

Comparative Study of Equations of State for the Dew Curves Calculation in High Pressure Natural Gas Mixtures*

Estudio Comparativo de Ecuaciones de Estado para el Cálculo de Curvas de Rocío en Mezclas de Gas Natural a Alta Presión*

Estudo Comparativo de Equações de Estado para o Cálculo de Curvas de Orvalho em Misturas de Gás Natural de alta Pressão*

Natalia Prieto Jiménez **

Germán González Silva ***

Alex Chaves Guerrero****

Universidad Industrial de Santander– Colombia

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**Candidata a Doctor en Ingeniería Química de la Universidad Industrial de Santander, Colombia. Filiación: Grupo de Investigación en Energía y Medio ambiente de la Universidad Industrial de Santander. Correo electrónico: natispj@gmail.com. Orcid: <https://orcid.org/0000-0001-9178-7758>

***Doctor en Ingeniería Química de la Universidade Estadual de Campinas (Brasil) y Profesor Asistente de la Escuela de Ingeniería de Petróleos de la Universidad Industrial de Santander. Filiación: Grupo de Modelamiento de Procesos de Hidrocarburos – Universidad Industrial de Santander. Correo electrónico: germangsilva@gmail.com Orcid: <https://orcid.org/0000-0002-4642-1092>

**** Doctor en Ingeniería Química- universidad Industrial de Santander. GIEMA: Grupo de Investigación en Energía y Medio Ambiente - Universidad Industrial de Santander. Bucaramanga-Colombia ORCID ID: <https://orcid.org/0000-0003-4481-4818>

Abstract

The success during the operation of natural gas processing plants depends on the correct estimation of thermodynamic properties of the system. This paper calculates the equilibrium curves of real and synthetic natural gas mixtures means of three Equations of State (EOS). These equilibrium curves were constructed and compared with experimental data found in the literature covered. The results showed that, above 4 MPa the Peng-Robinson equation presented a considerable deviation with respect to the experimental data,

reaching an absolute error of 4.36%; therefore, the GERG2008 equation is recommended for systems that operate at high pressures when the components present in the mixture apply.

Keywords:

Gas Mixtures, Dew curves, Equations of State; Peng-Robinson, Soave-Redlich-Kwong, GERG2008.

Resumen

El éxito durante la operación de plantas de tratamiento de gas natural depende de la correcta estimación de las propiedades termodinámicas del sistema. Este artículo calcula las curvas de equilibrio de mezclas de gas natural reales y sintéticas por medio de tres ecuaciones de estado (EOS). Estas curvas de equilibrio fueron construidas y comparadas con datos experimentales presentes en la literatura. Los resultados mostraron que, por encima de 4 MPa la ecuación de Peng-Robinson presentó una desviación considerable con respecto a los datos experimentales, alcanzando un error absoluto de 4,36%; por lo cual se recomienda la ecuación de GERG2008 para sistemas que operen a altas presiones cuando los componentes presentes en la mezcla apliquen.

Palabras clave:

Mezclas de gas, Curvas de rocío, Ecuaciones de estado, Peng-Robinson, Soave-Redlich-Kwong, GERG2008.

Resumo

O sucesso na operação de usinas de tratamento de gás natural depende da correta estimação das propriedades termodinâmicas do sistema. Este artigo calcula as curvas de equilíbrio de misturas de gás natural reais e sintéticas por meio de três equações de estado (EOS). As curvas de equilíbrio foram construídas e comparadas com dados experimentais presentes na literatura. Os resultados mostraram que, acima de 4 Mpa a equação de Peng-Robinson apresentou um desvio considerável em relação aos dados experimentais, atingindo um erro absoluto de 4,36%; por tanto, é recomendável a equação de GERG2008 para sistemas que operam

em alta pressão quando os componentes presentes no sistema apliquem.

Palavras-chave:

Misturas de gás, Curvas de orvalho, Equações de estado, Peng-Robinson, Soave-Redlich-Kwong, GERG2008.

Introduction

Natural gas is a complex mixture of hydrocarbons, composed mainly of methane (CH_4), with significant amounts of ethane (C_2H_6), propane (C_3H_8), butane (C_4H_{10}) and some traces of heavy hydrocarbons and certain inorganic compounds (Mokhatab, Poe, & Mak, 2015). Its use as a fossil fuel mostly depends on the treatment carried out before being sent to the transport network. The processing carried out on natural gas is divided into three stages: separation of the present phases; dew point adjustment and re-compression (Khanwelkar, 2015). Conventional processes of natural gas phases separation are carried out at operating pressures near atmospheric, thus the third stage is essential (Figure 1). According to data from Colombian natural gas plants, 12% of Operating Expenditures (OPEX) is exclusively directed to compression systems. The high energy costs generated by the re-compression led several authors to recommend that phase separation be carried out at high pressure (Austheim, Gjertsen, & Hoffmann, 2008; Brigadeau, 2007; Zaghoul, 2006). However, phase separation at high pressure is a complex process. As the operating pressure within the equipment increases, the density difference between the liquid phase and the gas phase considerably decreases, as the surface tension does, which leads to the condensation of fine droplets and therefore they are easily re-entrainment by the gas phase (Kharoua, Khezzer, & Saadawi, 2013; Laleh, Svrcek, & Monnery, 2012).

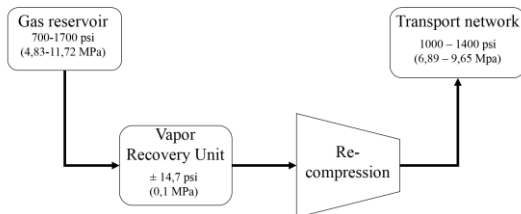


Figure 1. Schematic diagram of the conventional gas-liquid separation process.

For the proper design and analysis of separation systems and high-pressure natural gas processing plants, it is important to know the thermodynamic properties of the mixture, which can be calculated by means of Equations of State (EOS) or empirical correlations (Guo & Ghalambor, 2014; Shoaib, Bhran, Awad, El-Sayed, & Fathy, 2018). The most relevant properties of the mixture are revealed by the thermodynamic equilibrium curves, showing, in addition to the bubble and dew points, the zone where the liquid and gas phases coexist in equilibrium and, consequently, the range of operating conditions of pressure and temperature over which multiphase separation is possible (Jia, Wu, Li, & He, 2017). The cubic equations of Soave-Redlich-Kwong – SRK (Soave, 1972) and Peng-Robinson – PR (Peng & Robinson, 1976) are two EOS widely accepted and used at academic and industrial level for the calculation of thermodynamic properties of natural gas. Additionally, Kunz and Warner (Kunz & Wagner, 2012) developed a specific equation of state for natural gas, called GERG2008 (by the acronym of *Groupe Européen de Recherches Gazières* - European Gas Research Group and the year it was developed) that is based on the concept of multi-fluid and it is explicit for Helmholtz free energy. The correct estimation of the thermodynamics properties of two-phase and three-phase mixtures results in an important input parameter for the development of different processes in the *Oil&Gas* industry using numerical simulation through Computational Fluid Dynamics (CFD) techniques (González-Silva, Matos, Martignoni, & Mori, 2012; Jiménez, Hodapp, Silva, & Mori, 2010; Silva, Jiménez, & Salazar, 2012; Silva, Prieto, & Mercado, 2018).

