Isothermal vapour-liquid equilibrium measurements for the (R1234ze(E) + ethane) system at temperatures from 272.27 to 347.52 K

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Abstract

In this paper, new vapour-liquid equilibrium data of the (ethane + R1234ze(E)) binary system are presented. The measurements are performed at six isotherms ranging from 272.27 to 347.52 K and for pressures up to 4.89 MPa. The experiments were conducted by means of a "static-analytic" apparatus with phase analysis via gas chromatography, with resulting uncertainties of 0.03 K for temperature, 0.4 kPa for pressure and 0.0043 for vapour and liquid mole fractions.

The experimental data were correlated with Peng-Robinson equation of state using the classical van der Waals mixing rules. For the three temperatures above the critical temperature of pure ethane, the critical compositions and pressures of the binary mixtures were also calculated based on the extended asymptotic behaviour laws. The results obtained show a good agreement between experimental measurements and modelling, with a maximal deviation of 1.13% for bubble pressures and 1.23% for vapour molar fractions.

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1. Introduction

Over the last decades, fluorinated compounds have been thought to be one of the most important contributors to global warming potential (GWP). Refrigerants whose GWP value is above 150 are strictly prohibited in the domestic air conditioners and chillers according to the NO.517/2014 regulation[1] issued in May, 2014. Besides, according to the Kigali mendment to the Montreal Protocol, about 80 to 85 % of HFCs refrigerants will be phased out by the late 2040s[2]. Traditional refrigerants such as R134a have been widely used in the domestic field and commercial use. However, its high GWP value, which can reach 1300[3], has restricted its development. Consequently, refrigerants such as hydrofluorocarbons(HFCs), especially the fluorinated compounds which have a high GWP value will be banned or their use will be significantly reduced.

In recent years, finding new refrigerants to replace the traditional ones that are harmful to the ozone layer has been a hot research topic around the world. Hydrocarbons have been thought to be one of the most suitable substitutes but their GWP values are not negligible. Generally, hydrocarbons have smaller liquid densities than most of the fluorocarbons. Hence, the amount of charge decreases significantly with hydrocarbons which will help further relieve the direct emission of refrigerants. Among the hydrocarbons, ethane (R170), with a GWP value of 20, has been thought as a promising refrigerant. But, its flammability may cause concerns when applied in an application. It is believed that mixtures containing ethane can overcome these disadvantages and present a better thermal properties and a good performance as alternative refrigerants mixture. Currently, R1234ze(E) is considered to be one of the most promising new-generation refrigerant, categorized under the family of hydrofluoroolefines (HFOs). This refrigerant has a zero ozone depletion potential (ODP), a GWP of six and an extremely short atmospheric lifetime[4, 5]. It is an environmentally friendly refrigerant and can be widely used in the industrial house-hold areas.

Many scholars have published their experimental vapour-liquid equilibrium data of ethane or R1234ze(E) with other mixtures. In 2013, Nandi et al.[6] reported their experimental isothermal vapour-liquid equilibrium (VLE) data of ethane + n-perfluorooctane binary mixture with five isotherms ranging from 308.45 to 338.43 K and pressures up to 5.69 MPa. In their work, the VLE data were correlated with Peng-Robinson equation of state (PR-EoS)[7] associated with Mathias-Copeman α-function[8], and with Wong-Sandler (WS) mixing rules[9] incorporating the NRTL excess Gibbs energy model, and also with PR-EoS with the classical one-fluid mixing rules. The experimental results indicate that the thermodynamic model with the WS mixing rules produced a better correlation of the experimental data than did the classical one-fluid mixing rules. Kochenburger et al.[10] measured the VLE data of (ethane + R23) binary mixture by means of a "vapour recirculation apparatus". The results were also correlated with PR-EoS with van der Waals mixing rules, Mathias-Klotz-Prausnitz mixing rules[11] and Mathias-Copeman α-function. A positive azeotrope was found in the (ethane + R23) system. Other scholars have also done researches on R1234ze(E). In 2016, pressure-density-temperature-composition (P -ρ- T-x) measurements for (R1234ze(E) + R134a) binary mixture along with seven isotherms from 270.35 to 300.28 K were performed by Zhang et al.[12]. The PR-EoS with classical van der Waals one-fluid mixing rules and a

truncated virial equation of state were employed to correlate the experimental data of this system. Moreover, different models were employed to correlate the experimental data, and a good agreement was found between calculated and experimental results. Dong et al.[13] also measured the isothermal VLE values for the system of (R1234ze(E) + R134) at four temperatures from 258.15 to 288.15 K with an apparatus based on the "static-analytic" method. The correlation of VLE results were carried out by means of PR-EoS and Huron-Vidal mixing rules[14] involving the NRTL activity coefficient model. A good consistency is found between the experimental and calculated values through this model at each experimental point. The results show that an azeotropic behaviour is observed for this binary system. Besides, many other researchers (Di Nicola et al.[15], Koyama et al.[16]) have also measured the VLE data of the mixtures containing R1234ze(E).

