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Vapor-liquid equilibrium of binary systems containing low GWP refrigerants with cubic equations of state

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Abstract

Different Cubic Equations of State (CESs) were used to represent Vapor-Liquid Equilibrium (VLE) of binary systems containing low Global Warming Potential (GWP) refrigerants. The calculated VLE data from some of the widely used CESs were compared with the experimental data collected from the literature. To extend CESs to binary systems, van der Waals one-fluid mixing rules with a single binary interaction parameter were used. Although the deviations between the experimental and calculated values are generally low, a comparison of the results was performed to find the most accurate CES for VLE of the studied binary systems.

Keywords: HFO; refrigerant binary systems; VLE; cubic equation of state.

1. Introduction

The development of accurate models for representation of thermophysical properties and phase equilibria of low Global Warming Potential (GWP) refrigerants, such as HydroFluoro-Olefins (HFOs) and HydroChloroFluoro-Olefins (HCFOs), and their binary systems is of great importance to evaluate the potential performance of working fluids in refrigeration applications. One of the most used and reliable tool to descript the properties of refrigerants is the equation

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of state (EOS). Among the many EOSs developed over the years that ranged from simple equations with few constants to complex forms with many constants, the cubic equations of state (CESs) are still models of great interest in many engineering applications. The CESs are semi-empirical expressions derived from the original van der Waals equation of state [1] that are cubic in volume and combine a significant simplicity of calculation with a thermodynamic consistency. In particular, to enhance the representation of the vapor pressure and volumetric properties, a large number of CESs with different expressions and number of parameters were developed [2]. These models have been, and are still, widely used for the representation of the Vapor-Liquid Equilibrium (VLE) of refrigerant mixtures in the academic and industrial fields.

In this work, the VLE of 25 binary systems of alternative refrigerants were described using four two-parameters CESs. A wide comparison between the results of the studied CESs were performed on the basis of the experimental data available in literature. The CESs were extended to refrigerant binary systems through van der Waals one-fluid mixing rules with a single binary interaction parameter, also kwon as the standard one-binary-parameter mixing rules. In particular, this parameter was regressed by minimizing the deviations between the calculated and experimental pressure for complete datasets and by minimizing the deviations for each isotherm.

2. Cubic equations of state

In general, the two-parameters CESs have the following form:

$$P = \frac{R \cdot T}{(V - b)} - \frac{a(T)}{(V^2 + u \cdot V + w \cdot b^2)} \tag{1}$$

where P is the pressure, R is universal gas constant, V is molar volume, T is temperature, $a(T)$ is temperature dependent parameter, b is parameter not necessarily a function of the temperature, and u and w are two constants that define a specific CES. The parameters a and b in Eq. (1) are:

$$a(T) = \Omega_a \frac{R^2 \cdot T_c^2}{P_c} \alpha(T_r, \omega) \tag{2}$$

$$b = \Omega_b \frac{R \cdot T_c}{P_c} \tag{3}$$

where T_c is the critical temperature, P_c is the critical pressure, $\alpha(T_r, \omega)$ is a dimensionless function of the reduced temperature ($T_r = T \, T_c^{-1}$) and acentric factor (ω), and Ω_a and Ω_b are numerical constants which were calculated from the thermodynamic condition of the critical point. The α function used in this work has the same form of the expression introduced by Soave [3]:

$$\alpha(T_r, \omega) = \left[1 + K \cdot (1 - T_r)^{1/2}\right]^2 \tag{4}$$

where, in this case, k is a function of the acentric factor (ω); this parameter can be written as:

$$k(\omega) = c_0 + c_1 \cdot \omega - c_2 \cdot \omega^2 + c_3 \cdot \omega^3 \tag{5}$$

where c_0, c_1, c_2, and c_3 are coefficients specific for each CES.

Several two-parameters CESs, characterized by different levels of accuracy, have been proposed in literature by considering different values of u and w, including the well-known Redlich-Kwong-Soave (RKS) [3] and Peng-Robinson (PR) [4] EOSs. Every two-parameters CES is characterized by specific values of constants and critical compressibility factor, $Z_c = P_c\, V_c\, R^{-1}\, T_c^{-1}$. In order to obtain a more accurate description of specific properties for pure fluids and binary systems, many modifications of the proposed CESs were presented in literature. In this respect, the RKS and PR EOSs proposed by Stryjek [5–7], which guarantee a better representation of the saturated vapor pressure of mixture components, shown accurate representation of the vapor-liquid equilibrium (VLE) and critical state for the studied binary systems.

