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Research Article

Analysis of Thermodynamic Models for Liquid-Vapor Equilibrium: Evaluating Accuracy and Applicability

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Abstract

This article investigates the liquid-vapor equilibrium of four binary refrigerant systems: R134a + R290, R152a + R1234ze, R152a + R1243zf, and R1243zf + R134a. The study employs three thermodynamic models for accurate predictions: the Peng-Robinson equation with the classical mixture rule of van der Waals (vdW) and the Wilson model, the PC-SAFT equation, and the PR-MC-WS-NRTL model. Activity coefficients are determined using the Peng-Robinson equation with vdW and the Wilson model. The PC-SAFT equation and the PR-MC-WS-NRTL model are also applied to model the data. The calculated results show good agreement with reference data. Favorable agreements exist between the calculated results and the reference data, with relative errors remaining below (0.15 and 0.42) % for the molar fraction and the pressure, respectively. This research provides valuable insights into the accuracy and applicability of different thermodynamic models in predicting liquid-vapor equilibrium within refrigerant systems.

Keywords: *Equations of state; pc-saft; binary mixture; gwp; elv.*

1. Introduction

The international community has long recognized the urgent need to address the environmental impact of refrigerants, leading to the establishment of various protocols aimed at mitigating their harmful effects. Notable among these are the Montreal Protocol and the Kyoto Protocol. Initiated in 1987, the Montreal Protocol focuses on the phase-out of ozone-depleting substances, including certain refrigerants. Its success in phasing out substances like chlorofluorocarbons (CFCs) has significantly contributed to the healing of the ozone layer. In contrast, the Kyoto Protocol targets greenhouse gas emissions, including those associated with certain refrigerants. By setting binding emission reduction targets for industrialized nations, it seeks to combat climate change. These protocols collectively represent global efforts to strike a balance between the essential role of refrigerants in daily life and the imperative to protect the environment.

Understanding the thermodynamic behavior of refrigerants is crucial in designing efficient cooling systems. The utilization of the PR (Peng-Robinson) equation of state holds particular relevance, enabling accurate modeling of the thermodynamic properties of refrigerants. Incorporating activity parameters, such as those derived from models like NRTL (Non-Random Two-Liquid) or the Wilson model, significantly enhances the accuracy of thermodynamic predictions. These parameters account for non-ideal interactions among refrigerant molecules, thus considering the effects of non-ideality in mixtures. Additionally, the PC-SAFT (Perturbed Chain Statistical Associating Fluid Theory) approach provides an advanced alternative for modeling complex mixtures, offering an accurate representation of both liquid and gaseous phases. Integrating these models into cooling system designs allows optimization of energy performance and equipment reliability, contributing to more efficient and sustainable cooling solutions.

PC-SAFT (Perturbed-Chain Statistical Associating Fluid Theory) plays a pivotal role in the accurate prediction and calculation of liquid-vapor equilibrium, particularly in the realm of refrigerants and other complex fluids. It is a relatively new model that has been shown to be more accurate than traditional cubic equations of state such as Redlich-Kwong, Soave-Redlich-Kwong, and Peng-Robinson [**1**]. PC-SAFT takes into account the molecular structure of the fluids being modeled, allowing for better prediction of the behavior of mixtures with complex interactions. It has been used to model VLE in a variety of systems. including CO2 mixtures [2], and deep eutectic solvents and ionic liquids [3]. To use PC-SAFT for VLE calculations, the mixture composition, density, and temperature are utilized as independent variables [1].

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Researchers and engineers leverage PC-SAFT to explore the phase equilibria of refrigerants, enabling a comprehensive understanding of their thermodynamic properties. Recent studies. such as those by Gross and Sadowski [4] and Mollerup and Kontogeorgis [5], have demonstrated the effectiveness of PC-SAFT in providing precise predictions of phase behavior, critical points, and saturation properties. These references highlight the significance of PC-SAFT in advancing our comprehension of liquid-vapor equilibrium, paving the way for the development of environmentally sustainable cooling technologies.

The purpose of this study is to introduce a new simple calculation technique for predicting the binary interaction parameter [6] within the framework of the classical mixing rule. This technique aims to enhance the accuracy of isothermal (VLE) calculations in both subcritical and supercritical regions for various binary Refrigerant systems. Utilizing PR (with the incorporation of activity coefficients such as the Wilson and NRTL models) or PC-SAFT allows for a more accurate representation of non-ideal behavior, widely used in academic research.

The binary refrigerant systems considered in this study are: (R134a + R290) [7], (R152a + R1234ze (Z)) [8], (R152a + R1243zf) [8], and (R1243zf + R134a) [9]. These binary systems are noted for being environmentally friendly, possessing zero ozone depletion potential (ODP) and low global warming potential (GWP).

Tables 1 and 2 present the critical parameters, acentric factors, and environmental properties of the compounds in the binary blends, pure-component parameters for the PC-SAFT equation of state, and Mathias–Copeman coefficients.

Table 1. Critical parameters (Tc.Pc) and acentric factors (ω), ODP and GWP.

Compound	T_c/K	P_c/MPa	ω	ODP	GWP
R290	369.89	4.0593	0.1521	0	3
R1234ze	423.27	3.5330	0.327	0	<1
R1243zf	376.93	3.5182	0.261	0	<1
R134a	374.21	4.2512	0.327	0	1430
R152a	386.35	4.4990	0.226	0	124

2. Thermodynamic Models

In this study, we will use three different models to calculate the mole fractions of the liquid phase, the vapor phase, as well as the pressure.

2.1 Perturbed-Chain Statistical Fluid Theory. PC-SAFT Model

The first model is that of the PC-SAFT equation, which has demonstrated its capacity to predict the position of the azeotropeand determine the equilibrium values of liquid and vapour molar fractions. The equations have been described in previous works [10-11-12].

The PC-SAFT equation of state is a molecular-based model that considers molecules as chains of spherical segments. It represents the residual Helmholtz energy and incorporates various contributions. It's important to note that the terms of the equation of state corresponding to repulsive and dispersive effects require knowledge of three input parameters: the number of segments (m), the corresponding segment diameter (σ), and the segment dispersion energy parameter (ε/k). When dealing with mixtures, similar to cubic equations, it's necessary to define combination rules for the cross parameters of σ and ε. If i and j denote two segments, the commonly used rules are referred to as Lorentz-Berthelot combination rules.

$$\begin{cases} \varepsilon_{ij} = \sqrt{\varepsilon_i \varepsilon_j}\,(1 - k_{ij}) \\ \sigma_{ij} = \frac{\sigma_i + \sigma_j}{2} 1 - l_{ij}) \end{cases} \tag{1}$$

Where $k_{ij}$the adjustable binary interaction parameter is that canbe fitted to experimental datafor mixtures. In this model, the general definition of the Helmholtz energy is illustrated by

$$\begin{cases} \overline{a}^{res} = \overline{a}^{seg} + \overline{a}^{chain} + \overline{a}^{assoc} \\ P(T.v) = -\left(\frac{\partial \overline{a}^{res}}{\partial T}\right)_v \end{cases} \tag{2}$$

Where$\overline{a}^{seg}$ is the Helmholtz energy of the segment, including both hard-sphere reference and dispersion terms,

This$\overline{a}^{chain}$ is the contribution from chain formation and $\overline{a}^{assoc}$is the contribution of the associating interactions. Currently, most process simulators do not include association terms in their SAFT models.

