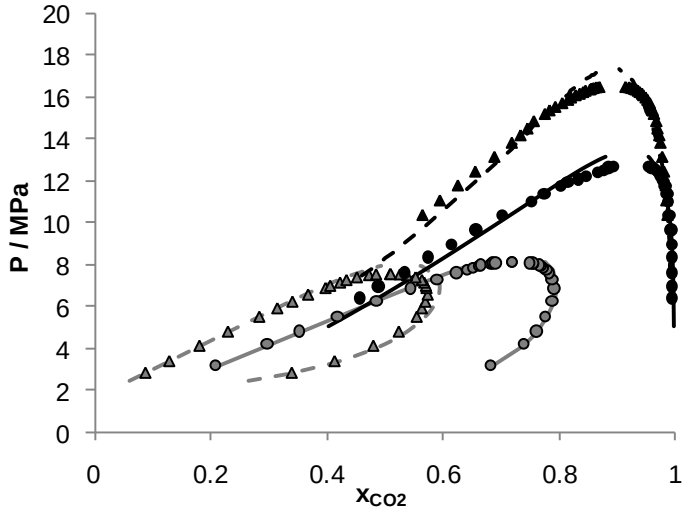


Fig. 1: Predicted VLE by SAFT-VR Mie for CO_2-*n*C_4 (grey) (Nagarajan 1985) and CO_2-*n*C_{10} (black) (Nagarajan 1986). Symbols: (●) T=344.3 and (▲) T=377.6K.

V**iscosity model**

TRAPP (TRAnsport Properties Prediction) is a predictive model based on the extended corresponding states theory (ECS) (K. C. M. and K. E. Gubbins 1976; Hanley and Cohen 1976), used to estimate the viscosity and thermal conductivity of pure fluids and their mixtures over the entire phase range. In the original TRAPP, propane was employed as reference fluid (Ely and Hanley 1983), although several reference fluids can be selected (Huber *et*. 1992). Accordingly, the residual viscosity of the mixture at a corresponding state point (T_0 and ρ_0) is given by:

$$\Delta\eta(\rho,T) = \eta_m - \eta_m^0 = F_{\eta m}[\eta_R - \eta_R^0] + \Delta\eta^{ENSKOG} \qquad \text{(Eq. 4)}$$

where $\eta_m{}^0$ is the viscosity of the mixture at low pressure evaluated by Herning and Zipperer (1936) approximation. The term η_R-$\eta_R{}^0$ is the residual viscosity of the reference fluid which is calculated as (Younglove and Ely 1987):

$$\eta_R - \eta_R^0 = G_1 \exp[\rho_0^{0.1} G_2 + \rho_0^{0.5}(\rho_{r,R} - 1) G_3] - G_1 \quad \text{(Eq. 5)}$$

where η_R is the real viscosity at T_0 and ρ_0 of the reference fluid, $\eta_R{}^0$ is the viscosity at low pressure and T_0, and $\rho_{r,R}$ is the reduced density calculated as $\rho_{r,R} = \rho_0 / \rho_{c,R}$. The corresponding state (T_0 and ρ_0) is calculated as $T_0 = T/f_m$ and $\rho_0 = \rho h_m$. G_1, G_2 and G_3 are parameters that can be calculated as:

$$G_1 = \exp(E_1 + E_2/T) \quad \text{(Eq. 6)}$$

$$G_2 = E_3 + E_4/T^{1.5} \quad \text{(Eq. 7)}$$

$$G_3 = E_5 + E_6/T + E_7/T^2 \quad \text{(Eq. 8)}$$

where the E_i parameters have been correlated for propane as reference fluid and are shown in **Table 3**.

Table 3: E_i parameter TRAPP Model (Poling et al. 2001).

