

VAPOUR PRESSURE OF ETHANOL AND 1-PROPANOL BINARY MIXTURES

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The vapour pressure of binary mixtures containing ethanol and 1-propanol were investigated at temperatures ranging from 274.15 to 443.15 K using two different setups with static methods. The measured values were fitted to a Clausius-Clapeyron type relationship. The heat of evaporation of mixtures was determined from the vapour-liquid equilibria data.

Keywords: vapour pressure, ethanol, 1-propanol, static method, pressure transmitters, Clausius-Clapeyron equation

1. Introduction

Investigation of thermodynamic properties of pure liquids and their mixtures is important in various fields of science, chemical engineering, economy and industry. Aliphatic alcohols are commonly applied in chemical, biological, and medical uses as solvents for fats, oils, resins, paints, and nitrocellulose with regard to the manufacture of goods from perfumes to brake fluids [1]. In addition, the studied solutions of ethanol (C_2H_5OH) and 1-propanol (C_3H_7OH) are also used as heat transfer fluids in heat reservoirs, solar heating systems, oxygenates in fuels, and cryogenic power generation systems [2]. For the design and modelling of such applications, the determination of flow in pipes, heat transfer, and mass transfer operations requires the knowledge of thermophysical properties. Density, vapour pressure, speed of sound, viscosity, and heat capacity often need to be defined for these purposes.

This work is a continuation of our previous publications in the field of thermophysical properties of alcohol and their solutions [3–6]. Hereby, the vapour pressure of binary solutions of $(1-x) C_2H_5OH + x C_3H_7OH$ were investigated. The vapour pressure data of binary solutions of ethanol and 1-propanol at different temperatures and concentrations were determined. After the analysis of the literature using “ThermoLit” from NIST, we concluded that only a few vapour pressure values for these systems have been reported to date [7–11].

Early studies by Parks and Schwenk [7] reported the vapour pressure of a $(1-x) C_2H_5OH + x C_3H_7OH$ mixture at 298.15 K using glass apparatus and the differential method. A good commercial grade ethanol ($w = 99.9\%$) and “refined” commercial 1-propanol ($w = 99.34\%$) were used during the preparation of solutions. Later, Udovenko and Frid [8] investigated the vapour pressure of the same mixture, but within a higher temperature range (323.15 to 353.15 K) using the dynamic method. The vapour liquid equilibria (VLE) of these systems were analysed using a refractometer. The activity coefficients γ of both pure components were calculated.

A series of studies in the early 90s included the work of Zielkiewicz [9], who studied the vapour pressure at 313.15 K using the static method. Dried ethanol and 1-propanol were used during the preparation of solutions. The temperature and pressure were controlled within ± 0.001 K and ± 0.004 kPa, respectively. Binary samples were prepared by weighing within an uncertainty of ± 0.0005 mole fractions. Solution preparations were carried out using the dry nitrogen process. Furthermore, Pradhan *et al.* [10] investigated the vapour pressures of ethanol and 1-propanol solutions at 303.15 K using the static method. For the fitting of obtained values a modified NRTL equation was used.

Quite recently, Cristino *et al.* [11] carried out high temperature VLE measurements for the system of ethanol and 1-propanol solutions within a temperature range of 403.2 to 423.2 K using a flow apparatus. Alcohols used during the preparation of solutions had a confirmed purity greater than 99.9 weight percent. The pressure was controlled using two pressure transducers within ranges of 0 – 0.4 (uncertainty of ± 0.0002 MPa) and 0–1.7 MPa (uncertainty of ± 0.0009 MPa). The

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Table 1. Summary of the vapour pressure P literature investigations of a $(1-x)$ $C_2H_5OH + x C_3H_7OH$ mixture.

Reference	Method	Properties	Temperature (T in K)	Concentration (x mole fraction)	Uncertainty ΔP	Fitted density equation	Purity	Source
Parks [7] 1924	GA	$P, T, \Delta H$	298.15	0.0000 to 1.0000			99.9% (Et) 99.34% (Pr)	CS
Udovenko [8] 1948	DM	P, T, γ	323.15 – 353.15	0.0000 to 1.0000		CC	ARG (Et) ARG (Pr)	R
Zielkiewicz [9] 1993	SM	P, T	313.15	0.0436 to 0.9291	± 0.004 kPa			POCh
Pradhan [10] 1993	SM	P, T	303.15	0.0306 to 0.9700	± 0.001 kPa		99.9% (Et) 99.6% (Pr)	AC
Cristino [11] 2015	FA	P, T, VLE	403.20 – 423.20	0.0017 to 0.9993	± 0.0002 – ± 0.0009 MPa	SAFT-VR	99.9% (Et) 99.9% (Pr)	P (Et) FS (Pr)