However, and to our knowledge, no data in the open literature can be found about the VLE data of (ethane + R1234ze(E)) binary mixture. In this work, the VLE data of this system were measured at six temperatures, three below ethane critical temperature and three above it. The measured data were correlated using PR-EoS with the classical van der Waals mixing rules. The experimental results in this work will help extend the database of the new refrigerant R1234ze(E) and its mixtures. Besides, the refrigerant ethane shows good performance in cryogenic systems. The research of ethane+R1234ze(E) will help promote the application of R1234ze(E) and its mixtures into industrial use such as the cryogenic field and ultra-cryogenic field after being well researched.

2. Experimental section

Materials. In Table 1 we list the suppliers of the two chemicals and the purities (given by the suppliers and verified by gas chromatography (GC) analysis). Except from a degassing at the moment of loading the products into the equilibrium cell, no further purification of the products was needed.

Table 1. Purities and suppliers of the chemicals used in this work

Chemicals	CAS Registry Number	Purity (GC)	Supplier
Ethane	74-84-0	99.95 vol%	MESSER
R1234ze(E)	29118-24-9	≥ 99.5 wt%	Climalife

Experimental apparatus. The measurements were conducted based on a static-analytic apparatus[17]. As it is shown in Figure 1, the key part of the apparatus is the equilibrium cell, which consists of a transparent tube held between two titanium flanges. The top flange is connected to a 100 Ω platinum resistance thermometer probe (Pt100), a pressure transducer, two inlet valves and two samplers. The bottom flange is connected to a Pt100 probe and two outlet valves. The temperature of the cell is measured by the two Pt100 probes, which measure the temperature at the top and the bottom of the cell, respectively. The temperature probes were calibrated against a standard probe (25 Ohms, TINSLEY, U.K.) which was certified by the 'Laboratoire National d'Essai' (Paris, France) based on the 1990 International

Temperature Scale (ITS 90). The accuracy of the platinum probes is ±0.03 K. The cell's pressure is measured by a pressure transducer (DRUCK, Model UNIK 500). Generally, two transducers are used to verify the precision of the measurement. One transducer is used for the low pressures measurements (0-50 bar) and another one for the high pressures measurements (up to 200 bar). The pressure transducers were calibrated with respect to a pressure automated calibration equipment (PACE 5000, GE Sensing and Inspection Technologies). The accuracies of the two pressure transducers are ±0.3 kPa and ±0.4 kPa respectively. In this study, the pressure in the cell was not higher than 50 bar, so only the low pressure transducer was used during the experiments. Before the measurements, the thermometer probes and the pressure transducers were calibrated. Both the thermometers and the pressure transducer are connected to an "Agilent" data acquisition system which is linked to a PC and provides real time reading of the values of temperature and pressure. The equilibrium cell is immersed in a liquid bath to maintain the desired temperature, and the temperature of the liquid bath is controlled by a PID controller, which provides and keeps the desired temperature with ±0.01 K. A magnetic stirrer coupled with an electric motor (HELDOLPH, model RZR2020) was held between the two flanges and help accelerate the mixing of the mixture. After the mixture reaches equilibrium, two electromagnetic capillary samplers ROLSI® are used to sample a small amount of each phase without perturbing the equilibrium conditions and send them into the GC to analyze the compositions of the vapour and liquid phases. The relative accuracies of mole numbers are ±1.2% for ethane and ±0.9 for R1234ze(E). Consequently the maximum uncertainty of mole fractions, calculated at $x_{ethane} = 0.5$ is $u_{max}(x,y) = 0.0043$.