E_1	-14.113294896
E_2	968.22940153
E_3	13.686545032
E_4	-12511.628378
E_5	0.01168910864
E_6	43.527109444
E_7	7659.4543472

In order to calculate the terms $F_{\eta m}$ and $\Delta\eta^{ENSKOG}$, the following mixing rules were applied:

$$h_m = \sum_i \sum_j y_i y_j h_{ij} \quad \text{(Eq. 9)}$$

$$f_m h_m = \sum_i \sum_j y_i y_j h_{ij} f_{ij} \quad \text{Eq. 10)}$$

$$h_{ij} = \frac{\left((h_i)^{1/3} + (h_j)^{1/3}\right)}{8} \quad \text{(Eq. 11)}$$

$$f_{ij} = (f_i f_j)^{1/2} \quad \text{(Eq. 12)}$$

where y_i is the mole fraction of component i; f_i and h_i are functions of the critical parameters and acentric factor defined as:

$$f_i = \frac{T_C}{T_C^R}[1 + (\omega - \omega^R)(0.05203 - 0.7498 \ln T_r)] \quad \text{(Eq. 13)}$$

$$h_i = \frac{\rho_C^R}{\rho_C} \frac{Z_C^R}{Z_C}[1 + (\omega - \omega^R)(0.1436 - 0.2822 \ln T_r)] \quad \text{(Eq. 14)}$$

The viscosity dimensional scaling factor can be calculated as:

$$F_{\eta m} = (M_R)^{-1/2}(h_m)^{-2}\sum_i\sum_j y_i y_j (f_{ij}M_{ij})^{1/2}(h_{ij})^{4/3} \quad \text{(Eq. 15)}$$

with

$$M_{ij} = \frac{2M_iM_j}{M_i + M_j} \quad \text{(Eq. 16)}$$

where M_i denotes the molecular weight.

The expression $\Delta\eta^{ENSKOG}$ is the residual viscosity which takes into account the differences in the molecules size based on the hard sphere assumption (Poling *et al*. 2001):

$$\Delta\eta^{ENSKOG} = \eta_m^{ENSKOG} - \eta_x^{ENSKOG} \quad \text{(Eq. 17)}$$

with

$$\eta_m^{ENSKOG} = \alpha\rho^2\sum_i\sum_j y_i y_j \sigma_{ij}^6 \eta_{ij}^0 g_{ij}^{hs} + \sum_i \beta_i Y_i \quad \text{(Eq. 18)}$$

where the α parameter is α=9.725x10^{-7} and σ_i is the hard-sphere diameter calculated as σ_i=4.771 $h_i^{1/3}$. The radial distribution function of the hard-sphere fluid is given by the following equation (Gubbins and Gray 1972):

$$g_{ij}^{hs} = \frac{1}{(1-\xi)} + \frac{3\xi}{(1-\xi)^2}\Phi_{ij} + \frac{2\xi^2}{(1-\xi)^3}\Phi_{ij}^2 \quad \text{(Eq. 19)}$$

where

$$\Phi_{ij} = \frac{\sigma_i\sigma_j}{2\sigma_{ij}}\frac{\sum_k y_k\sigma_k^2}{\sum_k y_k\sigma_k^3} \quad \text{(Eq. 20)}$$

$$\xi = \frac{\pi}{6}\rho\sum_k y_k\sigma_k^3 \quad \text{(Eq. 21)}$$

To calculate the expression $\Sigma\beta_i Y_i$ it is necessary to solve the linear system of equations of the form:

$$\sum_i B_{ij}\beta_i = Y_i \quad \text{(Eq. 22)}$$

with

$$Y_i = y_i\left[1 + \frac{18\pi}{15}\rho\sum_j y_j\frac{M_j}{M_i + M_j}\sigma_{ij}^3 g_{ij}^{hs}\right] \quad \text{(Eq. 23)}$$

$$B_{ij} = 2\sum_k y_i y_k \frac{g_{ik}}{\eta_{ik}^0}\left(\frac{M_k}{M_i + M_k}\right)^2\left[\left(1 + \frac{5M_i}{3M_k}\right)\delta_{ij} - \frac{2M_i}{3M_k}\delta_{jk}\right] \quad \text{(Eq. 24)}$$

where δ_{ij} is the Kronecker function which is a delta function (*i.e.*, δ_{ij} =1 if i=j and δ_{ij} =0 in other cases).