2.2 PR-MC-WS-NRTL Model

PR-MC-WS-NRTL model was applied by our group [11-13-14] to correlate the experimental data. The model is based on the Peng-Robinson equation of state (PR EoS), the Mathias-Copeman (MC) alpha function, the Wong-Sandler (WS) mixing rules involving the NRTL (Non-Random Two Liquids) model.

2.3 PR-vdW-Wilson Model

The PR-EoS is given by the following form:

$$p = \frac{RT}{v-b} - \frac{\alpha(T)}{v^2+2vb-b^2} \tag{3}$$

$$\alpha_i(T) = 0.457235\ \frac{R^2 T_{c.i}^2 \alpha_i(T)}{p_{c.i}} \tag{4}$$

$$b_i = 0.0077796\ \frac{RT_{c.i}}{p_{c.i}} \tag{5}$$

Where their alpha-function, it is given by:

$$\alpha_i(T) = [1 + (0.37464 + 1.5422\ \omega_i - 0.26992\omega_i^2)(1 - \sqrt{\frac{T}{T_{c.i}}}) \tag{6}$$

The fugacity coefficients in the liquid and vapor phases must be equal for the refrigerant blends to be in isothermal vapor-liquid equilibrium. The fugacity coefficient of species i can be expressed as follows when the Peng-Robinson Equation of State (PR-EoS) is used with the standard mixing rule (vdW) to forecast the isothermal vapor-liquid equilibrium:

$$\ln \emptyset_i^L = \frac{b_i}{b_m}(z-1) - \ln\left[z\left(1 - \frac{b_m}{v}\right)\right] + \frac{a_m}{2.828RTb_m}\left(\frac{b_i}{b_m} - \frac{2}{a_m}\sum_i x_i a_{ij}\right)\ln\left(\frac{1+2.414\frac{b_m}{v}}{1-0.414\frac{b_m}{v}}\right) \tag{7}$$

The mixing rule of van der Waals (vdW) is as follows:

$$\alpha_m = \sum_i \sum_j x_i x_j a_{ij} \tag{8}$$

$$b_m = \sum_i x_i b_i \tag{9}$$

With:

$$a_{ij} = (1 - k_{ij})\sqrt{a_i a_j} \tag{10}$$

Where $k_{ii} = 0$. $k_{ij}=k_{ji}$ is the binary interaction parameter.

The Wilson model is a thermodynamic model involving excess free enthalpy. It is a classic model used for predicting liquid-vapor equilibrium or estimating excess free enthalpy of miscible mixtures. In its initial form, the Wilson model ΔG^Eexpresses in the following expression:

$$\frac{G_m^E}{RT} = -\sum_i x_i \ln\left(1 - \sum_j x_j A_{ji}\right) \tag{11}$$

Where x_i is the mole fraction of species i and A_{ji} adjustable parameters such that A_{ii} = 0 and $A_{ij} \neq A_{ji} \neq 0$.
The activity coefficient is written as follows:

$$\ln \gamma_i = -\ln\left(1 - \sum_j x_{j/i}\right) + 1 - \sum_j \left[x_j\left(1 - A_{i/j}\right)/\left(1 - \sum_k x_k A_{k/j}\right)\right] \tag{12}$$

3. Results and Discussion

Three models (PC-SAFT, PR-MC-WS-NRTL, and PR-vdW-Wilson) have been proposed for the calculation of liquid and vapor mole fractions, as well as pressure. The critical temperature, critical pressure, and acentric factor of pure components, and the PC-SAFT parameter values of the selected compounds used in this study, are provided in Table 1, sourced from literature cited in the references.

The experimental and calculated compositions and pressures of the binary mixtures using (PC-SAFT), (PR-MC-WS-NRTL) model, and (PR-vdW-Wilson) model, respectively, are presented in Table 2 and implemented in Figures 1 through.

The parameters of (PC-SAFT), (PR-MC-WS-NRTL), and (PR-vdW-Wilson) models were obtained by minimizing the following objectivefunction:

$$F_{obj} = \frac{100}{N}\left[\sum_1^N \left(\frac{P_{exp} - P_{cal}}{P_{exp}}\right)^2 + \sum_1^N \left(\frac{z_{exp} - z_{cal}}{z_{exp}}\right)^2\right] \tag{13}$$

In order to assess the goodness of fit of our method, the deviations MRD (Mean Relative Deviation), and the Bias, applied on liquid and vapour phase mole fractions for mixtures, or pressure are calculated using Eq. (14) and Eq. (15) respectively:

$$MRD = \frac{100}{N}\sum\left|\left((U_{exp} - U_{cal})/U_{exp}\right)\right| \tag{14}$$

Table 2. Vapor–liquid equilibrium pressures and phase compositions for the system (R134a + R290) [7], (R152a + R1234ze (Z)) [8], (R152a + R1243zf) [8], and (R1243zf + R134a) [9],Δx and ΔP are deviations in liquid and vapor mole fractions, Calculated values are from PC-SAFT, PR-MC-WS-NRTL, and PR-vdW-Wilson.