Cubic Equations of State

The simple mathematical structure of the cubic equations of state allows its practical use in technical and academic applications (Galatro & Marín-Cordero, 2014). These two equations are characterized by having the same general form (Poling, Prausnitz, & Connell, 2000a), which can be expressed explicit in the pressure as shown in the equation (1) or depending on the compressibility factor (equations (2) to (4)):

$$P = \frac{RT}{V-b} - \frac{a}{V^2 + uV + wb^2} \quad (1)$$

$$Z^3 - (1 + B^* + uB^*)Z^2 + (A^* + wB^{*2} - uB^* - uB^{*2})Z - A^*B^* - wB^{*2} - wB^{*3} = 0 \quad (2)$$

$$A^* = \frac{aP}{R^2T^2} \quad (3)$$

$$B^* = \frac{bP}{RT} \quad (4)$$

Where P is pressure; T , temperature; V , molar volume of the mixture and R , the universal constant of gases in corresponding units. The value of the parameters u , w , a and b are defined according to the critical and reduced properties of the mixture (P_c, T_c, T_r) and Pitzer's acentric factor ω (Pitzer & Curl Jr, 1957), as shown in Table 1.

• Mixing rules

The parameters a and b of cubic equations of state require mixing rules. The parameter b represents the effective molar volume, in the case of mixtures containing molecules of approximately equal size; it is possible to express the mixing rule according to equation (5)

$$b = \sum_i x_i b_i \quad (5)$$

Table 1. Parameters used in cubic equations of state (Poling, Prausnitz, & Connell, 2000b).

Equation	Parameters
SRK	$u = 1$ $w = 0$ $b = \frac{0.08664RT_c}{P_c}$ $a = \frac{0.42748R^2T_c^2}{P_c} \left[1 + f_\omega \left(1 - T_r^{1/2} \right) \right]^2$ $f_\omega = 0.48 + 1.574\omega - 0.176\omega^2$
PR	$u = 2$ $w = -1$ $b = \frac{0.0778RT_c}{P_c}$ $a = \frac{0.45724R^2T_c^2}{P_c} \left[1 + f_\omega \left(1 - T_r^{1/2} \right) \right]^2$ $f_\omega = 0.3746 + 1.542\omega - 0.2699\omega^2$

Where x is used as a variable of generic composition for both liquid and vapor roots. The parameter a represents the molecular interactions of the mixture, therefore, its relationship with the internal energy of the mixture (U) must be carefully considered. The generating function for internal energy indicates the following relationship (Elliott & Lira, 1999):

$$\frac{U - U^{gi}}{RT} = -\frac{ap}{RT} \quad (6)$$

In a binary mixture there are three types of interactions for molecules 1 and 2: the molecule interacting with itself (1-1, 2-2) or interacting with each other (1-2). For a random fluid, the probability of finding the molecule (1) is the fraction x_1 . The probability of a 1-1 interaction is called conditional probability, this probability is calculated as the product of the individual probabilities x_1^2 . The probability of interaction 1-2 or 2-1 is x_1x_2 . If the attraction interactions are characterized by a_{11} , a_{22} and a_{12} ; the mixing rule for a is given by equation (7)

$$U - U^{gi} = -ap = -\rho(x_1^2a_{11} + 2x_1x_2a_{12} + x_2^2a_{22}) \quad (7)$$

The simplified form of equation (7) is represented by equations (8) and (9):

$$a = \sum_i \sum_j x_i x_j a_{ij} \quad (8)$$

$$a_{ij} = (a_i a_j)^{1/2} (1 - k_{ij}) \quad (9)$$

Where k_{ij} represents the binary combination parameter, which is tabulated in the literature.

Approach based on the multi-fluid concept

The multi-fluid approach uses equations of state in their fundamental form for each component of the mixture, in conjunction with functions developed for the binary mixtures between the components, in order to determine the residual behavior of the mixture (Kunz & Wagner, 2012). The GERG2008 EOS allows to calculate the thermodynamic properties of 21 components of natural gas and their binary mixtures as shown in Table 2. The Multi-Fluid Helmholtz-Free-Energy-Explicit approach, A which is a function of density (ρ), temperature (T) and the molar composition vector $\vec{x}$ contains the general form of the equation (10):

$$A(\rho, T, \vec{x}) = A^o(\rho, T, \vec{x}) + A^r(\rho, T, \vec{x}) \quad (10)$$

For modeling, the dimensionless Helmholtz free energy was used, as follows:

$$\alpha(\delta, \tau, \vec{x}) = \alpha^o(\rho, T, \vec{x}) + \alpha^r(\delta, \tau, \vec{x}) \quad (11)$$

Where α^o corresponds to the Helmholtz free energy component of the mixture as ideal gas, depending on $\rho, T, \vec{x}$:

$$\alpha^o(\rho, T, \vec{x}) = \sum_{i=1}^N x_i [\alpha_{oi}^o(\rho, T) + \ln x_i] \quad (12)$$

The term α^r is the residual component of Helmholtz free energy of the mixture, depending on $\delta, \tau, \vec{x}$.

$$\alpha^r(\delta, \tau, \vec{x}) = \sum_{i=1}^N x_i \alpha_{oi}^r(\delta, \tau) + \Delta \alpha^r(\delta, \tau, \vec{x}) \quad (13)$$

$$\alpha \equiv \frac{a}{RT} \quad (14)$$

$$\delta \equiv \frac{\rho}{\rho_r(\vec{x})} \quad (15)$$

$$\tau \equiv \frac{T_r(\vec{x})}{T} \quad (16)$$

Where δ is the reduced density of the mixture, τ is the inverse of the reduced temperature of the mixture and N is the maximum number of present components. α_{oi}^r is the residual part of the Helmholtz free energy reduced from component i and $\Delta\alpha^r$ it is the generating function. Equation (13) takes into account the residual behavior of the mixture, evaluated at the reduced properties δ and τ . Another way to express the residual Helmholtz free energy is through equation (17); where the first term corresponds to the linear contribution of the reduced residual Helmholtz free energy, multiplied by the molar fractions x_i . The double sum of the next term is equivalent to the generating function.

$$\begin{aligned} \alpha^r(\delta, \tau, \vec{x}) &= \sum_{i=1}^N x_i \alpha_{oi}^r(\delta, \tau) \\ &+ \sum_{i=1}^{N-1} \sum_{j=i+1}^N x_i x_j F_{ij} \alpha_{ij}^r(\delta, \tau) \end{aligned} \quad (17)$$

The dimensionless form of Helmholtz free energy as the ideal gas of component i in equation (12) is defined as:

$$\begin{aligned} \alpha_{oi}^o(\rho, T) &= \ln\left(\frac{\rho}{\rho_{c,i}}\right) \\ &+ \frac{R^*}{R} \left[n_{oi,1}^o + n_{oi,2}^o \frac{T_{c,i}}{T} + n_{oi,3}^o \ln\left(\frac{T_{c,i}}{T}\right) \right] \\ &+ \sum_{k=4,6} n_{oi,k}^o \ln\left(\left| \sinh\left(v_{oi,3}^o \frac{T_{c,i}}{T}\right) \right|\right) \\ &- \sum_{k=5,7} n_{oi,k}^o \ln\left(\left| \cosh\left(v_{oi,3}^o \frac{T_{c,i}}{T}\right) \right|\right) \end{aligned} \quad (18)$$

Where $\rho_{c,i}$ and $T_{c,i}$ are the critical parameters of the pure components. For a specific binary generating function, the adjustable factor F_{ij} normally it is represented as one (1) if the mixture exists and zero (0) if it does not exist. The residual part of the

Helmholtz free energy for the compound as the ideal gas is represented by the equation (19).