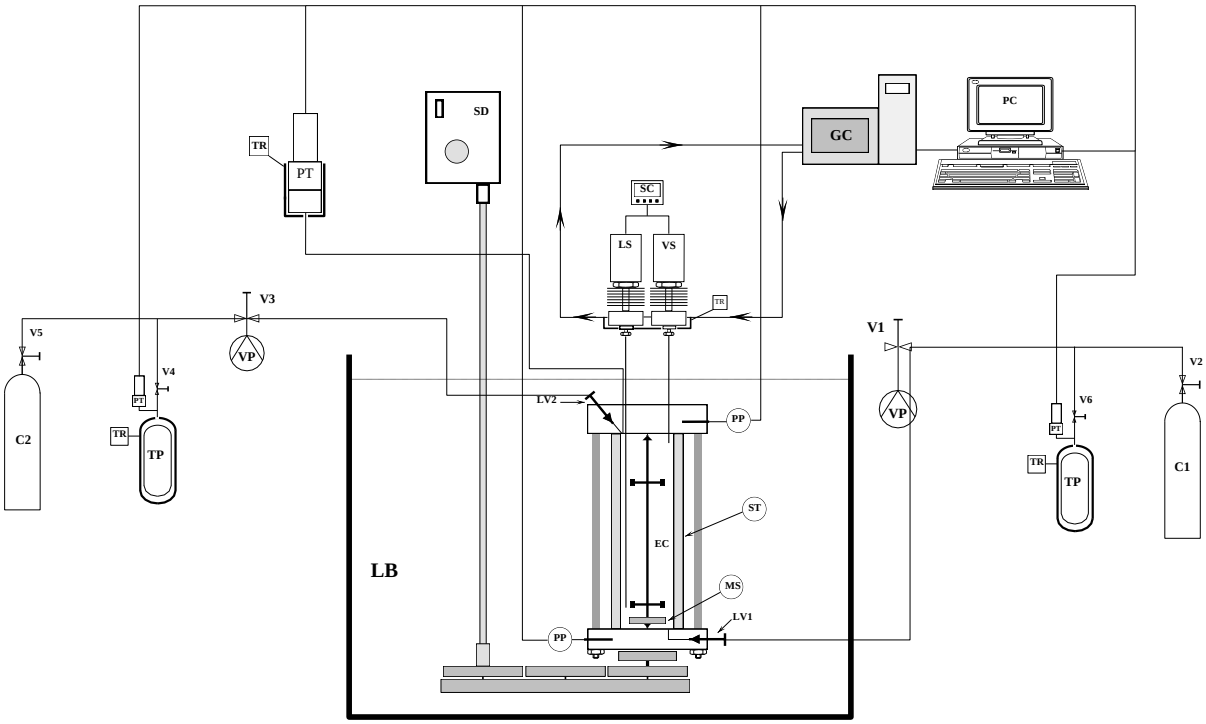


Figure 1. Schematic diagram of the static-analytic apparatus. C1: more volatile compound; C2: less

volatile compound; EC: equilibrium cell; GC: gas chromatograph; LB: liquid bath; LS: liquid sampler; LV: loading valve; MS: magnetic stirrer; PC: personal computer; PP: platinum resistance thermometer probe; PT: pressure transducer; SC: sample controlling; SD: stirring device; ST: sapphir tube; TP: thermal press; TR: temperature regulator; Vi: valve; VP: vacuum pump; VS: vapour sampler.

Experimental Procedure. First of all, the equilibrium cell and the connecting parts are evacuated and put under vacuum by means of a vacuum pump. About 5 cm^3 of the heavier component (R1234ze(E) in this study) is introduced into the cell. Thereafter, a small amount of ethane is introduced into the equilibrium cell from a thermal press (where it is stored). It is assumed to reach the equilibrium conditions when the temperatures and the pressures stay nearly constant for at least 10 min, under continuous stirring. Once the equilibrium is obtained, the liquid and vapour samples are sent to the GC for analysis. For each equilibrium point, at least six samples are taken, for the determination of the average composition and to check the measurements repeatability (less than 1%). Then a further volume of ethane is introduced into the cell and another measurement is started. This procedure is continued until the existence points can cover the whole composition range. Once one isotherm is finished, the cell is emptied, put under vacuum and the same procedure is started for another isotherm.

3. Data treatment and modelling

The critical temperatures (T_c) and pressures (P_c) and acentric factors (ω) for pure ethane and R1234ze(E) which are collected from the software *REFPROP v9.0*[18] are provided in Table 2.

Table 2. Thermal properties for ethane and R1234ze(E) pure component (Source REFPROP 9.0)

Component	T_c /K	P_c /MPa	Acentric factor ω
Ethane	305.32	4.8722	0.0995
R1234ze(E)	382.52	3.6363	0.313

The experimental VLE data of (ethane + R1234ze(E)) system was correlated with a homemade software developed at the Centre of Thermodynamics of Processes (CTP) - MINES ParisTech. The data were correlated using PR-EoS[7] combined with the classical van der Waals mixing rules.

For the three isotherms higher than the critical temperature of pure ethane, the extended asymptotic behaviour laws described by Ungerer et al.[19] (Eqs. (1) and (2)) were used for the data treatment and the estimation of critical compositions and pressures.

$$y - x = \lambda_1(P_c - P) + \mu(P_c - P)^{\beta} \quad (1)$$

$$\frac{x+y}{2} - x_c = \lambda_2(P_c - P) \quad (2)$$

Where x and y stand for the molar fraction of the liquid phase and vapour phase at pressure P, respectively. x_c and P_c represent the critical composition and critical pressure, respectively. β is a characteristic universal exponent, whose value equals to 0.325. λ_1, λ_2 and μ are adjustable coefficients regressed from a set of several isothermal coexistence data points (P, x, y) below the critical point.

4. Results and Discussions

The experimental VLE measurements of (ethane + R1234ze(E)) mixture were performed at six temperatures (272.27, 283.36, 298.04, 308.04, 323.18 and 347.52) K and for pressures up to 4.89 MPa. In addition, the experimental data were correlated with PR-EoS. The experimental VLE data and the calculated ones are reported in Table 3.