Experimental data			PC-SAFT				PR–vdW- WILSON				PR-MC-WS-NRTL			
P_{exp}/ MPa	x_{1exp}	y_{1exp}	P_{cal}/ MPa	y_{1cal}	ΔP/ MPa	Δy_1	P_{cal}/ MPa	y_{1cal}	ΔP/ MPa	Δy_1	x_{1cal}	y_{1cal}	Δx_1	Δy_1
R134a + R290 [7]														
							T =253.15 K							
0.2441	0.0000	0.0000	0.2470	0.0000	0.0030	0.0000	0.2445	0.0000	-0.0004	0.0000	0.0000	0.0000	0.0000	0.0000
0.2686	0.0689	0.1418	0.2750	0.1500	0.0060	0.0080	0.2692	0.1417	-0.0006	0.0001	0.0694	0.1462	-0.0005	-0.0044
0.2844	0.1470	0.2375	0.2900	0.2340	0.0050	-0.0030	0.2844	0.2357	0.0000	0.0018	0.1460	0.2387	0.0010	-0.0012
0.2936	0.2517	0.3037	0.2960	0.2910	0.0030	-0.0120	0.2933	0.3101	0.0003	-0.0064	0.2502	0.3100	0.0015	-0.0063
0.2954	0.3482	0.3501	0.2970	0.3200	0.0010	-0.0300	0.2954	0.3525	0.0000	-0.0024	0.3510	0.3492	-0.0028	0.0010
0.2938	0.4719	0.3830	0.2940	0.3440	0.0000	-0.0390	0.2940	0.3882	-0.0002	-0.0052	0.4736	0.3774	-0.0017	0.0056
0.2883	0.6383	0.3892	0.2840	0.3800	-0.0040	-0.0090	0.2888	0.4195	-0.0005	-0.0303	0.6348	0.4031	0.0035	-0.0139
0.2570	0.8468	0.5130	0.2390	0.5000	-0.0180	-0.0130	0.2762	0.4561	-0.0192	0.0569	0.8548	0.4823	-0.0080	0.0307
0.1319	1.0000	1.0000	0.1320	1.0000	0.0010	0.0000	0.1327	1.0000	-0.0008	0.0000	1.0000	1.0000	0.0000	0.0000
							T =273.15 K							
0.4725	0.0000	0.0000	0.4780	0.0000	0.0060	0.0000	0.4745	0.0000	-0.0020	0.0000	0.0000	0.0000	0.0000	0.0000
0.5093	0.0516	0.1046	0.5180	0.1120	0.0090	0.0070	0.5155	0.1129	-0.0062	-0.0083	0.0523	0.1100	-0.0007	-0.0054
0.5430	0.1256	0.2105	0.5510	0.2080	0.0080	-0.0030	0.5521	0.2184	-0.0091	-0.0079	0.1251	0.2104	0.0007	0.0001
0.5668	0.2248	0.2913	0.5700	0.2800	0.0030	-0.0110	0.5766	0.3051	-0.0098	-0.0138	0.2242	0.2954	0.0006	-0.0041
0.5759	0.3449	0.3606	0.5740	0.3330	-0.0020	-0.0270	0.5866	0.3685	-0.0107	-0.0079	0.3453	0.3592	-0.0004	0.0014
0.5716	0.4951	0.4132	0.5660	0.3820	-0.0060	-0.0320	0.5847	0.4173	-0.0131	-0.0041	0.4942	0.4107	0.0010	0.0025
0.5542	0.6585	0.4459	0.5400	0.4400	-0.0150	-0.0060	0.5737	0.4541	-0.0195	-0.0082	0.6550	0.4576	0.0035	-0.0117
0.5136	0.8035	0.5300	0.4840	0.5330	-0.0290	0.0030	0.5540	0.4896	-0.0404	0.0404	0.8067	0.5243	-0.0028	0.0057
0.2938	1.0000	1.0000	0.2890	1.0000	-0.0040	0.0000	0.2928	1.0000	0.0010	0.0000	1.0000	1.0000	0.0000	0.0000

Table 2. Vapor–liquid equilibrium pressures and phase compositions for the system (R134a + R290) [7], (R152a + R1234ze (Z)) [8], (R152a + R1243zf) [8], and (R1243zf + R134a) [9], Δx and ΔP are deviations in liquid and vapor mole fractions, Calculated values are from PC-SAFT, PR-MC-WS-NRTL, and PR-vdW-Wilson“(continue)”.