Table 2. Components of natural gas and its specific EOS of the pure component (Kunz & Wagner, 2012).

Comp.	EOS Ref.	Range of validity	
		T^*	P^*
Main components			
CH_4	(Klimeck, 2000)	90 - 623	300
N_2	(Klimeck, 2000)	63 - 700	300
CO_2	(Klimeck, 2000)	216 - 900	300
C_2H_6	(Klimeck, 2000)	90 - 623	300
Secondary alkanes			
C_3H_8	(Span & Wagner, 2003)	85 - 623	100
nC_4H_{10}	(Span & Wagner, 2003)	134 - 693	70
iC_4H_{10}	(Span & Wagner, 2003)	113 - 573	35
nC_5H_{12}	(Span & Wagner, 2003)	143 - 573	70
iC_5H_{12}	(Lemmon & Span, 2006)	112 - 500	35
nC_6H_{14}	(Span & Wagner, 2003)	177 - 548	100
nC_7H_{16}	(Span & Wagner, 2003)	182 - 523	100
nC_8H_{18}	(Span & Wagner, 2003)	216 - 548	100
nC_9H_{20}	(Lemmon & Span, 2006)	219 - 600	800
$nC_{10}H_{22}$	(Lemmon & Span, 2006)	243 - 675	800
Other secondary components			
H_2	(Kunz & Wagner, 2012)	14 – 700	300
O_2	(Span & Wagner, 2003)	54 – 303	100
CO	(Lemmon & Span, 2006)	68 – 400	100
H_2O	(Kunz & Wagner, 2012)	273 – 1273	100
H_2S	(Lemmon & Span, 2006)	187 – 760	170
He	(Kunz & Wagner, 2012)	2,2 – 573	100
Ar	(Span & Wagner, 2003)	83 - 520	100

* T in K and P in MPa

$$\begin{aligned} \alpha_{oi}^r(\delta, \tau) &= \sum_{k=1}^{K_{Pol,i}} n_{oi,k} \delta^{d_{oi,k}} \tau^{t_{oi,k}} \\ &+ \sum_{k=K_{Pol,i}+1}^{K_{Exp,i}} n_{oi,k} \delta^{d_{oi,k}} \tau^{t_{oi,k}} \exp(-\delta^{C_{oi,k}}) \end{aligned} \quad (19)$$

The function $\alpha_{oi}^r(\delta, \tau)$ of equation (17) depends only on the reduced variables of the mixture δ and τ , it is represented in equation (20):

$$\begin{aligned} \alpha_{ij}^r(\delta, \tau) &= \sum_{k=1}^{K_{Pol,ij}} n_{ij,k} \delta^{d_{ij,k}} \tau^{t_{ij,k}} \\ &+ \sum_{k=K_{Pol,i}+1}^{K_{Exp,i}} n_{oi,k} \delta^{d_{oi,k}} \tau^{t_{oi,k}} \exp \left[-\eta_{ij,k} (\delta - \varepsilon_{ij,k})^2 - \beta_{ij,k} (\delta - \gamma_{ij,k}) \right] \end{aligned} \quad (20)$$

The reduced variables of the mixture δ and τ are calculated from equations (15) and (16) respectively by means of dependent functions of the composition, as shown in the equations (21) and (22). The binary parameters $\beta_{v,ij}$ and $\gamma_{v,ij}$ of equation (21) and $\beta_{T,ij}$ and $\gamma_{T,ij}$ of equation (22), as well as the critical parameters $\rho_{c,i}$ and $T_{c,i}$ are tabulated in Kunz and Wagner (Kunz & Wagner, 2012).

Mixing Rules

For the binary mixtures that are formed from the 21 components of the GERG2008 model, five different mixing rules are used, shown in Figure 2.

$$\begin{aligned} \frac{1}{\rho_r(\vec{x})} &= \sum_{i=1}^N x_i^2 \frac{1}{\rho_{c,i}} \\ &+ \sum_{i=1}^{N-1} \sum_{j=i+1}^N 2x_i x_j \beta_{v,ij} \gamma_{v,ij} \frac{x_i + x_j}{\beta_{v,ij}^2 x_i + x_j} \\ &\cdot \frac{1}{8} \left(\frac{1}{\rho_{c,i}^3} + \frac{1}{\rho_{c,j}^3} \right)^3 \end{aligned} \quad (21)$$

$$\begin{aligned} T_r(\vec{x}) &= \sum_{i=1}^N x_i^2 T_{c,i} \\ &+ \sum_{i=1}^{N-1} \sum_{j=i+1}^N 2x_i x_j \beta_{T,ij} \gamma_{T,ij} \frac{x_i + x_j}{\beta_{T,ij}^2 x_i + x_j} (T_{c,i}, T_{c,j})^{\frac{1}{2}} \end{aligned} \quad (22)$$