Interfacial tension model

The density gradient theory (DGT) has been used to compute the interfacial tension. This model is based

on the square gradient of van der Waals (Rowlinson 1979) and on the reformulation of Cahn and Hilliard 1958 to compute interfacial tension values from bulk phase properties such as density and composition. The DGT, when coupled with appropriate thermodynamic models, has been successfully applied in the prediction of interfacial properties of a wide class of systems and interfaces. The reader is referred to the study of Pereira et al. (2015) and references within for more details. In this work, the SAFT-VR Mie EoS was used to estimate the bulk equilibrium of the investigated systems and DGT used to predict interfacial properties. The main equations within the DGT framework are given below.

In summary, by applying the minimization criterion of the Helmholtz energy to planar interfaces, the interfacial tension values with respects to the density of a reference component is given by Miqueu *et al.* 2004 and 2005:

$$IFT = \int_{\rho_{ref}^{V}}^{\rho_{ref}^{L}} \sqrt{2\Delta\Omega(\rho)\sum_i\sum_j c_{ij}\frac{d\rho_i}{d\rho_{ref}}\frac{d\rho_j}{d\rho_{ref}}}\,d\rho_{ref} \qquad \text{(Eq. 25)}$$

where ρ^{L}_{ref} and ρ^{V}_{ref} are the bulk phase densities and the *ref* subscript denotes the reference component of the mixture. $\Delta\Omega$ is the variation of the grand thermodynamic potential which is related to the Helmholtz free energy by the following equation:

$$\Delta\Omega = f_0(\rho) - \sum_i \rho_i\mu_i + p \qquad \text{(Eq. 26)}$$

where f_0 is the Helmholtz free energy density of the homogeneous fluid at local density, μ_i are the chemical potential of each component and p is the pressure at equilibrium. The methodology described by Miqueu *et al.* 2004 was here followed for determining the density distribution of each component across the interface.

In **Eq. 25**, c_{ij} is the cross influence parameter and the mixing rule used based on the geometric mean of the pure component influence parameters (Carey 1979) and it is given by

$$c_{ij} = (1+\beta_{ij})\sqrt{c_i c_j} \qquad \text{(Eq. 27)}$$

where β_{ij} is the binary interaction parameter and c_i and c_j are the pure component influence parameters. In this work, the binary parameter β_{ij} has been fixed to 0, making the calculation of the interfacial tension of the systems investigated fully predictive. The influence parameters c_i and c_j can be derived from theoretical expressions (Freeman and McDonald 1973). Nevertheless, the parameters are generally correlated using Surface Tension (ST) data from pure substances, by rewriting Eq. 25 for c_i as follows:

$$c_i = \frac{1}{2}\left[\frac{ST}{\int_{\rho^V}^{\rho^L}\sqrt{f_0 - \rho\mu + p}\,d\rho}\right]^2 \qquad \text{(Eq. 28)}$$

There are different approaches for calculating the influence parameters of each pure component (Miqueu *et al.* 2004). In this work, c_i has been taken as a constant value calculated from ST data far from the critical point (Lafitte et al. 2010). Hence, the influence parameters used were adjusted against ST data (NIST webbook 2013) of the pure components at a reduced temperature t_r=0.7. The parameters used are reported in **Table 4**.

Table 4: Density gradient theory influence parameters used in Eq. 26.

Substance	c_i / J·m^5·mol^{-2}

CO_2	2.37×10^{-20}
n-butane	1.68×10^{-19}
n-decane	9.51×10^{-19}

Results

Viscosity

The viscosity of CO_2 systems was calculated using the TRAPP model and the densities computed from the SAFT-VR Mie EoS. A literature review of the available experimental data on the viscosity of some mixtures of relevance for CCS operations was carried and the results listed in **Table 5**. As can be seen in Table 5, experimental data on this property are available in a broad range of temperatures but, in most studies, pressure is limited to either atmospheric pressure or to a maximum pressure of 2.6MPa. Only in the study of Chapoy et al. 2013 this property has been investigated for temperatures in the range (273-423) K and pressures up to 150MPa. The viscosity studies were conducted in the single phase region, *i.e.* Gas (G) phase at low pressures, Liquid (L) above saturation pressure or in the supercritical (SC) region, as described in Table 5.