T =293.15 K														
0.8365	0.0000	0.0000	0.8420	0.0000	0.0060	0.0000	0.8365	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.8942	0.0508	0.0967	0.9070	0.1020	0.0130	0.0060	0.9015	0.1036	-0.0073	-0.0069	0.0521	0.1011	-0.0013	-0.0044
0.9500	0.1208	0.1962	0.9630	0.1940	0.0130	-0.0020	0.9597	0.2029	-0.0097	-0.0067	0.1207	0.1962	0.0001	0.0000
0.9729	0.1616	0.2379	0.9840	0.2330	0.0110	-0.0050	0.9820	0.2460	-0.0091	-0.0081	0.1603	0.2384	0.0013	-0.0005
0.9971	0.2199	0.2859	1.0020	0.2770	0.0050	-0.0090	1.0034	0.2950	-0.0063	-0.0091	0.2196	0.2894	0.0003	-0.0035
1.0197	0.5370	0.3684	1.0160	0.3450	-0.0040	-0.0230	1.0229	0.3691	-0.0032	-0.0007	0.3455	0.3677	-0.0002	0.0007
1.0088	0.7359	0.5069	0.9930	0.4270	-0.0160	-0.0230	1.0158	0.4413	-0.0070	0.0656	0.5321	0.4489	0.0016	0.0005
0.9241	0.8860	0.6741	0.8830	0.5640	-0.0420	0.0050	0.9585	0.5292	-0.0344	0.1449	0.7719	0.5611	-0.0006	-0.0013
0.5717	1.0000	1.0000	0.5640	1.0000	-0.0080	0.0000	0.5717	1.0000	0.0000	0.0000	1.0000	1.0000	0.0000	0.0000
R152a + R1234ze [8]														
T =273.15 K														
0.2166	0.0000	0.0000	0.2140	0.0000	-0.0030	0.0000	0.2165	0.0000	0.0001	0.0000	0.0000	0.0000	0.0000	0.0000
0.2185	0.0385	0.0503	0.2170	0.0490	-0.0020	-0.0010	0.2222	0.0579	-0.0037	-0.0076	0.0444	0.0624	-0.0059	-0.0121
0.2261	0.1453	0.1778	0.2240	0.1790	-0.0020	0.0010	0.2333	0.1870	-0.0072	-0.0092	0.1483	0.1843	-0.0030	-0.0065
0.2308	0.2306	0.2710	0.2290	0.2760	-0.0010	0.0050	0.2394	0.2752	-0.0086	-0.0042	0.2284	0.2700	0.0023	0.0010
0.2379	0.3639	0.4111	0.2370	0.4180	0.0000	0.0060	0.2465	0.4047	-0.0086	0.0064	0.3640	0.4113	-0.0001	-0.0001
0.2446	0.4863	0.5296	0.2440	0.5390	0.0000	0.0090	0.2514	0.5215	-0.0068	0.0081	0.4933	0.5432	-0.0070	-0.0136
0.2507	0.6311	0.6665	0.2520	0.6740	0.0010	0.0080	0.2559	0.6595	-0.0052	0.0070	0.6099	0.6580	0.0213	0.0085
0.2547	0.7325	0.7567	0.2570	0.7650	0.0020	0.0080	0.2585	0.7556	-0.0038	0.0011	0.6982	0.7446	0.0343	0.0121
0.2592	0.8479	0.8639	0.2610	0.8670	0.0020	0.0030	0.2611	0.8632	-0.0019	0.0007	0.8146	0.8559	0.0333	0.0080
0.2623	0.9537	0.9573	0.2660	0.9590	0.0030	0.0020	0.2631	0.9592	-0.0008	-0.0019	0.9191	0.9555	0.0346	0.0018
0.2636	1.0000	1.0000	0.2670	1.0000	0.0040	0.0000	0.2640	1.0000	-0.0004	0.0000	1.0000	1.0000	0.0000	0.0000
T =293.15 K														
0.4273	0.0000	0.0000	0.4220	0.0000	-0.0050	0.0000	0.4273	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.4318	0.0386	0.0486	0.4270	0.0480	-0.0050	0.0000	0.4322	0.0481	-0.0004	0.0005	0.0436	0.0578	-0.0050	-0.0092
0.4441	0.1443	0.1725	0.4400	0.1740	-0.0040	0.0010	0.4448	0.1730	-0.0007	-0.0005	0.1427	0.1763	0.0016	-0.0038
0.4545	0.2299	0.2661	0.4500	0.2700	-0.0050	0.0040	0.4543	0.2678	0.0002	-0.0017	0.2279	0.2681	0.0020	-0.0020
0.4681	0.3625	0.4033	0.4650	0.4090	-0.0040	0.0060	0.4678	0.4058	0.0003	-0.0025	0.3625	0.4037	0.0000	-0.0004
0.4784	0.4844	0.5239	0.4770	0.5300	-0.0010	0.0060	0.4789	0.5253	-0.0005	-0.0014	0.4790	0.5168	0.0054	0.0071
0.4900	0.6308	0.6623	0.4900	0.6680	0.0000	0.0060	0.4908	0.6625	-0.0008	-0.0002	0.6324	0.6635	-0.0016	-0.0012
0.4974	0.7329	0.7557	0.4990	0.7610	0.0010	0.0050	0.4980	0.7557	-0.0006	0.0000	0.7301	0.7561	0.0028	-0.0004
0.5054	0.8478	0.8613	0.5080	0.8640	0.0020	0.0020	0.5052	0.8600	0.0002	0.0013	0.8354	0.8546	0.0124	0.0067
0.5102	0.9536	0.9569	0.5150	0.9580	0.0040	0.0010	0.5108	0.9569	-0.0006	0.0000	0.9396	0.9553	0.0140	0.0016
0.5124	1.0000	1.0000	0.5170	1.0000	0.0050	0.0000	0.5129	1.0000	-0.0005	0.0000	1.0000	1.0000	0.0000	0.0000
T =313.15 K														
0.7665	0.0000	0.0000	0.7570	0.0000	-0.0100	0.0000	0.7665	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.7735	0.0385	0.0474	0.7660	0.0470	-0.0080	0.0000	0.7752	0.0469	-0.0017	0.0005	0.0430	0.0543	-0.0045	-0.0069
0.7967	0.1441	0.1689	0.7880	0.1700	-0.0080	0.0010	0.7975	0.1689	-0.0008	0.0000	0.1522	0.1816	-0.0081	-0.0127
0.8125	0.2286	0.2609	0.8060	0.2630	-0.0070	0.0030	0.8137	0.2606	-0.0012	0.0003	0.2293	0.2645	-0.0007	-0.0036
0.8359	0.3623	0.3996	0.8310	0.4030	-0.0050	0.0030	0.8367	0.3979	-0.0008	0.0017	0.3628	0.3999	-0.0005	-0.0003
0.8520	0.4847	0.5174	0.8520	0.5230	0.0000	0.0060	0.8551	0.5175	-0.0031	-0.0001	0.4705	0.5047	0.0142	0.0127
0.8726	0.6301	0.6584	0.8740	0.6610	0.0010	0.0020	0.8740	0.6549	-0.0014	0.0035	0.6307	0.6568	-0.0006	0.0017
0.8847	0.7319	0.7506	0.8870	0.7540	0.0020	0.0040	0.8856	0.7496	-0.0009	0.0010	0.7378	0.7571	-0.0059	-0.0065

Table 2. Vapor–liquid equilibrium pressures and phase compositions for the system (R134a + R290) [7], (R152a + R1234ze (Z)) [8], (R152a + R1243zf) [8], and (R1243zf + R134a) [9], Δx and ΔP are deviations in liquid and vapor mole fractions, Calculated values are from PC-SAFT, PR-MC-WS-NRTL, and PR-vdW-Wilson"(continue)".

0.8961	0.8474	0.8585	0.9000	0.8600	0.0040	0.0010	0.8969	0.8568	-0.0008	0.0017	0.8477	0.8594	-0.0003	-0.0008