	He	H ₂	H ₂ O	CO	H ₂	O ₂	Ar	nC ₄ H ₁₀	nC ₅ H ₁₂	nC ₆ H ₁₄	nC ₇ H ₁₆	nC ₈ H ₁₈	nC ₉ H ₂₀	nC ₁₀ H ₂₂	nC ₁₁ H ₂₄	nC ₁₂ H ₂₆	nC ₁₃ H ₂₈	nC ₁₄ H ₃₀	nC ₁₅ H ₃₂	nC ₁₆ H ₃₄	nC ₁₇ H ₃₆	nC ₁₈ H ₃₈	nC ₁₉ H ₄₀	nC ₂₀ H ₄₂	nC ₂₁ H ₄₄	nC ₂₂ H ₄₆	nC ₂₃ H ₄₈	nC ₂₄ H ₅₀	nC ₂₅ H ₅₂	nC ₂₆ H ₅₄	nC ₂₇ H ₅₆	nC ₂₈ H ₅₈	nC ₂₉ H ₆₀	nC ₃₀ H ₆₂	nC ₃₁ H ₆₄	nC ₃₂ H ₆₆	nC ₃₃ H ₆₈	nC ₃₄ H ₇₀	nC ₃₅ H ₇₂	nC ₃₆ H ₇₄	nC ₃₇ H ₇₆	nC ₃₈ H ₇₈	nC ₃₉ H ₈₀	nC ₄₀ H ₈₂	nC ₄₁ H ₈₄	nC ₄₂ H ₈₆	nC ₄₃ H ₈₈	nC ₄₄ H ₉₀	nC ₄₅ H ₉₂	nC ₄₆ H ₉₄	nC ₄₇ H ₉₆	nC ₄₈ H ₉₈	nC ₄₉ H ₁₀₀	nC ₅₀ H ₁₀₂	nC ₅₁ H ₁₀₄	nC ₅₂ H ₁₀₆	nC ₅₃ H ₁₀₈	nC ₅₄ H ₁₁₀	nC ₅₅ H ₁₁₂	nC ₅₆ H ₁₁₄	nC ₅₇ H ₁₁₆	nC ₅₈ H ₁₁₈	nC ₅₉ H ₁₂₀	nC ₆₀ H ₁₂₂	nC ₆₁ H ₁₂₄	nC ₆₂ H ₁₂₆	nC ₆₃ H ₁₂₈	nC ₆₄ H ₁₃₀	nC ₆₅ H ₁₃₂	nC ₆₆ H ₁₃₄	nC ₆₇ H ₁₃₆	nC ₆₈ H ₁₃₈	nC ₆₉ H ₁₄₀	nC ₇₀ H ₁₄₂	nC ₇₁ H ₁₄₄	nC ₇₂ H ₁₄₆	nC ₇₃ H ₁₄₈	nC ₇₄ H ₁₅₀	nC ₇₅ H ₁₅₂	nC ₇₆ H ₁₅₄	nC ₇₇ H ₁₅₆	nC ₇₈ H ₁₅₈	nC ₇₉ H ₁₆₀	nC ₈₀ H ₁₆₂	nC ₈₁ H ₁₆₄	nC ₈₂ H ₁₆₆	nC ₈₃ H ₁₆₈	nC ₈₄ H ₁₇₀	nC ₈₅ H ₁₇₂	nC ₈₆ H ₁₇₄	nC ₈₇ H ₁₇₆	nC ₈₈ H ₁₇₈	nC ₈₉ H ₁₈₀	nC ₉₀ H ₁₈₂	nC ₉₁ H ₁₈₄	nC ₉₂ H ₁₈₆	nC ₉₃ H ₁₈₈	nC ₉₄ H ₁₉₀	nC ₉₅ H ₁₉₂	nC ₉₆ H ₁₉₄	nC ₉₇ H ₁₉₆	nC ₉₈ H ₁₉₈	nC ₉₉ H ₂₀₀	nC ₁₀₀ H ₂₀₂	nC ₁₀₁ H ₂₀₄	nC ₁₀₂ H ₂₀₆	nC ₁₀₃ H ₂₀₈	nC ₁₀₄ H ₂₁₀	nC ₁₀₅ H ₂₁₂	nC ₁₀₆ H ₂₁₄	nC ₁₀₇ H ₂₁₆	nC ₁₀₈ H ₂₁₈	nC ₁₀₉ H ₂₂₀	nC ₁₁₀ H ₂₂₂	nC ₁₁₁ H ₂₂₄	nC ₁₁₂ H ₂₂₆	nC ₁₁₃ H ₂₂₈	nC ₁₁₄ H ₂₃₀	nC ₁₁₅ H ₂₃₂	nC ₁₁₆ H ₂₃₄	nC ₁₁₇ H ₂₃₆	nC ₁₁₈ H ₂₃₈	nC ₁₁₉ H ₂₄₀	nC ₁₂₀ H ₂₄₂	nC ₁₂₁ H ₂₄₄	nC ₁₂₂ H ₂₄₆	nC ₁₂₃ H ₂₄₈	nC ₁₂₄ H ₂₅₀	nC ₁₂₅ H ₂₅₂	nC ₁₂₆ H ₂₅₄	nC ₁₂₇ H ₂₅₆	nC ₁₂₈ H ₂₅₈	nC ₁₂₉ H ₂₆₀	nC ₁₃₀ H ₂₆₂	nC ₁₃₁ H ₂₆₄	nC ₁₃₂ H ₂₆₆	nC ₁₃₃ H ₂₆₈	nC ₁₃₄ H ₂₇₀	nC ₁₃₅ H ₂₇₂	nC ₁₃₆ H ₂₇₄	nC ₁₃₇ H ₂₇₆	nC ₁₃₈ H ₂₇₈	nC ₁₃₉ H ₂₈₀	nC ₁₄₀ H ₂₈₂	nC ₁₄₁ H ₂₈₄	nC ₁₄₂ H ₂₈₆	nC ₁₄₃ H ₂₈₈	nC ₁₄₄ H ₂₉₀	nC ₁₄₅ H ₂₉₂	nC ₁₄₆ H ₂₉₄	nC ₁₄₇ H ₂₉₆	nC ₁₄₈ H ₂₉₈	nC ₁₄₉ H ₃₀₀	nC ₁₅₀ H ₃₀₂	nC ₁₅₁ H ₃₀₄	nC ₁₅₂ H ₃₀₆	nC ₁₅₃ H ₃₀₈	nC ₁₅₄ H ₃₁₀	nC ₁₅₅ H ₃₁₂	nC ₁₅₆ H ₃₁₄	nC ₁₅₇ H ₃₁₆	nC ₁₅₈ H ₃₁₈	nC ₁₅₉ H ₃₂₀	nC ₁₆₀ H ₃₂₂	nC ₁₆₁ H ₃₂₄	nC ₁₆₂ H ₃₂₆	nC ₁₆₃ H ₃₂₈	nC ₁₆₄ H ₃₃₀	nC ₁₆₅ H ₃₃₂	nC ₁₆₆ H ₃₃₄	nC ₁₆₇ H ₃₃₆	nC ₁₆₈ H ₃₃₈	nC ₁₆₉ H ₃₄₀	nC ₁₇₀ H ₃₄₂	nC ₁₇₁ H ₃₄₄	nC ₁₇₂ H ₃₄₆	nC ₁₇₃ H ₃₄₈	nC ₁₇₄ H ₃₅₀	nC ₁₇₅ H ₃₅₂	nC ₁₇₆ H ₃₅₄	nC ₁₇₇ H ₃₅₆	nC ₁₇₈ H ₃₅₈	nC ₁₇₉ H ₃₆₀	nC ₁₈₀ H ₃₆₂	nC ₁₈₁ H ₃₆₄	nC ₁₈₂ H ₃₆₆	nC ₁₈₃ H ₃₆₈	nC ₁₈₄ H ₃₇₀	nC ₁₈₅ H ₃₇₂	nC ₁₈₆ H ₃₇₄	nC ₁₈₇ H ₃₇₆	nC ₁₈₈ H ₃₇₈	nC ₁₈₉ H ₃₈₀	nC ₁₉₀ H ₃₈₂	nC ₁₉₁ H ₃₈₄	nC ₁₉₂ H ₃₈₆	nC ₁₉₃ H ₃₈₈	nC ₁₉₄ H ₃₉₀	nC ₁₉₅ H ₃₉₂	nC ₁₉₆ H ₃₉₄	nC ₁₉₇ H ₃₉₆	nC ₁₉₈ H ₃₉₈	nC ₁₉₉ H ₄₀₀	nC ₂₀₀ H ₄₀₂	nC ₂₀₁ H ₄₀₄	nC ₂₀₂ H ₄₀₆	nC ₂₀₃ H ₄₀₈	nC ₂₀₄ H ₄₁₀	nC ₂₀₅ H ₄₁₂	nC ₂₀₆ H ₄₁₄	nC ₂₀₇ H ₄₁₆	nC ₂₀₈ H ₄₁₈	