Table 5: Literature experimental data for the viscosity of CO_2-mixtures with N_2, O_2, and Ar.

Source	System	Phase	T/K	P/MPa	N	Uncertainty
Kestin and Leidenfrost (1959)	CO_2/N_2, CO_2/N_2	G	293	0.1-2.17	28	±0.05%
Kestin et al. (1966)	CO_2/N_2, CO_2/Ar	G	293-304	0.1-2.6	83	±0.1%
Gururaja et al. (1967)	CO_2/N_2, CO_2/O_2	G	298	0.1	17	
Kestin and Ro (1974)	CO_2/N_2, CO_2/Ar, CO_2/N_2/Ar	G	297-773	0.1	64	±0.15%
Kestin et al. (1977)	CO_2/O_2	G	298-674	0.1	10	±0.3%
Hobley et al. (1989)	CO_2/Ar	G	301-521	0.1	24	<0.7%
Chapoy et al. (2013)	$CO_2/N_2/O_2$/Ar	G/L/ SC	273-423	1.5-150	38	
				Total	**264**	

The modeling results for the viscosity of the CO_2-rich systems are shown in **Table 6**, along with the percentage absolute average deviation (%AAD) between predicted and experimental viscosity values for each studied system. A total of 264 experimental points were considered and an overall %AAD of 2.68 % was calculated for the viscosity values predicted with the TRAPP+SAFT-VR Mie model. This value represents half of the deviation reported by Huber (1996) and Poling et al. (2001) and obtained with the TRAPP model combined with a modified BWR-EoS. Nonetheless, the largest deviations with the TRAPP+SAFT-VR Mie model were obtained at high pressures and low temperatures, as depicted in **Fig. 2**, with a maximum deviation of 9.3% for the multicomponent system (90%CO_2, 5%O_2, 2%Ar and 3%N_2).

Table 6: Absolute average deviation of the viscosity calculations.

System	x_{CO2}	N	T/K	P/MPa	%AAD
CO_2/N_2	0.19-0.90	95	293-773	0.1-2.6	3,12
CO_2/Ar	0.22-0.92	96	293-773	0.1-2.6	1,28
CO_2/O_2	0.19-0.92	19	298-674	0.1	3,25

$CO_2/N_2/Ar$	0.25-0.53	16	297-673	0.1	1,39
$CO_2/N_2/O_2/Ar$	0.9	38	273-423	1.5-150	5.15
				Total	**2.68%**