0.9049	0.9535	0.9566	0.9100	0.9570	0.0050	0.0000	0.9058	0.9560	-0.0009	0.0006	0.9501	0.9551	0.0034	0.0016
0.9082	1.0000	1.0000	0.9140	1.0000	0.0060	0.0000	0.9093	1.0000	-0.0011	0.0000	1.0000	1.0000	0.0000	0.0000
R152a + R1243zf [8]														
							T =273.15 K							
0.2682	0.0000	0.0000	0.2690	0.0000	0.0010	0.0000	0.2695	0.0000	-0.0013	0.0000	0.0000	0.0000	0.0000	0.0000
0.2704	0.0837	0.0902	0.2710	0.0890	0.0000	-0.0010	0.2710	0.0880	-0.0006	0.0022	0.0815	0.0870	0.0022	0.0033
0.2735	0.2127	0.2200	0.2730	0.2210	0.0000	0.0010	0.2726	0.2171	0.0009	0.0029	0.2088	0.2164	0.0039	0.0037
0.2744	0.3103	0.3134	0.2740	0.3170	0.0000	0.0040	0.2732	0.3113	0.0012	0.0021	0.3074	0.3119	0.0029	0.0016
0.2747	0.4082	0.4093	0.2750	0.4120	0.0000	0.0030	0.2733	0.4043	0.0014	0.0050	0.4080	0.4088	0.0002	0.0005
0.2750	0.5073	0.5029	0.2750	0.5070	0.0000	0.0040	0.2729	0.4981	0.0021	0.0048	0.5067	0.5035	0.0006	-0.0006
0.2742	0.5961	0.5885	0.2750	0.5920	0.0000	0.0030	0.2722	0.5827	0.0020	0.0058	0.5979	0.5923	-0.0018	-0.0038
0.2734	0.6891	0.6763	0.2750	0.5850	0.0010	-0.0910	0.2710	0.6729	0.0024	0.0034	0.6874	0.6785	0.0017	-0.0022
0.2710	0.8159	0.8000	0.2720	0.8060	0.0010	0.0060	0.2688	0.8003	0.0022	-0.0003	0.8139	0.8015	0.0020	-0.0015
0.2674	0.9206	0.9094	0.2690	0.9190	0.0020	0.0100	0.2663	0.9112	0.0011	-0.0018	0.9235	0.9137	0.0024	-0.0042
0.2636	1.0000	1.0000	0.2670	1.0000	0.0040	0.0000	0.2640	1.0000	-0.0004	0.0000	1.0000	1.0000	0.0000	0.0000
							T =293.15 K							
0.5077	0.0000	0.0000	0.5080	0.0000	0.0010	0.0000	0.5100	0.0000	-0.0023	0.0000	0.0000	0.0000	0.0000	0.0000
0.5129	0.0824	0.0910	0.5130	0.0890	0.0000	-0.0020	0.5182	0.0913	-0.0053	-0.0003	0.0867	0.1242	-0.0043	-0.0332
0.5186	0.2125	0.2218	0.5200	0.2240	0.0010	0.0030	0.5250	0.2180	-0.0064	0.0038	0.2172	0.2234	-0.0047	-0.0016
0.5218	0.3094	0.3176	0.5230	0.3210	0.0010	0.0040	0.5270	0.3082	-0.0052	0.0094	0.3135	0.3144	-0.0041	0.0032
0.5241	0.4088	0.4130	0.5260	0.4180	0.0020	0.0050	0.5274	0.4009	-0.0033	0.0121	0.4109	0.4146	-0.0021	-0.0016
0.5254	0.5070	0.5083	0.5280	0.5120	0.0020	0.0030	0.5266	0.4943	-0.0012	0.0140	0.5077	0.5143	-0.0007	-0.0060
0.5253	0.5959	0.5948	0.5280	0.5960	0.0030	0.0410	0.5250	0.5809	0.0003	0.0139	0.5954	0.6021	0.0005	-0.0072
0.5248	0.6889	0.6824	0.5280	0.6850	0.0030	0.0030	0.5229	0.6738	0.0019	0.0086	0.6857	0.6894	0.0032	-0.0070
0.5225	0.8156	0.8057	0.5250	0.8080	0.0030	0.0030	0.5192	0.8040	0.0033	0.0017	0.8110	0.8107	0.0047	-0.0050
0.5169	0.9203	0.9122	0.5210	0.9150	0.0040	0.0020	0.5157	0.9145	0.0012	-0.0023	0.9117	0.9163	0.0086	-0.0041
0.5124	1.0000	1.0000	0.5170	1.0000	0.0050	0.0000	0.5129	1.0000	-0.0005	0.0000	1.0000	1.0000	0.0000	0.0000
							T =313.15 K							
0.8809	0.0000	0.0000	0.8810	0.0000	0.0000	0.0000	0.8843	0.0000	-0.0034	0.0000	0.0000	0.0000	0.0000	0.0000
0.8906	0.0831	0.0916	0.8910	0.0900	0.0000	-0.0020	0.8999	0.0899	-0.0093	0.0017	0.0874	0.1056	-0.0043	-0.0140
0.9033	0.2121	0.2224	0.9030	0.2250	0.0000	0.0020	0.9134	0.2150	-0.0101	0.0074	0.2173	0.2419	-0.0051	-0.0195
0.9106	0.3098	0.3194	0.9110	0.3230	0.0000	0.0030	0.9185	0.3068	-0.0079	0.0126	0.3146	0.3357	-0.0048	-0.0163
0.9156	0.4075	0.4161	0.9170	0.4190	0.0010	0.0030	0.9209	0.3995	-0.0053	0.0166	0.4118	0.4268	-0.0043	-0.0107
0.9199	0.5079	0.5149	0.9210	0.5160	0.0010	0.0010	0.9213	0.4969	-0.0014	0.0180	0.5114	0.5190	-0.0035	-0.0041
0.9218	0.5959	0.5976	0.9240	0.6000	0.0020	0.0020	0.9205	0.5841	0.0013	0.0135	0.5968	0.5987	-0.0009	-0.0011
0.9215	0.6898	0.6881	0.9250	0.6900	0.0030	0.0020	0.9187	0.6791	0.0028	0.0090	0.6890	0.6868	0.0009	0.0013
0.9201	0.8153	0.8095	0.9230	0.8120	0.0030	0.0020	0.9153	0.8082	0.0048	0.0013	0.8124	0.8070	0.0029	0.0025
0.9138	0.9200	0.9150	0.9190	0.9170	0.0050	0.0020	0.9120	0.9169	0.0018	-0.0019	0.9186	0.9163	0.0014	-0.0013
0.9082	1.0000	1.0000	0.9140	1.0000	0.0060	0.0000	0.9093	1.0000	-0.0011	0.0000	1.0000	1.0000	0.0000	0.0000
R1243zf + R134a [9]														
							T =243.15 K							
0.0845	0.0000	0.0000	0.0840	0.0000	-0.0010	0.0000	0.0843	0.0000	-0.0843	0.0000	0.0000	0.0000	0.0000	0.0000
0.0855	0.1640	0.1770	0.0860	0.1770	0.0000	0.0000	0.0858	0.1751	-0.0858	0.0019	0.1674	0.1730	-0.0034	0.0040

Table 2. Vapor–liquid equilibrium pressures and phase compositions for the system (R134a + R290) [7], (R152a + R1234ze (Z)) [8], (R152a + R1243zf) [8], and (R1243zf + R134a) [9], Δx and ΔP are deviations in liquid and vapor mole fractions, Calculated values are from PC-SAFT, PR-MC-WS-NRTL, and PR-vdW-Wilson"(continue)".