nC ₂₀₉ H ₄₂₀	nC ₂₁₀ H ₄₂₂	nC ₂₁₁ H ₄₂₄	nC ₂₁₂ H ₄₂₆	nC ₂₁₃ H ₄₂₈	nC ₂₁₄ H ₄₃₀	nC ₂₁₅ H ₄₃₂	nC ₂₁₆ H ₄₃₄	nC ₂₁₇ H ₄₃₆	nC ₂₁₈ H ₄₃₈	nC ₂₁₉ H ₄₄₀	nC ₂₂₀ H ₄₄₂	nC ₂₂₁ H ₄₄₄	nC ₂₂₂ H ₄₄₆	nC ₂₂₃ H ₄₄₈	nC ₂₂₄ H ₄₅₀	nC ₂₂₅ H ₄₅₂	nC ₂₂₆ H ₄₅₄	nC ₂₂₇ H ₄₅₆	nC ₂₂₈ H ₄₅₈	nC ₂₂₉ H ₄₆₀	nC ₂₃₀ H ₄₆₂	nC ₂₃₁ H ₄₆₄	nC ₂₃₂ H ₄₆₆	nC ₂₃₃ H ₄₆₈	nC ₂₃₄ H ₄₇₀	nC ₂₃₅ H ₄₇₂	nC ₂₃₆ H ₄₇₄	nC ₂₃₇ H ₄₇₆	nC ₂₃₈ H ₄₇₈	nC ₂₃₉ H ₄₈₀	nC ₂₄₀ H ₄₈₂	nC ₂₄₁ H ₄₈₄	nC ₂₄₂ H ₄₈₆	nC ₂₄₃ H ₄₈₈	nC ₂₄₄ H ₄₉₀	nC ₂₄₅ H ₄₉₂	nC ₂₄₆ H ₄₉₄	nC ₂₄₇ H ₄₉₆	nC ₂₄₈ H ₄₉₈	nC ₂₄₉ H ₅₀₀	nC ₂₅₀ H ₅₀₂	nC ₂₅₁ H ₅₀₄	nC ₂₅₂ H ₅₀₆	nC ₂₅₃ H ₅₀₈	nC ₂₅₄ H ₅₁₀	nC ₂₅₅ H ₅₁₂	nC ₂₅₆ H ₅₁₄	nC ₂₅₇ H ₅₁₆	nC ₂₅₈ H ₅₁₈	nC ₂₅₉ H ₅₂₀	nC ₂₆₀ H ₅₂₂	nC ₂₆₁ H ₅₂₄	nC ₂₆₂ H ₅₂₆	nC ₂₆₃ H ₅₂₈	nC ₂₆₄ H ₅₃₀	nC ₂₆₅ H ₅₃₂	nC ₂₆₆ H ₅₃₄	nC ₂₆₇ H ₅₃₆	nC ₂₆₈ H ₅₃₈	nC ₂₆₉ H ₅₄₀	nC ₂₇₀ H ₅₄₂	nC ₂₇₁ H ₅₄₄	nC ₂₇₂ H ₅₄₆	nC ₂₇₃ H ₅₄₈	nC ₂₇₄ H ₅₅₀	nC ₂₇₅ H ₅₅₂	nC ₂₇₆ H ₅₅₄	nC ₂₇₇ H ₅₅₆	nC ₂₇₈ H ₅₅₈	nC ₂₇₉ H ₅₆₀	nC ₂₈₀ H ₅₆₂	nC ₂₈₁ H ₅₆₄	nC ₂₈₂ H ₅₆₆	nC ₂₈₃ H ₅₆₈	nC ₂₈₄ H ₅₇₀	nC ₂₈₅ H ₅₇₂	nC ₂₈₆ H ₅₇₄	nC ₂₈₇ H ₅₇₆	nC ₂₈₈ H ₅₇₈	nC ₂₈₉ H ₅₈₀	nC ₂₉₀ H ₅₈₂	nC ₂₉₁ H ₅₈₄	nC ₂₉₂ H ₅₈₆	nC ₂₉₃ H ₅₈₈	nC ₂₉₄ H ₅₉₀	nC ₂₉₅ H ₅₉₂	nC ₂₉₆ H ₅₉₄	nC ₂₉₇ H ₅₉₆	nC ₂₉₈ H ₅₉₈	nC ₂₉₉ H ₆₀₀	nC ₃₀₀ H ₆₀₂	nC ₃₀₁ H ₆₀₄	nC ₃₀₂ H ₆₀₆	nC ₃₀₃ H ₆₀₈	nC ₃₀₄ H ₆₁₀	nC ₃₀₅ H ₆₁₂	nC ₃₀₆ H ₆₁₄	nC ₃₀₇ H ₆₁₆	nC ₃₀₈ H ₆₁₈	nC ₃₀₉ H ₆₂₀	nC ₃₁₀ H ₆₂₂	nC ₃₁₁ H ₆₂₄	nC ₃₁₂ H ₆₂₆	nC ₃₁₃ H ₆₂₈	nC ₃₁₄ H ₆₃₀	nC ₃₁₅ H ₆₃₂	nC ₃₁₆ H ₆₃₄	nC ₃₁₇ H ₆₃₆	nC ₃₁₈ H ₆₃₈	nC ₃₁₉ H ₆₄₀	nC ₃₂₀ H ₆₄₂	nC ₃₂₁ H ₆₄₄	nC ₃₂₂ H ₆₄₆	nC ₃₂₃ H ₆₄₈	nC ₃₂₄ H ₆₅₀	nC ₃₂₅ H ₆₅₂	nC ₃₂₆ H ₆₅₄	nC ₃₂₇ H ₆₅₆	nC ₃₂₈ H ₆₅₈	nC ₃₂₉ H ₆₆₀	nC ₃₃₀ H ₆₆₂	nC ₃₃₁ H ₆₆₄	nC ₃₃₂ H ₆₆₆	nC ₃₃₃ H ₆₆₈	nC ₃₃₄ H ₆₇₀	nC ₃₃₅ H ₆₇₂	nC ₃₃₆ H ₆₇₄	nC ₃₃₇ H ₆₇₆	nC ₃₃₈ H ₆₇₈	nC ₃₃₉ H ₆₈₀	nC ₃₄₀ H ₆₈₂	nC ₃₄₁ H ₆₈₄	nC ₃₄₂ H ₆₈₆	nC ₃₄₃ H ₆₈₈	nC ₃₄₄ H ₆₉₀	nC ₃₄₅ H ₆₉₂	nC ₃₄₆ H ₆₉₄	nC ₃₄₇ H ₆₉₆	nC ₃₄₈ H ₆₉₈	nC ₃₄₉ H ₇₀₀	nC ₃₅₀ H ₇₀₂	nC ₃₅₁ H ₇₀₄	nC ₃₅₂ H ₇₀₆	nC ₃₅₃ H ₇₀₈	nC ₃₅₄ H ₇₁₀	nC ₃₅₅ H ₇₁₂	nC ₃₅₆ H ₇₁₄	nC ₃₅₇ H ₇₁₆	nC ₃₅₈ H ₇₁₈	nC ₃₅₉ H ₇₂₀	nC ₃₆₀ H ₇₂₂	nC ₃₆₁ H ₇₂₄	nC ₃₆₂ H ₇₂₆	nC ₃₆₃ H ₇₂₈	nC ₃₆₄ H ₇₃₀	nC ₃₆₅ H ₇₃₂	nC ₃₆₆ H ₇₃₄	nC ₃₆₇ H ₇₃₆	nC ₃₆₈ H ₇₃₈	nC ₃₆₉ H ₇₄₀	nC ₃₇₀ H ₇₄₂	nC ₃₇₁ H ₇₄₄	nC ₃₇₂ H ₇₄₆	nC ₃₇₃ H ₇₄₈	nC ₃₇₄ H ₇₅₀	nC ₃₇₅ H ₇₅₂	nC ₃₇₆ H ₇₅₄	nC ₃₇₇ H ₇₅₆	nC ₃₇₈ H ₇₅₈	nC ₃₇₉ H ₇₆₀	nC ₃₈₀ H ₇₆₂	nC ₃₈₁ H ₇₆₄	nC ₃₈₂ H ₇₆₆	nC ₃₈₃ H ₇₆₈	nC ₃₈₄ H ₇₇₀	nC ₃₈₅ H ₇₇₂	nC ₃₈₆ H ₇₇₄	nC ₃₈₇ H ₇₇₆	nC ₃₈₈ H ₇₇₈	nC ₃₈₉ H ₇₈₀	nC ₃₉₀ H ₇₈₂	nC ₃₉₁ H ₇₈₄	nC ₃₉₂ H ₇₈₆	nC ₃₉₃ H ₇₈₈	nC ₃₉₄ H ₇₉₀	nC ₃₉₅ H ₇₉₂	nC ₃₉₆ H ₇₉₄	nC ₃₉₇ H 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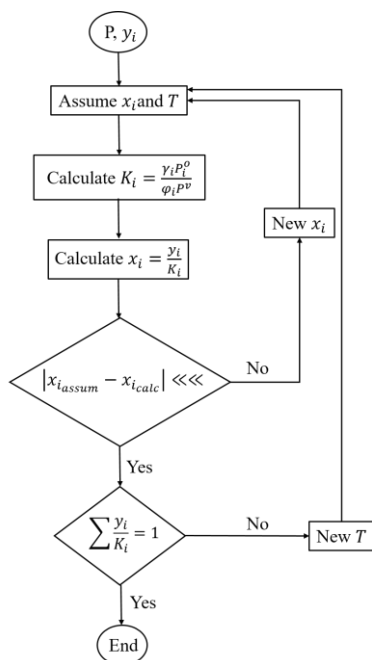