In Figure 2, we present the experimental data and the correlated ones with PR-EoS. From this Figure, it is obvious that PR-EoS makes a good calculation for the VLE of the studied mixture, especially for the isotherms lower than the critical temperature of pure ethane. Figure 3 indicates the composition dependency of the relative volatility for the measured temperatures. A very good agreement between the experimental relative volatility and the calculated values using PR-EoS can be observed.

Table 3. Experimental and Calculated VLE data of (ethane (1) + R1234ze(E) (2)) system[a].

Experimental data							Calculated data			
P_{exp}/MPa	n_x[b]	$x_{1,exp}$	u(x)[a]	n_y[b]	$y_{1,exp}$	u(y)[a]	P_{cal}/MPa	$y_{1,cal}$	ΔP[b]/MPa	Δy[b]
272.27 K										
0.3904	6	0.0511	0.0004	6	0.4650	0.0023	0.3861	0.4574	0.0043	0.0076
0.6966	6	0.1489	0.0013	6	0.7045	0.0018	0.6941	0.7022	0.0025	0.0023
0.9920	6	0.2610	0.0025	6	0.7980	0.0015	0.9978	0.7987	-0.0058	-0.0007
1.294	6	0.4009	0.0022	6	0.8526	0.0011	1.3096	0.8549	-0.0156	-0.0023
1.5932	6	0.5716	0.0023	6	0.8924	0.0009	1.6134	0.8942	-0.0202	-0.0018
1.8437	6	0.7267	0.0018	6	0.9227	0.0006	1.8558	0.9233	-0.0121	-0.0006
2.0045	6	0.8247	0.0013	6	0.9436	0.0005	2.0143	0.9439	-0.0098	-0.0003
2.1571	6	0.9093	0.0007	6	0.9664	0.0003	2.1654	0.9665	-0.0083	-0.0001
283.36 K										
0.7483	11	0.1104	0.0009	10	0.5880	0.0024	0.7407	0.5796	0.0076	0.0084
1.1312	10	0.2227	0.0018	10	0.7304	0.0020	1.1270	0.7279	0.0042	0.0025
1.4943	9	0.3482	0.0021	9	0.8014	0.0015	1.4989	0.8021	-0.0046	-0.0007
1.8646	11	0.5016	0.0022	9	0.8514	0.0012	1.8814	0.8529	-0.0168	-0.0015
2.2451	9	0.6754	0.0020	15	0.8940	0.0011	2.2592	0.8944	-0.0141	-0.0004
2.7069	7	0.8780	0.0010	6	0.9490	0.0006	2.7189	0.9488	-1.8409	0.0002
298.04 K										
0.9368	9	0.0904	0.0008	6	0.4610	0.0022	0.9252	0.4545	0.0116	0.0066
1.5016	8	0.2206	0.0015	9	0.6648	0.0022	1.4863	0.6606	0.0153	0.0042
2.0673	8	0.3751	0.0021	8	0.7590	0.0019	2.0671	0.7618	0.0002	-0.0027
2.6204	8	0.5464	0.0026	8	0.8235	0.0013	2.6253	0.8243	-0.0049	-0.0008
3.1639	6	0.7198	0.0028	6	0.8762	0.0012	3.1577	0.8751	0.0062	0.0010
3.7306	8	0.8873	0.0013	8	0.9370	0.0006	3.7342	0.9361	-0.0036	0.0009
308.04 K										