0.0861	0.2310	0.2460	0.0860	0.2440	0.0000	-0.0020	0.0863	0.2427	-0.0863	0.0033	0.2385	0.2410	-0.0075	0.0051

0.0869	0.3620	0.3670	0.0870	0.3680	0.0000	0.0010	0.0869	0.3699	-0.0869	-0.0029	0.3645	0.3564	-0.0025	0.0106
0.0866	0.4760	0.4750	0.0870	0.4740	0.0000	-0.0010	0.0870	0.4770	-0.0870	-0.0020	0.4755	0.4604	0.0005	0.0146
0.0866	0.5820	0.5800	0.0870	0.5720	0.0000	-0.0080	0.0869	0.5755	-0.0869	0.0045	0.5810	0.5586	0.0010	0.0214
0.0857	0.6740	0.6530	0.0860	0.6590	0.0000	0.0060	0.0865	0.6615	-0.0865	-0.0085	0.6635	0.6420	0.0105	0.0110
0.0846	0.8230	0.8040	0.0850	0.8060	0.0000	0.0020	0.0854	0.8064	-0.0854	-0.0024	0.8135	0.7852	0.0095	0.0188
0.0835	0.9120	0.8930	0.0840	0.9010	0.0000	0.0080	0.0843	0.8995	-0.0843	-0.0065	0.9025	0.8515	0.0095	0.0415
0.0819	1.0000	1.0000	0.0830	1.0000	0.0010	0.0000	0.0828	1.0000	-0.0828	0.0000	1.0000	1.0000	0.0000	0.0000
T =263.15 K														
0.2010	0.0000	0.0000	0.1990	0.0000	-0.0020	0.0000	0.2006	0.0000	-0.2006	0.0000	0.0000	0.0000	0.0000	0.0000
0.2020	0.1630	0.1670	0.2010	0.1690	-0.0010	0.0020	0.2014	0.1634	-0.2014	0.0036	0.1630	0.1671	0.0000	-0.0001
0.2030	0.2440	0.2460	0.2010	0.2470	-0.0010	0.0010	0.2016	0.2436	-0.2016	0.0024	0.2450	0.2427	-0.0010	0.0033
0.2030	0.3510	0.3490	0.2010	0.3480	-0.0010	-0.0010	0.2016	0.3482	-0.2016	0.0008	0.3500	0.3335	0.0010	0.0155
0.2020	0.4660	0.4550	0.2010	0.4540	-0.0010	-0.0010	0.2011	0.4590	-0.2011	-0.0040	0.4605	0.4285	0.0055	0.0265
0.2010	0.5640	0.5480	0.2000	0.5460	-0.0010	-0.0020	0.2002	0.5526	-0.2002	-0.0046	0.5560	0.5157	0.0080	0.0323
0.1980	0.7060	0.6830	0.1970	0.6830	-0.0010	0.0000	0.1981	0.6885	-0.1981	-0.0055	0.6945	0.6642	0.0115	0.0188
0.1960	0.8060	0.7850	0.1940	0.7840	-0.0020	-0.0010	0.1958	0.7868	-0.1958	-0.0018	0.7955	0.7760	0.0105	0.0090
0.1920	0.8910	0.8760	0.1920	0.8750	0.0000	-0.0010	0.1931	0.8748	-0.1931	0.0012	0.8924	0.8718	-0.0014	0.0042
0.1870	1.0000	1.0000	0.1880	1.0000	0.0010	0.0000	0.1881	1.0000	-0.1881	0.0000	1.0000	1.0000	0.0000	0.0000
T =283.15 K														
0.4140	0.0000	0.0000	0.4090	0.0000	-0.0050	0.0000	0.4146	0.0000	-0.4146	0.0000	0.0000	0.0000	0.0000	0.0000
0.4150	0.1630	0.1630	0.4120	0.1640	-0.0030	0.0010	0.4157	0.1635	-0.4157	-0.0005	0.1603	0.1630	0.0027	0.0000
0.4150	0.2450	0.2410	0.4120	0.2430	-0.0030	0.0020	0.4153	0.2422	-0.4153	-0.0012	0.2470	0.2430	-0.0020	-0.0020
0.4130	0.3580	0.3470	0.4100	0.3480	-0.0030	0.0010	0.4137	0.3485	-0.4137	-0.0015	0.3737	0.3525	-0.0157	-0.0055
0.4090	0.4680	0.4400	0.4080	0.4500	-0.0010	0.0100	0.4108	0.4507	-0.4108	-0.0017	0.4699	0.4373	-0.0019	0.0027
0.4060	0.5660	0.5420	0.4040	0.5410	-0.0020	-0.0010	0.4071	0.5423	-0.4071	-0.0003	0.5540	0.5160	0.0120	0.0260
0.3980	0.7110	0.6800	0.3960	0.6820	-0.0010	0.0020	0.3996	0.6819	-0.3996	-0.0019	0.6955	0.6666	0.0155	0.0134
0.3920	0.8060	0.7820	0.3900	0.7800	-0.0020	-0.0020	0.3932	0.7786	-0.3932	0.0034	0.7940	0.7806	0.0120	0.0014
0.3850	0.8770	0.8600	0.3850	0.8570	0.0000	-0.0030	0.3874	0.8550	-0.3874	0.0050	0.8743	0.8685	0.0027	-0.0085
0.3740	1.0000	1.0000	0.3740	1.0000	0.0010	0.0000	0.3754	1.0000	-0.3754	0.0000	1.0000	1.0000	0.0000	0.0000

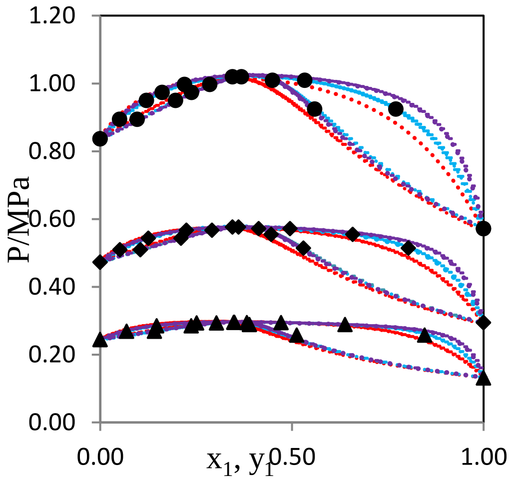

Figure 1. VLE for the system R134a + R290 [7] at different temperatures: (▲) 253.15 K. (♦) 273.15 K. (●) 293.15 K; .... PC-SAFT model........... PR-MC-WS-NRTL model........... PR-vdW- Wilson model.

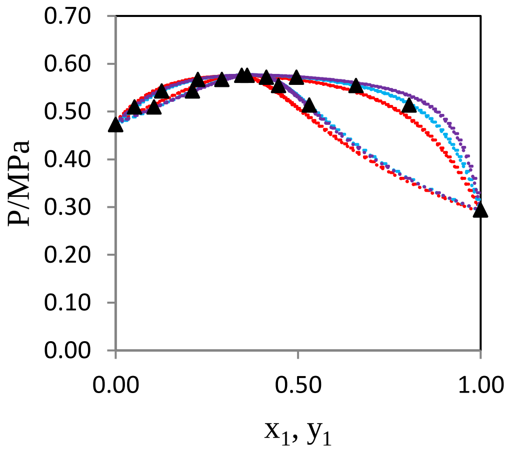

Figure 2. VLE for the system R134a + R290[7] at (▲) 273.15 K.......... PC-SAFT model. .... PR-MC-WS-NRTL model. .... PR-vdW- Wilson model.