Figure 3. Algorithm for the calculation of dew points.

After its validation, the selected equation of state was used to estimate the dew points and the cricondenbar and cricondentherm points of natural gas mixtures from Colombian oil fields; considered “low impact”, for having a production volume lower than 1 MMSCFD. The application of this study is focused on the determination of the appropriate conditions, limits and operating intervals on which it is possible to have a liquid gas mixture in equilibrium without reaching the supercritical fluid zone. The results will facilitate studies of phase separation systems of natural gas at high pressure by gravitational, centrifugal or supersonic techniques, in plants where the cost of re-compression is considerably high.

• Characterization of mixtures

In this study, several mixtures of natural gas with potential for phase separation in a range of operating pressure between 4 and 10 MPa were selected. In this regard, Valiollahi *et al* (2016) collected a set of 36 mixtures of natural gas, of which, 23 of them have the present components in the GERG2008 state equation and 5 have high separation potential at high pressure according to

the analysis of the experimental equilibrium curves. Table 3 presents the molar percentage of each of the mixtures used in the thermodynamic analysis and the Table 5 shows the results of the experimental equilibrium curves of the mixtures, which were compared with the curves constructed by different EOS.

Table 3. Molar percentage of selected natural gas mixtures and its respective reference.

ID	Gas 1	Gas 2	Gas 3	Gas 4	Gas 5
Ref.	(Jarne et al., 2004)	(Mørch et al., 2006)	(Mørch et al., 2006)	(Avila, et al., 2002)	(Jarne et al., 2004)
CH ₄	84.45	96.611	93.6	83.35	69.11
N ₂	0.772	0	0	5.651	1.559
CO ₂	1.7	0	0	0.284	25.91
C ₂ H ₆	8.683	0	2.630	7.526	2.62
C ₃ H ₈	3.297	0	1.49	2.009	0.423
nC ₄ H ₁₀	0.589	1.475	1.49	0.52	0.104
iC ₄ H ₁₀	0.293	1.527	0.795	0.305	0.105
nC ₅ H ₁₂	0.086	0.385	0	0.144	0.023
iC ₅ H ₁₂	0.084	0	0	0.12	0.034
nC ₆ H ₁₄	0.05	0	0	0.068	0.11
nC ₇ H ₁₆	0	0	0	0.014	0
nC ₈ H ₁₈	0	0	0	0.011	0

• Colombian natural gas mixtures

In addition to the real and synthetic natural gas mixtures that were extracted from the literature (Avila et al., 2002; Jarne et al., 2004; Mørch et al., 2006), two mixtures of natural gas from Colombian reservoirs were analyzed. The molar percentage of the Colombian natural gas mixtures used during the present study are shown in Table 4.

Table 4. Molar percentage of gases from Colombian fields.

%mol	Gas 6	Gas 7	%mol	Gas 6	Gas 7
CH ₄	97.299	82.867	nC ₅ H ₁₂	0.013	0.024
N ₂	1.507	0.541	iC ₅ H ₁₂	0.023	0.033
CO ₂	0.095	2.008	nC ₆ H ₁₄	0.009	0.006
C ₂ H ₆	0.69	8.828	nC ₇ H ₁₆	0.004	0.004
C ₃ H ₈	0.241	3.86	nC ₈ H ₁₈	0.002	0.001
nC ₄ H ₁₀	0.062	1.19	nC ₉ H ₂₀	0.001	0
iC ₄ H ₁₀	0.054	0.638			

Table 5. Values of the cricondenthem point (K), cricondenbar point (MPa) and experimental equilibrium curves for different mixtures of natural gas (Avila et al., 2002; Jarne et al., 2004; Mørch et al., 2006).

Gas 1		Gas 2		Gas 3		Gas 4		Gas 5	
CricT (K)	261.4	CricT (K)	268.3	CricT (K)	278.4	CricT (K)	273.5	CricT (K)	252.2
CricB (MPa)	8.18	CricB (MPa)	--	CricB (MPa)	--	CricB (MPa)	9.23	CricB (MPa)	--
T(K)	P(MPa)	T(K)	P(MPa)	T(K)	P(MPa)	T(K)	P(MPa)	T(K)	P(MPa)
217.9	0.12	263.7	7.92	268.7	9.32	243.4	0.2	213.6	0.12
219.6	0.14	265.3	7.54	271.2	9.04	249.8	0.32	216.4	0.12
222.6	0.21	266.1	7.06	272.4	8.52	254.1	0.5	217.5	0.13
228.1	0.31	267	6.56	274.3	8.1	258.3	0.7	219.5	0.14
232.1	0.41	267.8	6.04	275.3	7.68	262.4	0.99	222	0.17
235.9	0.53	268.3	5.56	276.1	7.16	266.7	1.47	224.5	0.21
238.7	0.64	268.2	5.06	277.6	6.64	269.7	2.01	228.5	0.29
240.7	0.73	268	4.58	278.4	6.18	271.4	2.46	231.5	0.38
242.5	0.86	267.4	4.1	278.3	5.64	272.7	3.06	233.4	0.44
244.4	0.98	266.4	3.58	278.4	5.12	273.2	3.51	235.6	0.52
247.1	1.18	264.9	3.08	277.5	4.62	273.3	4	237.8	0.63
259.9	6.24	262.8	2.6	276.6	4.08	273.5	4.5	239.4	0.72
259.1	6.51	259.9	2.08	275.7	3.6	273.2	5	241.3	0.85
258.1	6.76	255.9	1.58	273.6	3.1	272.5	5.5	243.2	1.01
256.5	6.98	249.7	1.06	271.3	2.6	271.8	6.01	245.1	1.2
255.9	7.19	241.9	0.64	268.5	2.08	269.4	6.62	246.7	1.42
254.2	7.54			264.1	1.56	267.6	7.14	248.9	1.8
253.5	7.59			258.1	1.06	264.6	7.53	249.8	2.02
252.4	7.77			247.9	0.58	262.1	8.02	250.5	2.22
251.4	7.78			240.8	0.34	257.9	8.48	251.1	2.42
234	7.92					252.1	8.85	251.5	2.64
234.8	7.93					246.1	9.14	251.9	2.88
						241.9	9.23	252.1	3.14
						231.4	9.05	252.2	3.37
						228.1	8.87	252.2	3.83
						223.3	8.28	251.3	4.5
						215.7	7.42	250.7	4.79
								249.5	5.2
								248.1	5.61
								246.4	6.02

Results and discussion

The discussion of results is divided into the analysis of the cricondenthem and cricondenbar points of the mixtures presented in the studies by Mørch et al. (2006), Jarne et al. (2004) and Avila et al. (2002), the construction and comparison of equilibrium curves and the analysis of natural gas mixtures from

Colombian fields. Figure 4 shows the experimental values and the results of the calculation using the PR, SRK and GERG2008 state of equations for each of the selected natural gas mixtures. Table 6 also contains the absolute error percentage of each state equation compared to the experimental data.