1.1942	6	0.0968	0.0010	10	0.4235	0.0022	1.1880	0.4244	0.0062	-0.0009
1.8909	6	0.2402	0.0020	6	0.6325	0.0021	1.8928	0.6342	-0.0019	-0.0017
2.4105	6	0.3620	0.0038	6	0.7151	0.0019	2.4273	0.7164	-0.0168	-0.0013
2.9802	6	0.5065	0.0023	6	0.7775	0.0016	2.9989	0.7770	-0.0187	0.0005
3.3693	6	0.6090	0.0024	6	0.8124	0.0013	3.3813	0.8106	-0.0120	0.0018
3.6941	6	0.6926	0.0020	6	0.8400	0.0012	3.6949	0.8371	-0.0008	0.0029
3.8753	6	0.7377	0.0020	6	0.8550	0.0012	3.8701	0.8521	0.0052	0.0029
3.9801	6	0.7634	0.0021	6	0.8641	0.0011	3.9732	0.8611	0.0069	0.0030
4.0936	6	0.7896	0.0016	6	0.8740	0.0011	4.0815	0.8709	0.0121	0.0031
4.1923	6	0.8123	0.0014	6	0.8833	0.0010	4.1782	0.8799	0.0141	0.0034
4.2905	8	0.8342	0.0013	6	0.8927	0.0009	4.2745	0.8891	0.0160	0.0036
4.3973	6	0.8579	0.0015	6	0.9035	0.0009	4.3823	0.8999	0.0150	0.0036
4.4872	6	0.8771	0.0011	6	0.9128	0.0008	4.4727	0.9093	0.0145	0.0035
4.5974	6	0.8989	0.0012	6	0.9242	0.0006	4.5787	0.9209	0.0187	0.0033
4.6925	6	0.9168	0.0009	6	0.9331	0.0007	4.6685	0.9311	0.0240	0.0020
4.7444	6	0.9282	0.0008	6	0.9401	0.0006	4.7266	0.9380	0.0178	0.0021
					323.18 K					
1.5920	6	0.0932	0.0009	6	0.3493	0.0023	1.5866	0.3450	0.0054	0.0044
2.3031	6	0.2159	0.0023	6	0.5373	0.0023	2.3092	0.5363	-0.0061	0.0010
2.8707	7	0.3178	0.0030	6	0.6247	0.0021	2.8655	0.6205	0.0052	0.0042
3.4876	6	0.4402	0.0028	6	0.6893	0.0020	3.4867	0.6851	0.0009	0.0042
3.9825	6	0.5400	0.0022	6	0.7267	0.0020	3.9643	0.7228	0.0182	0.0039
4.0939	6	0.5622	0.0024	6	0.7346	0.0017	4.0683	0.7301	0.0256	0.0045
4.1929	6	0.5824	0.0022	6	0.7393	0.0019	4.1624	0.7364	0.0305	0.0029
4.2968	6	0.6022	0.0021	6	0.7461	0.0019	4.2542	0.7424	0.0426	0.0037
4.3923	6	0.6222	0.0022	6	0.7523	0.0016	4.3466	0.7481	0.0457	0.0042
4.4933	6	0.6435	0.0020	25	0.7584	0.0017	4.4445	0.7539	0.0488	0.0045
4.5919	7	0.6642	0.0020	6	0.7630	0.0016	4.5391	0.7592	0.0528	0.0039

4.6906	6	0.6843	0.0020	6	0.7669	0.0016	4.6299	0.7638	0.0607	0.0031
4.7916	6	0.7062	0.0019	6	0.7689	0.0016	4.7265	0.7679	0.0651	0.0010
4.8921	6	0.7328	0.0018	6	0.7630	0.0020	4.8840	0.7328	0.0081	0.0302
					347.52 K					
2.1602	6	0.0486	0.0005	6	0.1497	0.0016	2.1722	0.1489	-0.0120	0.0008
2.4859	6	0.0919	0.0010	6	0.2424	0.0017	2.5076	0.2428	-0.0217	-0.0004
3.0070	6	0.1617	0.0012	6	0.3520	0.0020	3.0291	0.3482	-0.0221	0.0038
3.4895	6	0.2291	0.0018	6	0.4214	0.0027	3.5078	0.4168	-0.0183	0.0046
4.0030	6	0.3035	0.0019	6	0.4773	0.0022	4.0034	0.4681	-0.0004	0.0092
4.1732	6	0.3309	0.0019	6	0.4915	0.0022	4.1756	0.4819	-0.0024	0.0096
4.3157	6	0.3529	0.0020	6	0.4994	0.0024	4.3090	0.4912	0.0067	0.0083
4.4097	6	0.3683	0.0021	8	0.5043	0.0023	4.3994	0.4966	0.0103	0.0077
4.5135	6	0.3849	0.0022	6	0.5065	0.0046	4.4936	0.5016	0.0199	0.0049
4.5818	6	0.3979	0.0022	12	0.5092	0.0029	4.5647	0.5047	0.0171	0.0045
4.7294	6	0.4254	0.0021	7	0.5105	0.0023	4.7053	0.5088	0.0241	0.0017
4.7882	6	0.4395	0.0021	6	0.5061	0.0022	4.7705	0.5092	0.0177	-0.0031
4.8303	6	0.4534	0.0022	6	0.4973	0.0025	4.8283	0.5081	0.0020	-0.0108

[a] Standard uncertainties *u* are reported. u(T) = 0.017K and u(P) = 0.23 kPa.

[b] n_x, n_y: number of taken samples. $\triangle P=P_{cal} - P_{exp}$. $\triangle y=y_{cal} - y_{exp}$,