GA, glass apparatus; ΔH , heat of mixing; P , vapour pressure; T , temperature; x , mole fraction; Et, Ethanol; Pr, 1-Propanol; CS, commercial sample; DM, dynamic method; γ , activity coefficient; CC, Clapeyron-Clausius equation; ARG, analytical reagent grade; R, Reachim, USSR; SM, static method; POCh, Avantor Performance Materials Poland S.A.; AC, Aldrich Chemical; FA, flow apparatus; SAFT-VR, statistical associating fluid theory; VLE, vapour-liquid equilibrium; P, Panreac; FS, Fisher Scientific.

temperature was measured using a platinum resistance thermometer with an uncertainty of ± 0.1 K. The statistical associating fluid theory for potentials of variable range (SAFT-VR) was used to model the systems and found to accurately reproduce the experimental data. Using this analytical method the uncertainty of solution preparation was ± 0.0001 mole fractions.

The outcome of a literature survey summarised in Table 1 is that only small temperature, pressure, and concentration intervals were investigated to date in addition to older literature examples decades ago that may have used out-dated measurement techniques.

In this work, the vapour pressures of binary $(1-x)$ $C_2H_5OH + x C_3H_7OH$ solutions were investigated using two highly accurate, fully automatic static experimental setups and ultrapure Merck quality chemicals.

2. Experimental

2.1. Samples and Measurements

Ultra-pure ethanol EMPLURA® ($w = 99.995\%$, CAS No. 71-36-3, Art. Nr. 8.22262.2500) and 1-propanol Analyse EMSURE® ACS, Reag. Ph Eur ($w = 99.995\%$, CAS No. 71-23-8, Art. Nr. 1009971000) were purchased from Merck Schuchardt OHG, Germany. The samples were used without further purification. They were carefully degassed in glass flasks with special vacuum leak-proof valves before measurements were taken. The water content is determined by Karl Fischer titration and was determined to be less than a mass fraction of 20 ppm.

2.2. Experimental Procedure

The vapour pressure measurements of binary solutions of $(1-x)$ $C_2H_5OH + x C_3H_7OH$ were measured using two high-accuracy static experimental setups [12–14]. The glass cells were used for vapour pressure measurements lower than ambient pressure at temperatures from

274.15 to 323.15 K. The metal cell was used for the higher temperature range of 323.15–433.15 K using the static method [12–14]. The glass cell method consists of absolute and differential parts (if the vapour pressure is smaller than the uncertainty of the absolute cell, 30 Pa). The vapour pressure of the solution was always higher than the uncertainty of measurements between 274.15 and 323.15 K. The measurements within this temperature range were carried out only using the absolute cell of installation. The internal volume of the glass cell in absolute measurements is approximately 78.56 cm³, and the volume of steel tube cells is 1 cm³. The glass cell static method consists of a bolted-top cell in a water-bath kept at constant temperature (± 0.01 K) using a thermostat.

The vapour pressure was measured using a calibrated high accuracy sensor head [Type 615A connected to the signal conditioner Type 670A, MKS Baratron, USA] attached to the top of the cell of various Keller pressure transmitters: maximum pressure of 300,000 Pa with an uncertainty of $\Delta P = \pm(400$ to 1,500) Pa, maximum pressure of 1,000,000 Pa with an uncertainty of $\Delta P = \pm(1,000$ to 5,000) Pa and maximum pressure of 1,600,000 Pa with an uncertainty of $\Delta P = \pm(2,000$ to 8,000) Pa. The experimental uncertainty of the pressure in the absolute vapour pressure measurement using the glass cell is ± 10 –30 Pa.

The internal volume of the measurement cell is approximately 140 cm³. Temperatures were measured using two different platinum resistance thermometers, PT-100. The second platinum resistance thermometer, PT-100, transfers the measured temperature in the computer via an Omega PT-104A Channel RTD Input Data Acquisition Module (Omega Engineering, Inc., USA) for the measuring of temperature, with an accuracy of ± 0.001 K. Experiments were carried out starting from a low temperature (333.15 K) to a high temperature (433.15 K) at 10 K intervals.