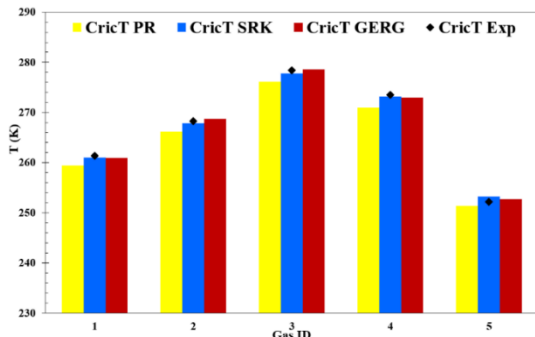


Figure 4. Experimental and simulated value of the cricondentherm point for each of the analysis gases.

Table 6. . Calculation of absolute error for each one of the cricondentherm points, using the PR, SRK and GERG2008 EOS vs. Experimental data.

Cric (K)	Gas1	%e	Gas2	%e	Gas3	%e
Exp.	261.4	--	268.3	--	278.4	--
PR	259.4	0.77	266.2	0.78	276.1	0.83
SRK	261	0.15	267.8	0.19	277.8	0.22
GERG2008	260.9	0.19	268.7	0.15	278.6	0.07
	Gas4	%e	Gas5	%e		
Expl	273.5	--	252.2	--		
PR	271	0.91	251.4	0.32		
SRK	273.2	0.11	253.2	0.40		
GERG2008	273	0.18	252.7	0.20		

In the case of the cricondentherm point, it is observed that, for three of the five experimental mixtures, the lowest percentage of error was presented using the GERG2008 state equation. The maximum percentage reached was using the PR equation in gas 4 (0.91%). However, since all the percentages of error are so low, it is considered that it is not a sufficient measure for the selection and that other properties must be analyzed. Similarly to the cricondentherm point, Figure 5 and Table 7 present the comparison and the calculation of percentage of error of the cricondenbar points for the mixtures of natural gas that are adapted to the process of stage separation at high pressure. According to the available experimental data, it is observed that both the SRK and GERG2008 state equations have a low percentage of error when calculating the values of the maximum pressure where the liquid and gas phases coexist in equilibrium.

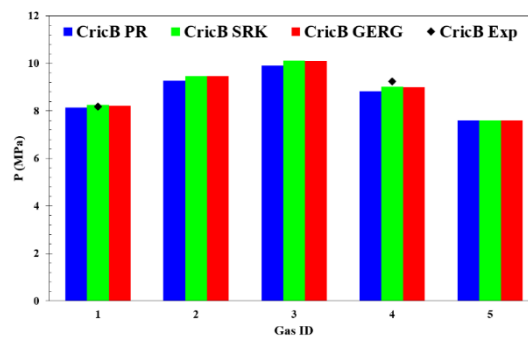


Figure 5. Experimental and simulated value of the cricondenbar point for each of the analysis gases.

Table 7. Calculation of absolute error for each one of the cricondenbar points, using the PR, SRK and GERG2008 EOS vs. Experimental data.

Cric (MPa)	Gas1	%e	Gas2	%e	Gas3	%e
Exp.	8.18	--	--	--	--	--
PR	8.134	0.56	9.269	--	9.902	--
SRK	8.247	0.82	9.466	--	10.11	--
GERG2008	8.218	0.46	9.459	--	10.09	--
	Gas4	%e	Gas5	%e		
Expl	9.23	--	--	--		
PR	8.828	4.36	7.595	--		
SRK	9.02	2.28	7.606	--		
GERG2008	8.999	2.50	7.596	--		

Once again, in light of this information, any of the three state equations would be adequate to estimate the cricondenbar and cricondentherm points; so it is not enough to make a decision, it is necessary to study the behavior of the pressure depending on the temperature for each of the mixtures of Table 3. After analyzing the cricondentherm and cricondenbar points of the five natural gas mixtures, the pressure profiles were plotted as a function to temperature (Figure 8) and later they were compared with experimental data.

The calculated equilibrium curves show that at low pressures, all the state equations analyzed have a good correlation with the experimental data, thus the equation that is used to estimate the thermodynamic properties of the mixtures is considered indifferent, that is, any of the three works with relatively low error rates. However, if the objective is to operate high-pressure multiphase separation systems, not all state equations are

adequate; as it is the case of the Peng-Robinson equation, where in none of the dew curves the real behavior of the mixture was represented, especially near the cricondentherm and cricondenbar points; in this last point the Peng equation Robinson reaches the maximum error rate of 4.36% for gas 4 (Table 7).

Discarding the Peng-Robinson's equation of state for the calculation of the dew curve of high-pressure natural gas mixtures, the state equation was established that estimates the thermodynamic properties of the multi-component system. In Figure 8, it is possible to observe that in spite of not existing a significant difference between the two remaining equations of state, the dew curve calculated with the GERG2008 equation is the one that better correlates the experimental data of the literature; this trend is more evident in the results of gases 2, 3, and 4. As a special case, gas 5 starts with a favorable trend for the GERG2008 equation and as the pressure increases, the Soave-Redlich-Kwong equation is closer to the real trend in a small section of the dew curve, unfortunately there is no experimental data in the region of the cricondenbar point.

Application to Colombian natural gas mixtures

Figure 8 suggests that the GERG2008 state equation manages to adequately reproduce the thermodynamic behavior of different types of synthetic and real mixtures of natural gas. For this reason, two real mixtures of natural gas from Colombian production fields were chosen (Table 4). The first (Figure 6) corresponds to a "dry" natural gas, mainly composed of methane and therefore has a low separation potential, due to the lack of heavy hydrocarbons (C_5H_{12} hereinafter), this gas is especially suitable for use in electric power generation systems.

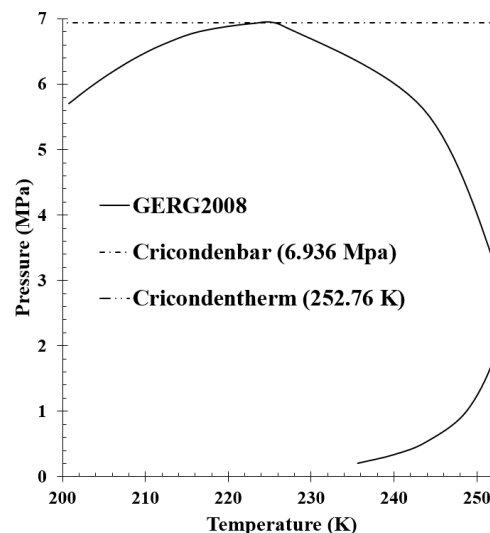


Figure 6. Colombian natural gas 6 phase envelope and properties calculated with the GERG2008 equation of state.