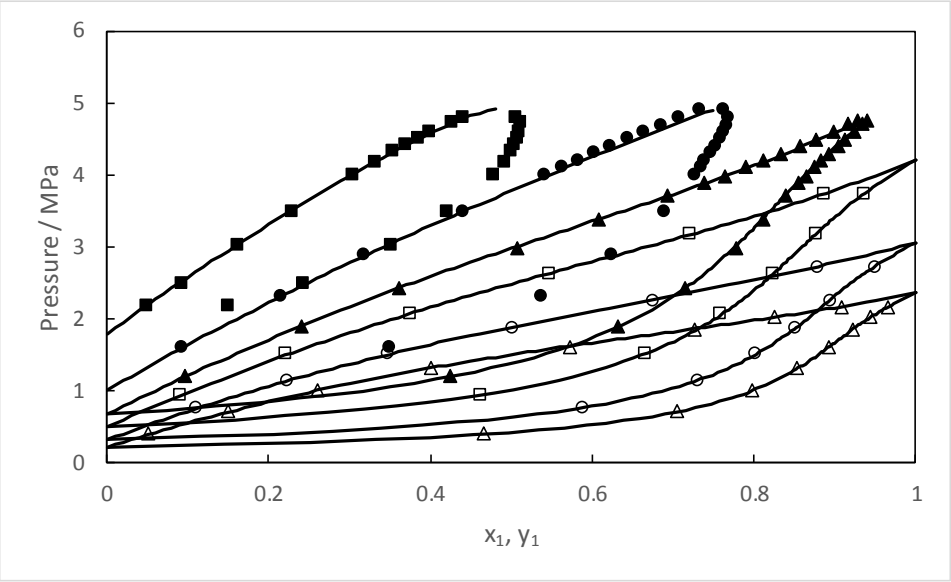


Figure 2. Phase diagrams for the (ethane (1) + R1234ze(E)(2)) system. (●) 272.27 K; (■) 283.36 K; (▲) 298.04 K; (◆) 308.04 K; (x) 323.18 K; (+) 347.52 K. Solid lines: PR-EoS.

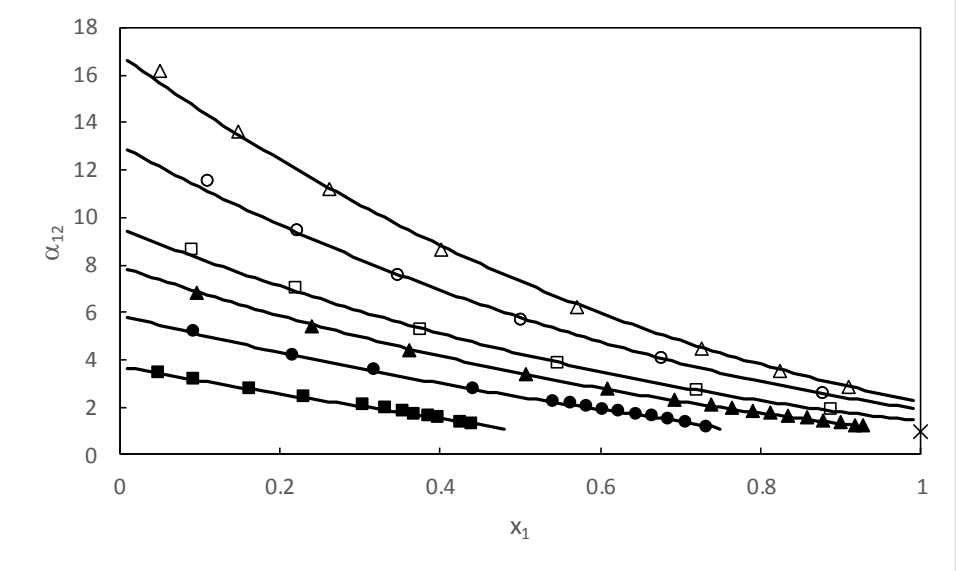


Figure 3. Relative volatility as function of molar fraction for the (ethane (1) + R1234ze(E)(2)) system. (●) 272.27 K; (■) 283.36 K; (▲) 298.04 K; (◆) 308.04 K; (x) 323.18 K; (+) 347.52 K. Solid lines: calculated using PR-EoS.

The temperature dependent parameter k_{ij} (see figure 4) were adjusted on experimental data (and listed in Table 4) considering the objective function given by Eq. (3).

$$F = \frac{100}{N}\left[\sum_1^N \left(\frac{P_{exp}-P_{cal}}{P_{exp}}\right)^2 + \sum_1^N \left(\frac{y_{exp}-y_{cal}}{y_{exp}}\right)^2\right] \quad (3)$$

Table 4. Adjusted k_{ij} and objective function values.

T/K	k_{ij}	F (Eq. 4)
272.27	0.1044	0.013
283.36	0.1028	0.007
298.04	0.1017	0.011
308.04	0.1051	0.057
323.18	0.1058	0.069
347.52	0.1177	0.002
$T>T_{c,ethane}$	0.1052	0.053