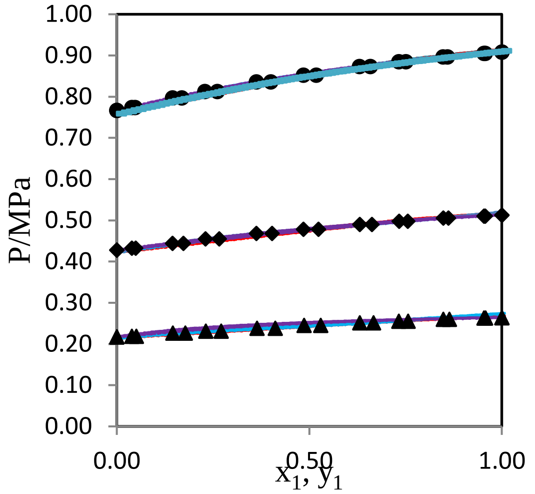

Figure 3. VLE for the system R152a + R1234ze[8] at different temperatures: (▲) 273.15 K. (♦) 293.15 K. (●) 313.15 K,.......... PC-SAFT model. .... PR-MC-WS-NRTL model. .... PR-vdW- Wilson model.

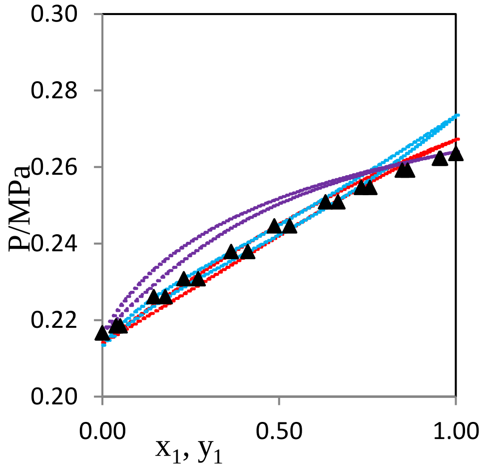

Figure 4. VLE for the system R152a + R1234ze [8] at (▲) 273.15 K.......... PC-SAFT model. .... PR-MC-WS-NRTL model. .... PR-vdW- Wilson model.

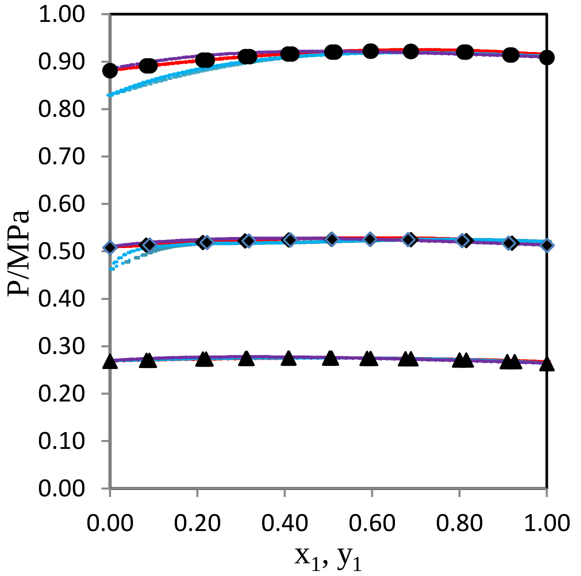

Figure 5. VLE for the system R152a + R1243zf[8] at different temperatures: (▲) 273.15 K. (♦) 293.15 K. (●) 313.15 K,.......... PC-SAFT model. .... PR-MC-WS-NRTL model. .... PR-vdW- Wilson model.

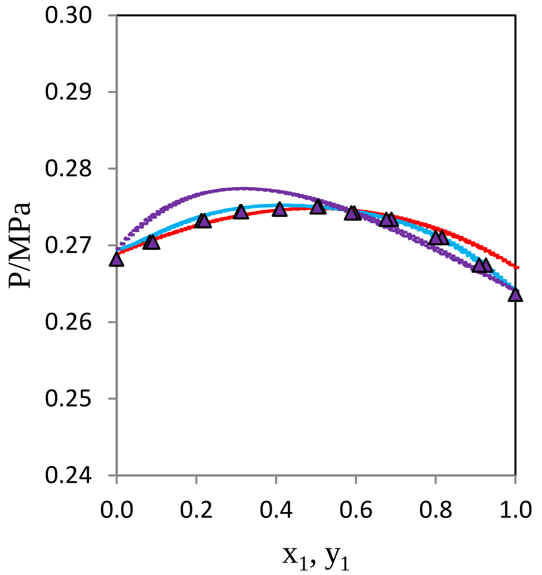

Figure 6. VLE for the system R152a + R1243zf[8] at (▲) 273.15 K.......... PC-SAFT model. .... PR-MC-WS-NRTL model. .... PR-vdW- Wilson model.

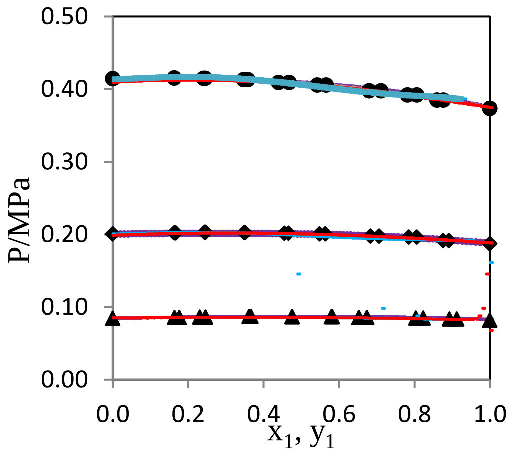

Figure 7. VLE for the system R1243zf + R134a[9] at different temperatures: (▲) 243.15 K. (♦) 263.15 K. (●) 283.15 K; .... PC-SAFT model. .... PR-MC-WS-NRTL model. .... PR-vdW- Wilson model.

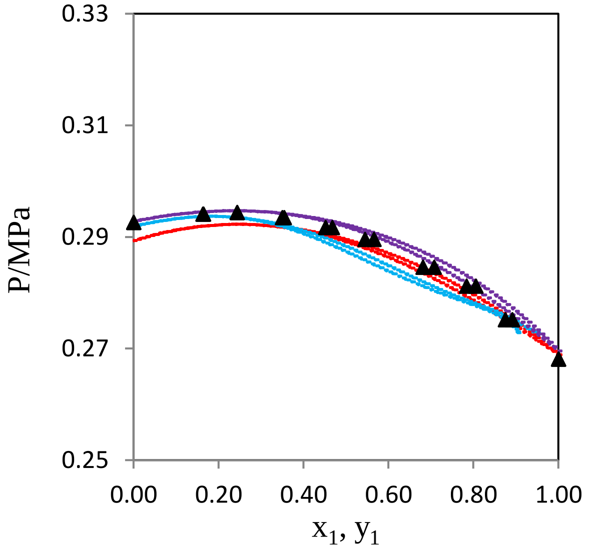

Figure 8. VLE for the system R1243zf + R134a [9] at (▲) 273.15 K.......... PC-SAFT model. .... PR-MC-WS-NRTL model. .... PR-vdW- Wilson model.