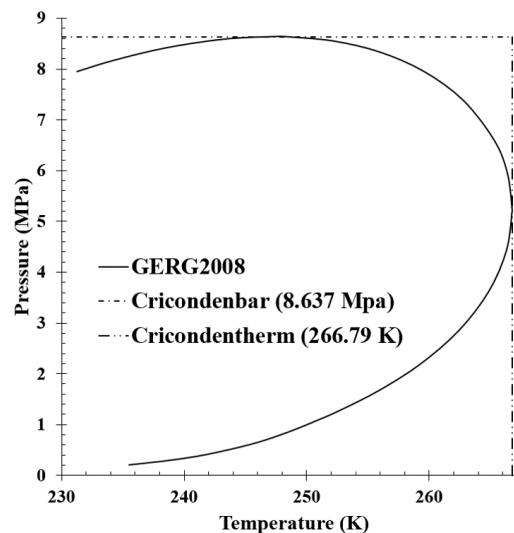


Figure 7. Colombian natural gas 7 phase envelope and properties calculated with the GERG2008 equation of state.

The second mixture (Figure 7) is a "wet" gas, in addition to methane contains considerable amounts of ethane, propane, butane, among others (Table 4), additionally has 2% of CO_2 ; this gas, apart from going through a phase separation process, could be subjected to a treatment with amines or membranes for its sweetening (removal or reduction of CO_2).

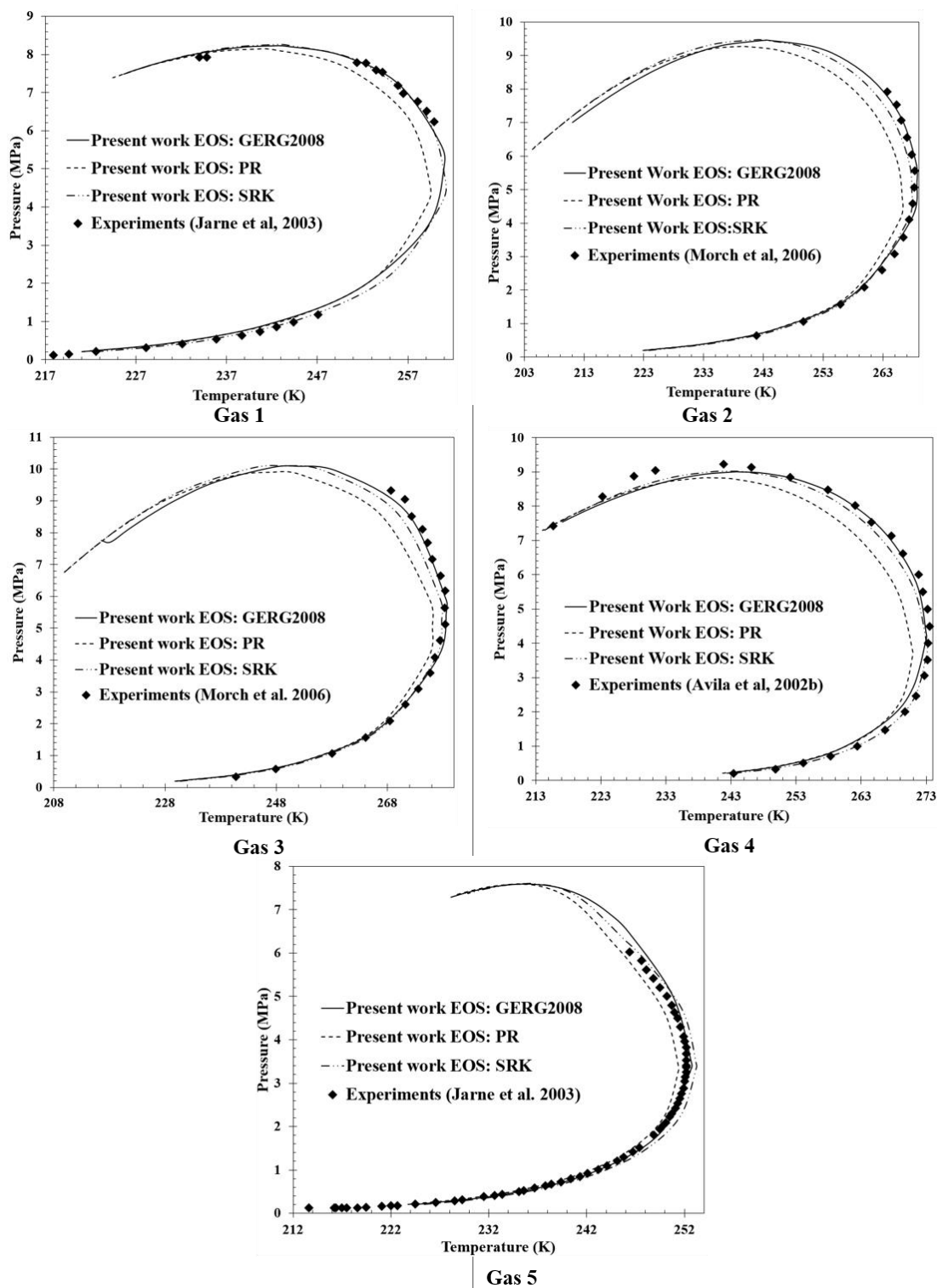


Figure 8 Comparison of the calculate phase equilibrium curves with the experimental data.

Both the dew curve and the cricondenthem and cricondenbar points show that for this humid gas there is a range of pressure and temperature conditions for which it is possible to carry out phase separation and recommend an operating pressure value in the separator close to the of the container well, which for Colombian natural gas wells is between 7 and 10 MPa.

Conclusion

From equilibrium experimental curves of five real and synthetic natural gas mixtures from the literature, equilibrium curves were constructed, compared and correlated using three different equations of state: Peng-Robinson, Soave-Redlich-Kwong and GERG2008. The results showed that there is no significant difference in the calculation of the cricondenthem points, presenting a maximum absolute error of 1% with respect to the real values. The calculation of the cricondenbar points yielded a maximum absolute error of 4.36% when the Peng-Robinson equation of state was used; therefore, both the Soave-Redlich-Kwong equation and the GERG2008 equation have low error rates, compared to the experimental points. Regarding the equilibrium curves, at low pressures the equation of state used to estimate the thermodynamic properties of natural gas is indifferent, however, as the pressure increases the Peng-Robinson equation is not suitable for represent the behavior of the multiphase and multicomponent system. The results suggest that, as long as the mixture contains the 21 present components in the GERG2008 state equation, this is the equation that best estimates the thermodynamic properties of the mixtures. In case of having additional components to those of Table 2 it is possible to use the Soave-Redlich-Kwong equation to calculate the equilibrium curves, since the results show that the error percentages are low for high pressure conditions. Finally, when performing the dew curves for two gas mixtures from Colombian fields using the GERG2008 state equation, it is possible to affirm that for "wet" gases there is a potential opportunity to carry out stage separation at high pressure, thus making it possible

to eliminate or reduce the expenses associated with re-compression during the natural gas processing.

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