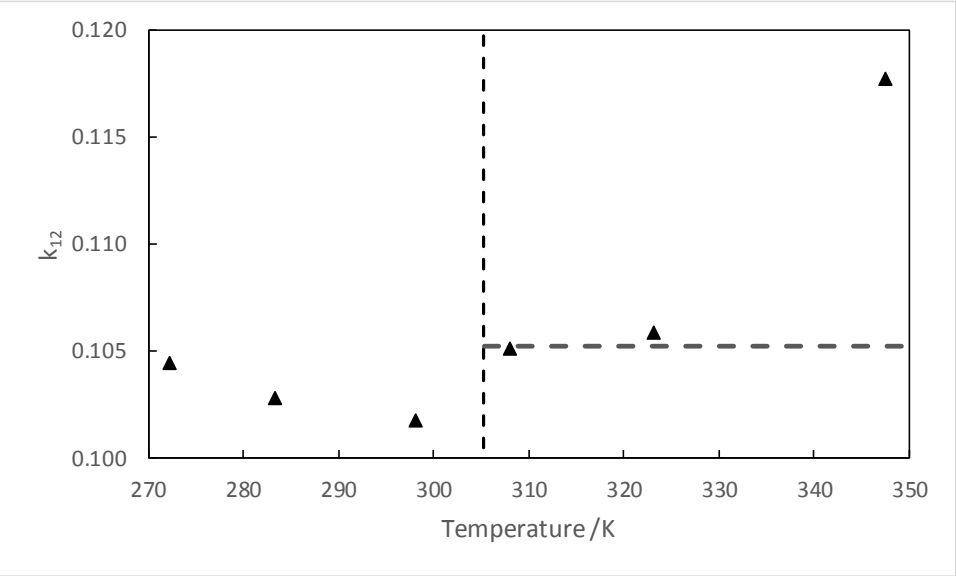


Figure 4. Plot of adjusted temperature dependent parameter k_{ij} versus temperature. Vertical dashed line: critical temperature of ethane. Horizontal dashed line: binary interaction parameter adjusted using all the data for $T>T_{c,ethane}$.

In order to quantify the goodness of the fit of PR-EoS to the experimental data, the mean relative absolute percentage deviations (MRD) and the bias on bubble pressures and vapour mole fractions were calculated. They are all listed in Table 5. The expressions of MRD and bias are defined by Eqs. (4) and (5) as follows:

$$\mathrm{MRD(U)} = \frac{100}{N}\sum\left[\left|\frac{U_{cal}-U_{exp}}{U_{exp}}\right|\right] \quad (4)$$

$$\mathrm{bias(U)} = \frac{100}{N}\sum\left[\frac{U_{cal}-U_{exp}}{U_{exp}}\right] \quad (5)$$

where U represents P or y_1 and N is the number of data points.

Table 5. MRD and Bias values for bubble pressures and vapour phase compositions.

T/K	Bias (P)/%	MRD(P)/%	Bias(y)/%	MRD(y)/%
272.27	-0.60	-0.05	0.71	0.14
283.36	-0.38	0.01	0.53	0.13
298.04	0.19	0.08	0.30	0.26
308.04	0.10	0.24	0.34	0.30
323.18	0.66	0.78	0.69	0.78
347.52	-0.05	0.69	0.38	1.14
$T > T_{c,ethane}$	0.69	1.13	0.78	1.23

Table 6. Critical pressures and critical compositions of the mixture at temperatures higher than the critical temperature of pure ethane.

T/K	P_c/MPa	$x_{c,ethane}$	$\mu * 10^3$	$\lambda_1 * 10^4$	$\lambda_2 * 10^4$
308.04	4.833	0.948	6	996	-1557
323.18	4.912	0.751	102	910	-1201
347.52	4.848	0.478	163	241	-938

For the three isotherms higher than 305.32 K and three isotherms lower than 305.32 K, two curves are correlated, respectively. Based on the extended asymptotic behaviour laws (Eqs. 1 and 2), the critical pressures and critical compositions were also calculated and their values were shown in Table 6. The estimated critical points based on experimental data are presented in Figure 5. Also, using our model and the adjusted parameter of Table 4 for $T>T_{c,ethane}$ (given the fact that the values of the BIP are very close, we prefer to use a non-temperature dependent BIP) and based on Heidemann and Khalil method[20], the critical line is calculated (Table 7) and presented in Figures 5 and 6. Based on these results, we can conclude that the model could represent the complete phase diagram of this system with a good accuracy, either at subcritical or supercritical conditions.