To evaluate the ability of the CESs to represent the VLE for low GWP refrigerant binary systems, the original RKS and PR EOSs were compared together with two CESs proposed by Stryjek [5–7]. The characteristics of the studied CESs (CESs' names, references to the original works and their respective values of u, w, coefficients, and Z_c) are reported in Table 1.

Table 1. Names, references to original work and coefficients of studied CESs.

CES Name	Ref.	u	w	Ω_a	Ω_b	Zc	c_0	c_1	c_2	c_3
RKS	[3]	1	0	0.4275	0.0866	0.333	0.4800	1.5740	0.1760	0.0000
PR	[4]	2	-1	0.4572	0.0778	0.307	0.3746	1.5423	0.2699	0.0000
PR mod	[6,7]	2	-1	0.4572	0.0778	0.307	0.3788	1.4895	0.1709	0.0194
Eq. 1	[6,7]	1	-1	0.4638	0.1074	0.333	0.3577	1.4713	0.1665	0.0183

The standard one-binary-parameter mixing rules, adopted to extend the selected CESs to refrigerant binary systems, are expressed as:

$$a_m = \sum_{i=1}^{Nc} \sum_{j=1}^{Nc} y_i \cdot y_j \cdot a_{ij} \tag{6}$$

$$b_m = \sum_{i=1}^{Nc} y_i \cdot b_i \tag{7}$$

$$a_{ij} = \left(a_i \cdot a_j\right)^{1/2}\left(1 - k_{ij}\right) \tag{8}$$

where Nc is the number of compounds, k_{ij} is the binary interaction parameter, $k_{ij} = k_{ij}$, and $k_{ii} = k_{jj} = 0$.

3. Vapor Liquid Equilibrium of binary systems containing low GWP refrigerants

After collecting reliable experimental data of VLE for low GWP refrigerant binary systems available in literature, the CESs were tested to represent VLE for these binary systems on the basis of the selected data. Since the simple models under analysis are often inaccurate in the description of the properties of systems containing components that were studied above their critical point, the calculation of VLE data for these binary systems were not considered.

The number of isotherms, the number of points, the references to the original sources and the ranges of temperature for the binary systems under analysis are reported in Table 2.

Table 2. VLE experimental data for low GWP refrigerant binary systems.

Binary system	N. isotherms (N. points)	Reference	T range (K)
R1233xf+R244bb	3(30)	[8]	323.15 - 303.15
R1234yf+R1233xf	3(30)	[8]	313.15 - 293.15
R1234yf+R152a	5(60)	[9]	323.15 - 283.15
R1234yf+R227ea	5(50)	[10]	323.15 - 283.15
R1234yf+R244bb	3(30)	[11]	313.15 - 293.15
R1234yf+R245cb	4(44)	[12]	313.15 - 283.15
R1234yf+R290	5(54)	[13]	293.15 - 253.15
R1234yf+R600a	5(60)	[14]	323.15 - 283.15
R1234ze(E)+R290	4(38)	[15]	283.15 - 258.15
R1234ze(E)+R600a	4(40)	[16]	288.15 - 258.15
R125+R1234yf	7(84)	[17]	333.21 - 273.15
R134+R1234ze(E)	4(40)	[18]	288.15 - 258.15
R134+R1234ze(Z)	5(71)	[19]	343.15 - 303.15
R134a+R1234yf	7(63)	[17]	333.21 - 273.31
R13I1+R1234ze(E)	5(55)	[20]	298.15 - 258.15
R143a + R1234yf	5(45)	[21]	323.15 - 283.15
R152a+R1234ze(E)	4(44)	[22]	288.15 - 258.15
R161+R1234yf	5(55)	[23]	323.15 - 283.15
	5(60)	[24]	323.15 - 283.15
R218+R1234yf	2(20)	[25]	272.81 - 223.08
R23+R1234yf	3(26)	[25]	273.25 - 193.19
R290+R1234ze(Z)	5(68)	[26]	293.15 - 253.15
R32+R1234yf	5(55)	[23]	323.15 - 283.15
	7(77)	[17]	333.15 - 273.15
R32+R1234ze(E)	5(15)	[23]	323.15 - 283.15
	5(65)	[27]	323.15 - 283.15
R600a+R1234ze(Z)	6(69)	[28]	353.15 - 303.15
R717+R1234yf	4(58)	[29]	283.15 - 253.15

Table 3 shows the Absolute Average Relative Deviation of pressure (AARD (ΔP) %) and the Absolute Average Deviation of vapor phase mole fraction (AAD (Δy_1)) for the studied binary systems considering k_{ij} as a constant. In fact, these coefficients were regressed from the complete datasets by minimizing the AARD (ΔP) %. The AARD of pressure and the AAD of vapor phase mole fraction are respectively defined as:

$$AARD(\Delta P)\% = \frac{100}{N}\sum_{i=1}^{N}\left|\frac{P_{i,\exp} - P_{i,calc}}{P_{i,\exp}}\right| \tag{6}$$

$$AAD(y1) = \sum_{i=1}^{N}\left|\frac{y_{1i,\exp} - y_{1i,calc}}{N}\right| \tag{7}$$

Table 3. Deviations between experimental and calculated VLE data for binary systems of low GWP refrigerants considering constant k_{ij}.