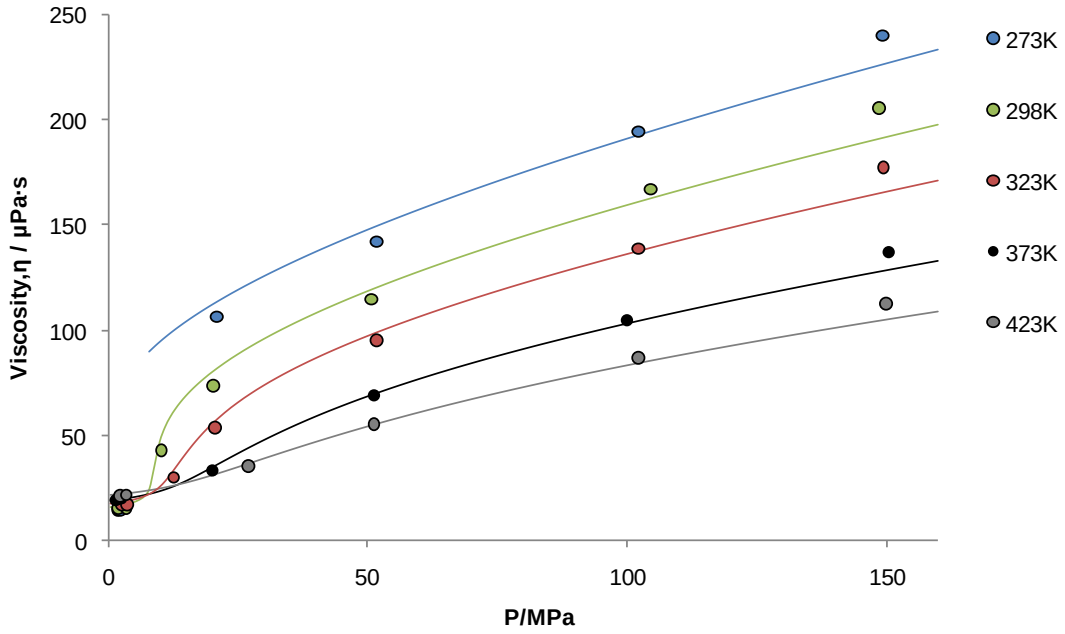


Fig. 2: Experimental (Chapoy et al. 2013) and predicted viscosity of a CO_2-rich system (90%CO_2, 5%O_2, 2%Ar and 3%N_2).

Interfacial tension

The saturated densities and IFT of CO_2-rich systems were modeled by coupling the DGT with the SAFT-VR Mie EoS. The results for the binaries CO_2-*n*-C_4 and CO_2-*n*-C_{10} and the ternary CO_2-*n*-C_4-*n*-C_{10} are plotted in **Fig. 3** and **Fig. 4**, respectively. As can be seen in Fig. 3 and Fig. 4, the DGT+SAFT-VR Mie predictions are in excellent agreement with the measured saturated density and IFT data of the systems considered. Largest deviations were observed close to the critical point, where the SAFT-VR Mie fails to capture the impact of pressure on the density of the bulk phases. Nonetheless, as can be seen in **Figure 5**, the maximum absolute deviation between predicted and measured IFT data was of 0.26mNm^{-1}. In general, the IFT values for the CO_2-*n*-C_4 system and the ternary system are slightly overestimated, whereas an inverse behavior was observed for the CO_2-*n*-C_{10} system.

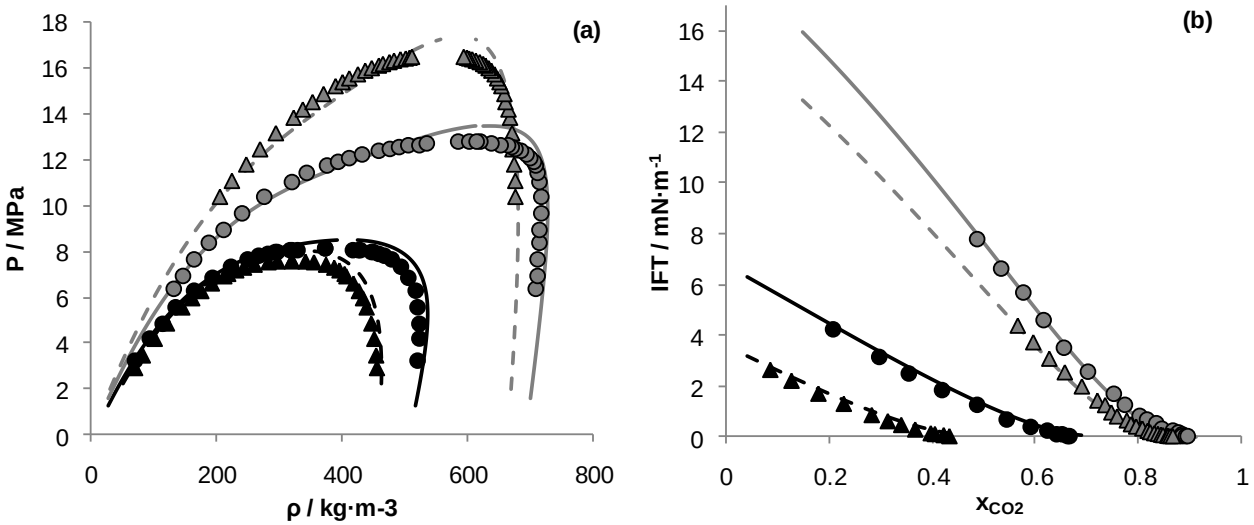


Fig. 3: Predicted density (a) and IFT (b) of CO_2-nC_4 (black) (Nagarajan 1985) [ref] and CO_2-nC_{10} (grey) (Nagarajan 1986). Symbols: (●) T=344.3 and (▲) T=377.6K.