Table 3. Pure-component parameters for the PC-SAFT equation of state.

m_1	m_2	m_3	σ_1 [Å]	σ_2 [Å]	σ_3 [Å]	$(\varepsilon/k)_1$ (K)	$(\varepsilon/k)_2$ (K)	$(\varepsilon/k)_3$ (K)
R134a +R290 [7]								
3.54	2.12	2.26	3.09	3.63	3.76	160.60	199.46	216.53
R152a + R1234ze [8]								
3.06	3.43	2.26	3.17	3.26	3.76	176.20	166.18	216.53
R152a + R1243zf [8]								
3.06	2.99	2.26	3.17	3.45	3.76	176.21	173.56	216.53
R1243zf + R134a [9]								
2.99	3.54	2.26	3.45	3.09	3.76	173.56	160.60	216.53

Table 4. Mathias–Copeman coefficients.

Coefficients	R134a	R290	R152a	R1234ze	R1243zf
C_1	0.850	0.600	0.835	0.861	0.850
C_2	0.007	-0.006	-0.631	-0.240	0.007
C_3	-0.054	0.174	0.790	0.602	-0.054

$$\mathrm{Bias} = \frac{100}{N}\sum\left((U_{exp} - U_{cal})/U_{exp}\right) \qquad (15)$$

Where N is the number of data points, and $U = x_i.y_i$ or P.

Figures (1-8) illustrate the variation of pressure as a function of the mole fraction of the most volatile pure component at different isotherms for the four binary systems: R134a + R290, 152a + R1234ze, R152a + R1243zf, and R1243zf + R134a, respectively. The curves represented by symbols depict the experimental values extracted from the literature, while the dashed curves in different colors represent the results calculated by the three chosen models (PC-SAFT, PR-MC-WS-NRTL and PR-vdW-Wilson). The liquid-vapour equilibrium data of these systems were studied within specific temperature interval: three isotherms (253.15, 273.15 and 293.15) K for the system R134a + R290, three isotherms (273.15. 293.15 and 313.15) K for the system R152a + R1234ze, three isotherms (273.15, 293.15 and 313.15) K for the R152a + R1243zf system, and three isotherms too (243.15, 263.15, 283.15) K for the R1243zf + R134a system. The R134a + R290 system exhibits homogeneous azeotropic behavior at maximum pressure, while the others are non-azeotropic systems. The boiling and dew curves of these binary systems are well represented by the three proposed models, where there is a good agreement between the points calculated using the chosen models and the experimental data. The values of the bias and MRD of each isotherm of the binary systems are illustrated in the Table 4.The relative differences with PC-SAFT model do not exceed (1.78 % for pressure, and 3.86 % for the vapour mole fraction), (1.47 % for pressure, and 3.87 % for the vapour mole fraction) with the PR-vdW-Wilson model, and (2.42 % for vapour mole fraction. and 0.68 % for the liquid mole fraction) with the PR-MC-WS-NRTL model for the R134a + R290 system. The relative differences with PC-SAFT model do not exceed (0.76 % for pressure. and 0.95 % for the vapour mole fraction), (1.80 % for pressure, and 2.93 % for the vapour mole fraction) with the PR-vdW-Wilson model, and (3.85 % for vapour mole fraction, and 3.93 % for the liquid mole fraction) with the PR-MC-WS-NRTL model for the R152a + R1234ze system. The relative differences with PC-SAFT model do not exceed (0.43 % for pressure, and 1.84 % for the vapour mole fraction), (0.54 % for pressure, and 2.82 % for the vapour mole fraction) with the PR-vdW-Wilson model, and (2.24 % for vapour mole fraction, and 1.29 % for the liquid mole fraction) with the PR-MC-WS-NRTL model for the R152a + R1243zf system. The relative differences with PC-SAFT model do not exceed (0.56 % for pressure, and 0.51 % for the vapour mole fraction), (0.56 % for pressure, and 0.84 % for the vapour mole fraction) with the PR-vdW-Wilson model, and (2.83 % for vapour mole fraction. and 1.67 % for the liquid mole fraction) with the PR-MC-WS-NRTL model for the R1243zf + R134a system.

4. Conclusion

The research aims to utilize three models, namely PC-SAFT, PR-MC-WS-NRTL, and PR-vdW-Wilson, for the computation of liquid and vapour mole fractions as well as pressure in binary systems. The investigation focuses on four binary systems: R134a + R290, 152a + R1234ze,R152a + R1243zf, and R1243zf + R134a. The study explores the liquid-vapor equilibrium data across different temperature intervals for each system. Notably, the R134a + R290 system exhibits homogeneous azeotropic behaviour at maximum pressure, while the others are non-azeotropic. The boiling and dew curves of these binary systems are effectively represented by the three proposed models. Demonstrating a favorable agreement between calculated and experimental data. The research evaluates the bias and mean relative differences (MRD) for each isotherm, with the relative differences for the liquid and vapor phases within acceptable limits, as exemplified by the R134a + R290 system.

List of symbols

$\bar{a}$	Reduced Helmholtz free energy
A	Parameter of the equation of state (energy parameter [J. m^3.mol^{-2}]).
B	Parameter of the equation of state (molar covolume parameter [m^3.mol^{-1}])
C	Cubic term of the Helmholtz free energy equation.
D	segment diameter of component
EoS	Equation of State
F_{obj}	objective function
G	Molar Gibbs energy [J. mol^{-1}].
K	Boltzmann constant
k_{ij}	Binary interaction parameter.
MC	Mathias-Copeman
NRTL	Non-Random- Two-Liquids
P	Pressure [MPa].
PR	Peng-Robinson
SAFT	Statistical Associating Fluid Theory
R	Gas constant [J.mol^{-1}.K^{-1}]
RE	Relative Error ($\Delta U, \Delta P$)
T	Temperature[K]
VLE	Vapour-Liquid Equilibrium
x	Liquid mole fraction
y	Vapor mole fraction
U	represents x, y or P
V	Total volume [m^3]
vdW	van der Waals
WS	Wong-Sandler

Greekletters

a	Alpha function.
σ	segment diameter
ω	Acentric factor

Subscripts and superscripts

assoc	associating interactions
c	Pure-component critical property.
cal	Calculated property.
chain	chain formation
Exp	Experimental property.
res	Residual property
i,j	Molecular species.

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