Binary systems	RKS		PR		PR mod		Eq. 1	
	AARD (ΔP) %	AAD ($\Delta y1$)	AARD (ΔP) %	AAD ($\Delta y1$)	AARD (ΔP) %	AAD ($\Delta y1$)	AARD (ΔP) %	AAD ($\Delta y1$)
R1233xf+R244bb	0.9	0.0105	0.96	0.0102	1	0.0103	1.03	0.0102
R1234yf+R1233xf	1.16	0.0024	1.36	0.0012	1.37	0.0009	1.42	0.001

Binary systems							
R1234yf+R152a	0.62	0.0029	0.24	0.0021	0.24	0.0019	0.2	0.0021
R1234yf+R227ea	0.8	0.0049	0.36	0.0034	0.4	0.0035	0.33	0.0034
R1234yf+R244bb	0.83	0.0028	0.37	0.0031	0.44	0.0033	0.38	0.0036
R1234yf+R245cb	0.8	0.0106	0.38	0.0118	0.38	0.0116	0.32	0.0119
R1234yf+R290	0.29	0.0016	0.19	0.0027	0.22	0.002	0.26	0.0024
R1234yf+R600a	1.01	0.0063	0.57	0.0052	0.61	0.0048	0.53	0.0049
R1234ze(E)+R290	0.34	0.0039	0.34	0.0041	0.32	0.0037	0.34	0.0036
R1234ze(E)+R600a	0.42	0.004	0.38	0.0028	0.43	0.003	0.49	0.0027
R125+R1234yf	0.72	0.012	0.4	0.0107	0.37	0.011	0.35	0.0108
R134+R1234ze(E)	0.37	0.0044	0.15	0.0032	0.18	0.0041	0.26	0.0041
R134+R1234ze(Z)	3.58	0.0138	3.41	0.0134	3.37	0.0135	3.37	0.0135
R134a+R1234yf	0.81	0.0056	0.38	0.0037	0.34	0.0039	0.28	0.0036
R13I1+R1234ze(E)	0.53	0.004	0.27	0.0044	0.21	0.0038	0.23	0.0039
R143a + R1234yf	0.81	0.0027	0.4	0.0015	0.41	0.0014	0.34	0.0017
R152a+R1234ze(E)	0.64	0.0025	0.44	0.0017	0.47	0.0023	0.52	0.0023
R161+R1234yf	0.84	0.0049	0.37	0.0023	0.41	0.0025	0.33	0.0022
	0.84	0.0047	0.45	0.0036	0.5	0.0037	0.44	0.0036
R218+R1234yf	0.72	0.0051	0.59	0.0049	0.69	0.0055	0.85	0.0056
R23+R1234yf	1.94	0.0028	1.06	0.0029	1.19	0.0034	1.18	0.0037
R290+R1234ze(Z)	1.09	0.0071	1.24	0.008	1.15	0.008	1.18	0.008
R32+R1234yf	0.89	0.0057	0.63	0.0038	0.65	0.004	0.61	0.0038
	0.82	0.0098	0.51	0.0091	0.54	0.0093	0.48	0.0092
R32+R1234ze(E)	0.73	0.0051	0.67	0.0032	0.67	0.0034	0.66	0.0031
	0.85	0.0055	0.59	0.0052	0.6	0.0051	0.58	0.005
R600a+R1234ze(Z)	0.59	0.0077	0.57	0.0095	0.55	0.0092	0.58	0.0096
R717+R1234yf	4.37	0.0419	4.19	0.0415	3.99	0.0415	3.93	0.0413
Average	1.06	0.0077	0.81	0.0071	0.81	0.0071	0.79	0.0071

The AARD (ΔP) % and AAD (Δy_1) for the selected binary systems calculated adopting a temperature dependent k_{ij} (T) for the standard one-binary-parameter mixing rules are reported in Table 4. In this case, k_{ij} were regressed by minimizing the AARD (ΔP) % for each isotherm.