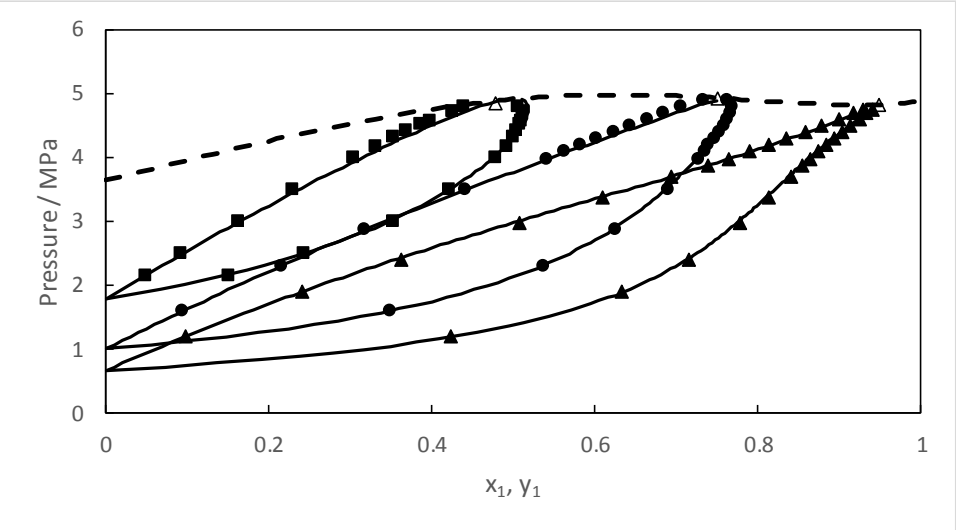
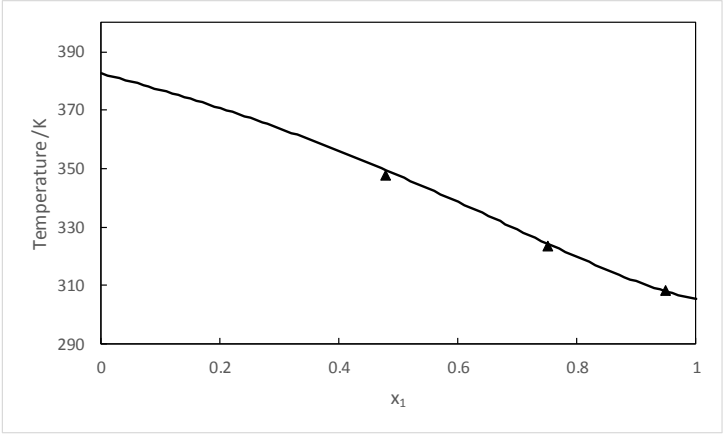


Figure 5. Phase diagrams for the (ethane (1) + R1234ze(E)(2)) system. (◆) 308.04 K; (x) 323.18 K; (+) 347.52 K. (Δ) Estimated critical points using experimental data. Solid lines: PR-EoS, Dashed line: critical line.

Table 7. Values of calculated critical temperature and pressure with temperature independent BIP.

x_1	T/K	P/MPa
0.00	382.52	3.64
0.10	376.93	3.96
0.20	370.71	4.26
0.30	363.78	4.53
0.40	356.09	4.76
0.50	347.65	4.91
0.60	338.57	4.98
0.70	329.11	4.96
0.80	319.76	4.89
0.90	311.37	4.82
1.00	305.32	4.87



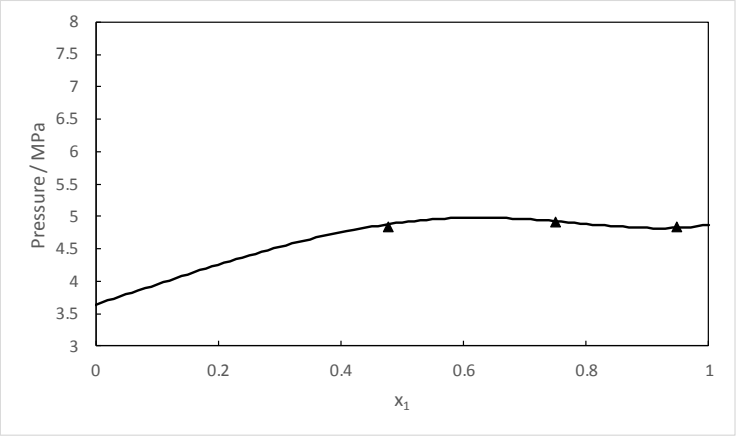


Figure 6. Critical temperature and pressure as a function of composition for the binary system ethane (1) + R1234ze(E)(2).

5. Conclusion

In this paper, new VLE data of the (ethane + R1234ze(E)) binary system are presented. This system can be classified as a type I according to van Konynenburg and Scott classification[21]. Experimental measurements were performed at six isotherms using a "static-analytic" apparatus with phase sampling and gas chromatography analysis, with resulting uncertainties of 0.03K for temperature, 0.4 kPa for pressure and 0.0043 for vapour and liquid mole fractions.

The experimental data were correlated using PR-EoS combined with the classical van der Waals mixing rules. For the three isotherms higher than the critical temperature of pure ethane, the critical pressures and critical compositions were calculated based on the extended asymptotic behaviour laws.

The results obtained indicate that the selected model reproduce accurately the experimental VLE data of (R1234ze(E) + ethane) mixture, with a maximal deviation of 1.13% for bubble pressure and 1.23% for vapour molar fraction.

These new data will be very valuable in selecting new working fluids for heat pump and refrigeration applications. They will be also very useful to extend the plethora of available

data and can be used to develop predictive models and fit their parameters.

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