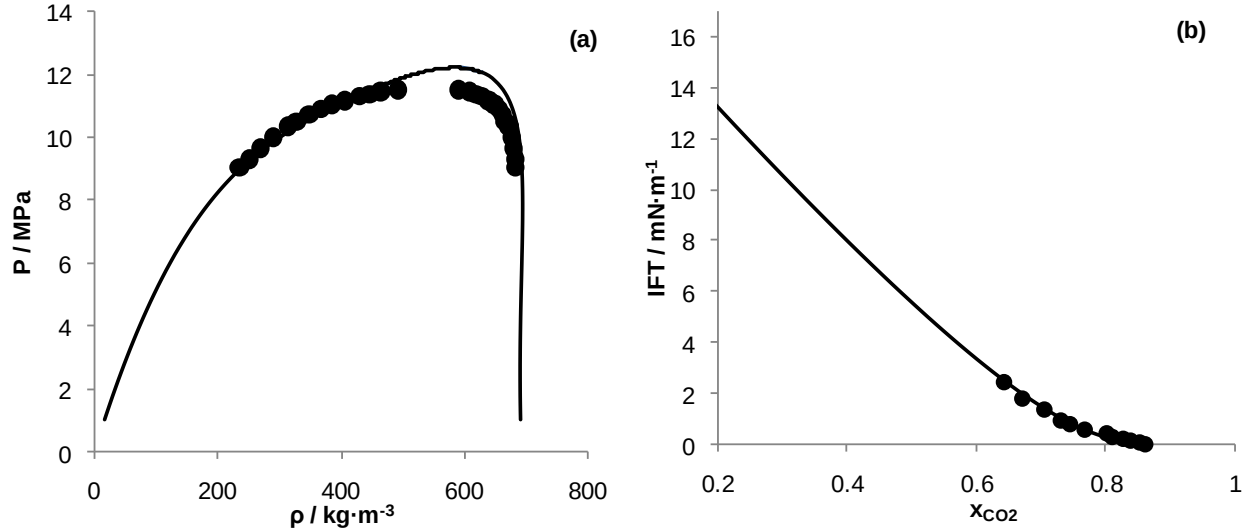


Figure 4: Predicted density (a) and IFT (b) of CO_2-nC_4-nC_{10} mixture at 344.3K (Nagarajan 1990).

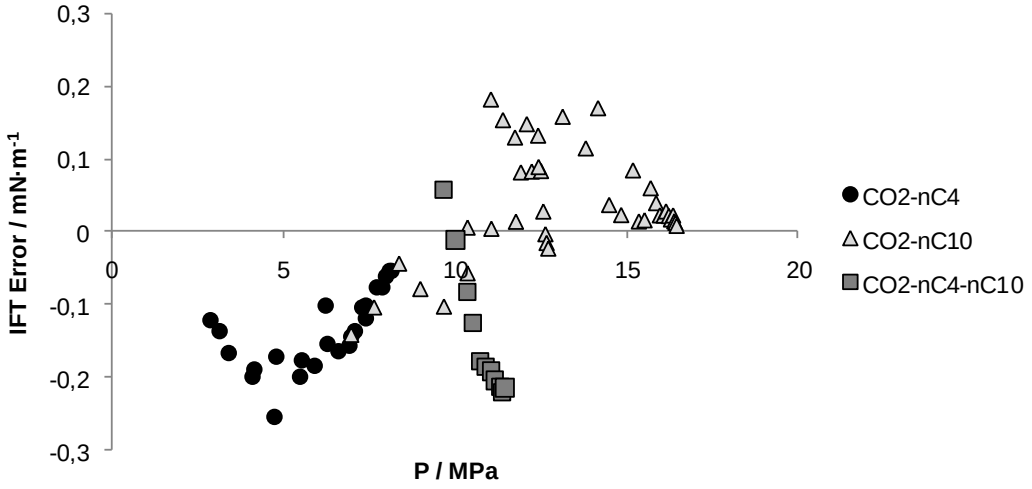


Fig. 5: Absolute deviation between prediction and experimental IFT data ($IFTerror=IFT^{EXP}-IFT^{CALC}$).