Table 4. Deviations between experimental and calculated VLE data for binary systems of low GWP refrigerants considering a temperature dependent k_{ij} (T).

Binary systems	RKS		PR		PR mod		Eq. 1	
	AARD (ΔP) %	AAD (Δy_1)	AARD (ΔP) %	AAD (Δy_1)	AARD (ΔP) %	AAD (Δy_1)	AARD (ΔP) %	AAD (Δy_1)
R1233xf+R244bb	0.80	0.0108	0.80	0.0108	0.80	0.0108	0.80	0.0107
R1234yf+R1233xf	0.65	0.0031	0.80	0.0029	0.80	0.0025	0.85	0.0026
R1234yf+R152a	0.56	0.0029	0.23	0.0022	0.24	0.0019	0.20	0.0021
R1234yf+R227ea	0.74	0.0049	0.34	0.0035	0.38	0.0035	0.32	0.0034
R1234yf+R244bb	0.60	0.0026	0.22	0.0030	0.30	0.0032	0.29	0.0034
R1234yf+R245cb	0.59	0.0109	0.22	0.0120	0.24	0.0119	0.20	0.0121
R1234yf+R290	0.23	0.0018	0.14	0.0025	0.14	0.0020	0.14	0.0023
R1234yf+R600a	0.99	0.0063	0.56	0.0052	0.60	0.0049	0.52	0.0049
R1234ze(E)+R290	0.31	0.0040	0.33	0.0043	0.31	0.0038	0.33	0.0039
R1234ze(E)+R600a	0.40	0.0041	0.28	0.0030	0.34	0.0032	0.34	0.0031
R125+R1234yf	0.60	0.0116	0.33	0.0104	0.33	0.0108	0.31	0.0106
R134+R1234ze(E)	0.18	0.0039	0.12	0.0033	0.14	0.0041	0.16	0.0041
R134+R1234ze(Z)	0.45	0.0055	0.43	0.0062	0.38	0.0059	0.45	0.0061
R134a+R1234yf	0.53	0.0053	0.22	0.0037	0.20	0.0039	0.18	0.0037
R13I1+R1234ze(E)	0.34	0.0041	0.26	0.0044	0.21	0.0038	0.20	0.0038
R143a + R1234yf	0.73	0.0023	0.26	0.0017	0.30	0.0016	0.22	0.0018
R152a+R1234ze(E)	0.53	0.0025	0.41	0.0016	0.43	0.0022	0.43	0.0022
R161+R1234yf	0.79	0.0048	0.33	0.0022	0.39	0.0025	0.31	0.0021
	0.79	0.0046	0.45	0.0036	0.49	0.0037	0.44	0.0036
R218+R1234yf	0.66	0.0053	0.45	0.0054	0.55	0.0056	0.62	0.0062
R23+R1234yf	1.62	0.0038	1.02	0.0029	1.15	0.0033	1.17	0.0037
R290+R1234ze(Z)	0.94	0.0070	1.05	0.0081	0.95	0.0081	0.97	0.0084
R32+R1234yf	0.88	0.0056	0.62	0.0039	0.64	0.0041	0.59	0.0039
	0.80	0.0097	0.51	0.0091	0.53	0.0093	0.48	0.0092
R32+R1234ze(E)	0.68	0.0050	0.60	0.0032	0.60	0.0034	0.58	0.0031
	0.84	0.0055	0.56	0.0052	0.56	0.0051	0.54	0.0051

R600a+R1234ze(Z)	0.54	0.0077	0.51	0.0094	0.48	0.0090	0.50	0.0094
R717+R1234yf	4.24	0.0413	4.05	0.0415	3.86	0.0411	3.80	0.0411
Average	0.80	0.0072	0.58	0.0068	0.58	0.0068	0.57	0.0068