In addition, the density profiles through the interface were computed with the DGT approach for the ternary system (90%CO_2, 6%*n*-butane and 4%*n*-decane) and the results plotted in **Fig. 5**. As depicted in **Fig. 5(a)**, the density profiles show a local enrichment of the interface with CO_2 molecules as a peak was noticeable in the density profile of carbon dioxide. The location of peak closer to the vapor side of the interface suggests that the absorption of CO_2 occurred at the surface of the liquid hydrocarbon phase. Furthermore, as can be seen in **Fig. 5(b),** the pressure increase lead to an increase of the interface thickness in this mixture and to a reduction of the local accumulation on light components, such as CO_2. Comparable results were obtained for the binary mixtures and the results are in agreement with those obtained by Miqueu et al. 2005.

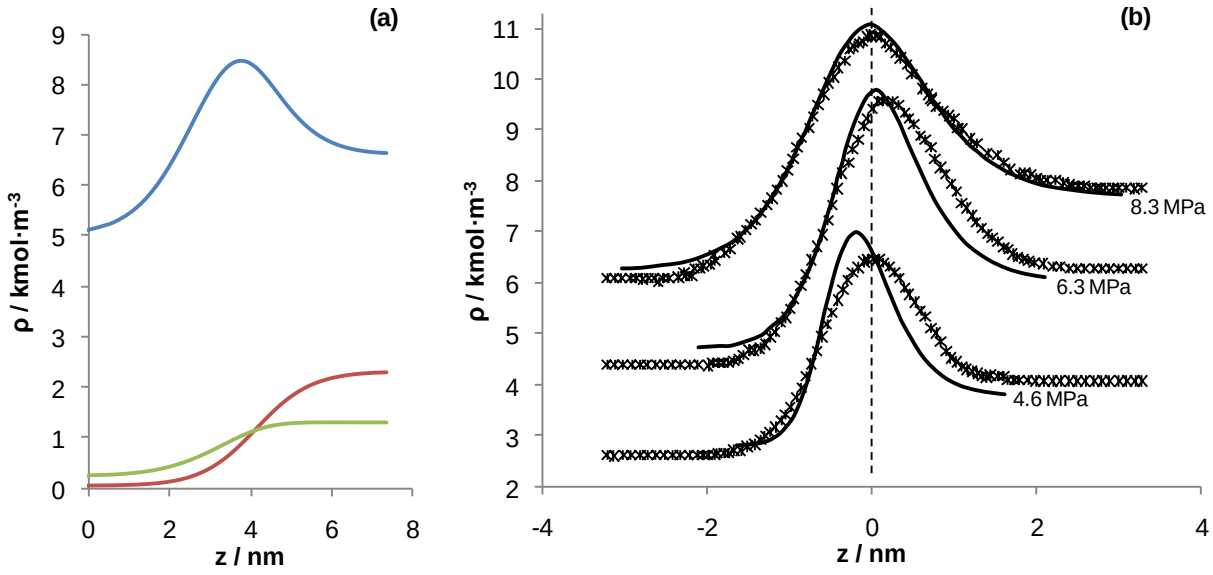


Fig. 5: (a) Density profiles calculated for each component in the CO_2-*n*C_4-*n*C_{10} mixture at 344.3K and 9.31MPa: CO2-(blue), n-butane (green) and n-decane (red). Total interface length of 7.36 nm. (b) Density profiles of carbon dioxide calculated for the CO_2-*n*C_{10} mixture at 344.3K at different pressures against the results of molecular simulation at same conditions (Müller and Mejía 2009).

Conclusions

The TRAPP model and the density gradient theory have been combined with the SAFT-VR Mie EoS to model the viscosity and IFT of CO_2-rich systems. The low deviations observed to measured density data showed the SAFT-VR Mie EoS good capabilities to describe the equilibrium of the studied systems with k_{ij} presented in this work. Furthermore, by using the computed bulk phase properties, the TRAPP and DGT models were able to predict the viscosity and IFT of CO_2-rich mixture with relative low deviations to experimental data, endorsing the capability of the methods for estimating key transport properties of fluids in CCS streams.

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