Table 3 and Table 4 show that the CESs generally ensure a good representation of the VLE for almost all the studied systems, adopting both constant k_{ij} and $k_{ij}(T)$. In the former case, the four CESs gave AARD (ΔP) % lower than 2% and AAD (Δy_1) lower than 0.01 for almost all the studied cases. However, as shown in Table 4, as expected the CESs with $k_{ij}(T)$ generally produced lower AARD(ΔP) % and AAD (Δy_1) than that of CESs with constant k_{ij}. As example, the improved description provided by PR EOS with k_{ij} regressed for each isotherm is shown in Fig. 1. This figure presents the behaviors of experimental and calculated pressure versus liquid and vapor molar fractions for R134 + R1234ze(Z) system at T = 303.15 K considering both constant k_{ij} and $k_{ij}(T)$ for the PR EOS. It is worthwhile noting that, as evident in Fig. 2 for the aforementioned binary system, a slightly variation of $k_{ij}(T)$ with the temperature guarantees an enhanced representation of the VLE. However, the CESs provided high values of AARD(ΔP) % and AAD ($\Delta y1$) for R717 + R1234yf in both the cases.
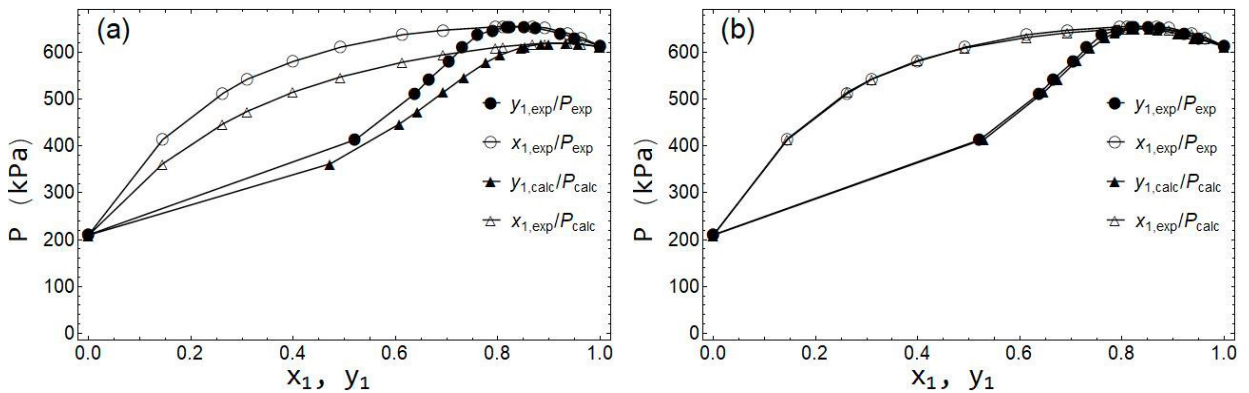

Fig. 1. $P-x-y$ diagram for R134 (1) + R1234ze(Z) (2) system at T = 303.15 K considering both (a) constant k_{ij} (k_{ij} = 0.11116) and (b) temperature variable k_{ij} (k_{ij} = 0.14952) adopting the PR EOS model.
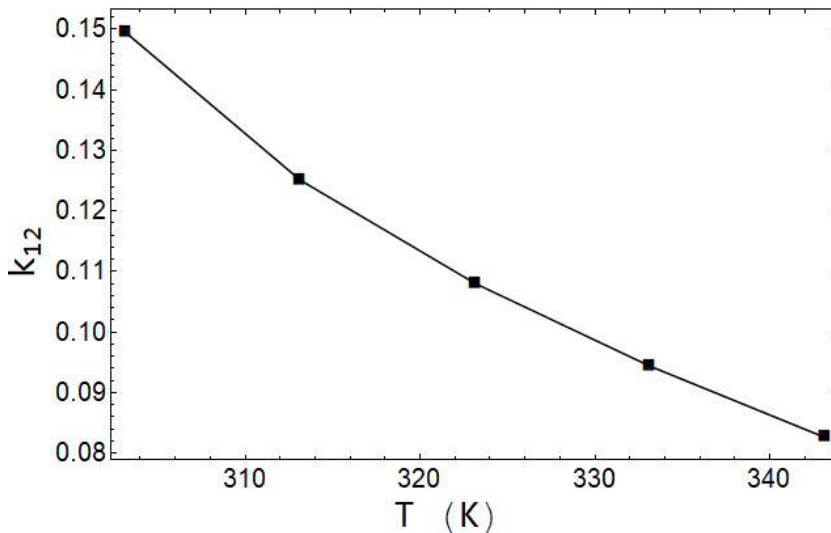

Fig. 2. $k_{ij}(T)$ as function of the temperature for R134 + R1234ze(Z) adopting the PR EOS model.

4. Conclusions

A literature survey of the experimental VLE data for binary systems of low GWP refrigerants is presented in this work.

These data were studied with the original RKS and PR EOSs. In addition, a modified PR EOS and a CES (Eq.1) proposed by Stryjek were also tested, since they produced lower deviations among the CESs proposed by the same author.

All the models provide satisfactory results for the VLE description, both when the standard one-binary-parameter mixing rules independent on temperature and the same mixing rules dependent on temperature were used.

At the end, the results are generally very good and confirm the representation ability of the CESs, in spite of their simplicity.

Acknowledgements

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