Before the experiments, the measurement cells were washed with water, methanol and acetone and then all residual fluids were removed. This procedure requires approximately 2 to 3 h or more to reach the

Table 2. Experimental mole fraction x of 1-propanol, and vapour pressure P (in Pa) of a solution of $(1-x)\text{C}_2\text{H}_5\text{OH} + x\text{C}_3\text{H}_7\text{OH}$ ^a

Temperature (K)	mole fraction of 1-propanol (x)							
	0.0000 ^b	0.0989	0.1918	0.4034	0.5935	0.7971	0.9038	1.0000 ^c
274.15	1684	1490	1374	1116	897	672	530	515
278.15	2248	1980	1843	1498	1219	902	739	697
283.15	3155	2810	2620	2149	1761	1330	1102	1008
293.15	5842	5390	5023	4186	3442	2670	2260	2034
303.15	10458	9762	9180	7780	6521	5150	4402	3854
313.15	18054	16872	15880	13558	11502	9260	8030	7048
323.15	29356	27918	26430	22909	19560	15890	13873	12273
333.15	46796	44590	42200	36700	31777	26000	22992	20472
343.15	71902	68812	65321	57200	49784	41400	36903	32867
353.15	108174	103196	98000	86256	75504	63400	56954	50997
363.15	157911	150535	143157	126500	111128	94202	85257	76746
373.15	224798	214272	203797	180800	159521	136296	123958	113402
383.15	313786	298327	284000	252345	223518	192294	176009	161109
393.15	429264	407124	387678	345105	306612	265259	244002	223982
403.15	576481	545340	519543	462803	412430	358865	331402	305477
413.15	759512	718454	684376	610504	545107	476594	441754	408702
423.15	982342	932045	887923	792004	708954	622271	578714	539077
433.15	1254038	1191945	1135123	1012845	907984	800473	746309	702376
443.15	1582042	1505202	1432927	1278187	1147706	1015139	949123	893968

^a Standard uncertainties u are $u(T) = 0.01$ K and $u(x) = 0.0001$ mole fractions and the combined expanded uncertainties U_c are $U_c(P) = 30$ Pa for $P < 0.1$ MPa, $U_c(P) = 1500$ Pa for $P < 3$ MPa, and $U_c(P) = 8000$ Pa for $P < 16$ MPa (level of confidence = 0.95); ^b The vapour pressure values of ethanol were taken from Ref. [15]; ^c The vapour pressure values of 1-propanol were taken from Ref. [16].

desired minimal pressure (20–30 Pa). Equilibration of the cells is a rapid process and a constant pressure in the stationary regime is reached within 15 minutes. Equilibrium pressure readings are performed in triplicate approximately 10 to 20 min intervals.

Specific quantities of ethanol and 1-propanol were evacuated, degassed in two separate flasks and connected using an adapter [12]. Ethanol flowed into a flask containing 1-propanol and the concentration of the solution was determined using the weight of the flask containing the solution on an electronic scale (Sartorius ED224S, Germany) with an uncertainty of 0.0001 g. A quantity of the solution was injected into the equilibrium cells up to approximately 50% of their volume.

The vapour pressures of the water, methanol, acetone, toluene, 1-butanol, *etc.* were measured as reference substances for testing both setups [12–14]. The experimental vapour pressure results were assessed to be reliable to within an average uncertainty of $\pm 0.05\%$ according to test measurements.

3. Results and Discussion

The measured experimental vapour pressures for an ethanol/1-propanol mixture within the temperature range of 274.15 to 433.15 K are listed in Table 2, and are also shown in Fig. 1. The vapour pressures of pure alcohols were taken from Refs. [15–16].

The experimental vapour pressure results, P in Pa of investigated solutions were fit to the Antoine equation:

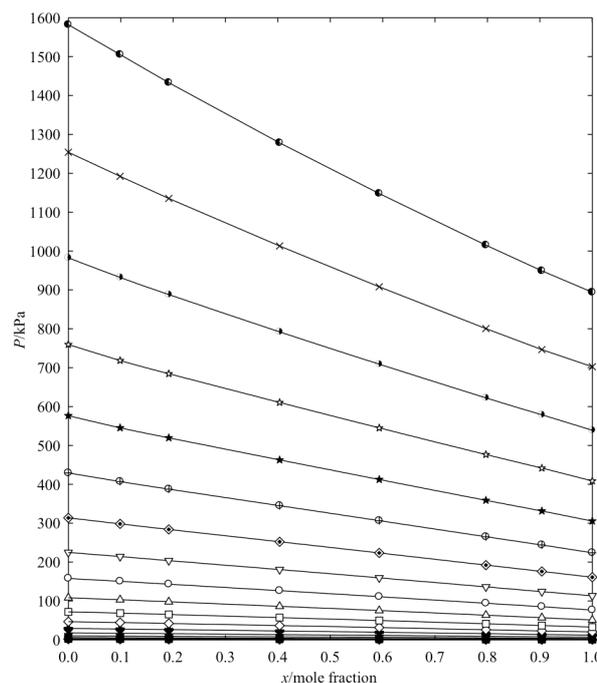


Figure 1. Plot of vapour pressure P (in kPa) of a $(1-x)\text{C}_2\text{H}_5\text{OH} + x\text{C}_3\text{H}_7\text{OH}$ solution mixture as a function of 1-propanol mole fraction x . \blacklozenge , 274.15 K; \blacksquare , 278.15 K; \blacktriangle , 283.15 K; \bullet , 293.15 K; \blacktriangledown , 303.15 K; \times , 313.15 K; \oplus , 323.15 K; \diamond , 333.15 K; \square , 343.15 K; \triangle , 353.15 K; \circ , 363.15 K; ∇ , 373.15 K; \diamond , 383.15 K; \oplus , 393.15 K; \star , 403.15 K; \star , 413.15 K; \bullet , 423.15 K; \times , 433.15 K; \circ , 443.15 K; lines fit to Eqs.(3) and (4).

$$\ln(P) = A^A - B^A / (T/K + C^A) \quad (1)$$

Table 3. Antoine parameters A^A , B^A , C^A and percent deviations ($\Delta P/P$ in %) as a function of 1-propanol mole fraction.

mole fraction	A^A	B^A	C^A	$\Delta P/P$
0.0000 ^a	23.1773	3461.23	-54.3818	0.6234
0.0989	22.8524	3275.81	-63.4603	0.0742
0.1918	22.7353	3228.33	-65.9886	0.0689
0.4034	22.4745	3118.11	-72.4808	0.1652
0.5935	22.3425	3077.68	-76.1926	0.2204
0.7971	22.2692	3064.56	-79.8646	0.3263
0.9038	22.1582	3009.02	-84.6711	0.1983
1.0000 ^b	22.7515	3373.18	-70.0769	0.8270

^a from Ref. [15]; ^b from Ref. [16].

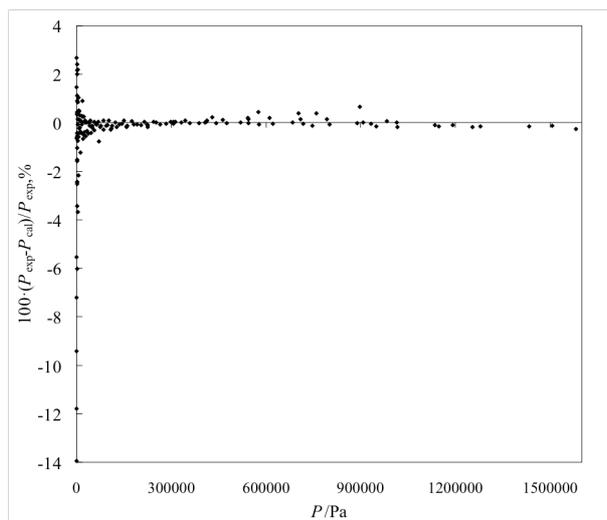


Figure 2. Deviation of experimental P_{exp} and calculated P_{cal} vapour pressure values versus pressure P using Eqs.(3) and (4) at various temperatures and mole fractions.

The fitted constants A^A , B^A , and C^A for the investigated solutions are summarised in Table 3 with the standard mean deviation defined as follows:

$$\delta P/P = 100/n \cdot \sum_{i=1}^n [(P_{\text{exp.}} - P_{\text{cal.}})/P_{\text{exp.}}] \quad (2)$$

From Table 3, it can be seen that coefficients A^A , B^A , and C^A exhibit non-trivial dependence from the mole fraction of 1-propanol. Fitting of these coefficients was a challenging task. Thus, we also used a Clausius–Clapeyron-type equation to obtain the vapour pressure results of the investigated solutions from mole fractions of 1-propanol:

$$\ln p = A^{\text{CC}} + \frac{B^{\text{CC}}}{T} + C^{\text{CC}} \ln T + D^{\text{CC}} T, \quad (3)$$

where P is vapour pressure in Pa; T is the temperature in K; and A^{CC} , B^{CC} , C^{CC} , and D^{CC} are the coefficients of the equation, depending on the mole fraction of the solvent as follows:

Table 4. Clausius - Clapeyron equation fitting parameters a_i , b_i , c_i , and d_i from Eqs.(3) and (4).

a_i	b_i	c_i	d_i
$a_0 = 103.156$	$b_0 = -7994.80$	$c_0 = -12.3406$	$d_0 = 0.0098481$
$a_1 = 251.788$	$b_1 = -8366.78$	$c_1 = -42.1398$	$d_1 = 0.0527419$
$a_2 = 222.344$	$b_2 = -7727.52$	$c_2 = -36.8168$	$d_2 = 0.0438405$
$a_3 = -446.740$	$b_3 = 14403.70$	$c_3 = 74.8888$	$d_3 = -0.0951465$

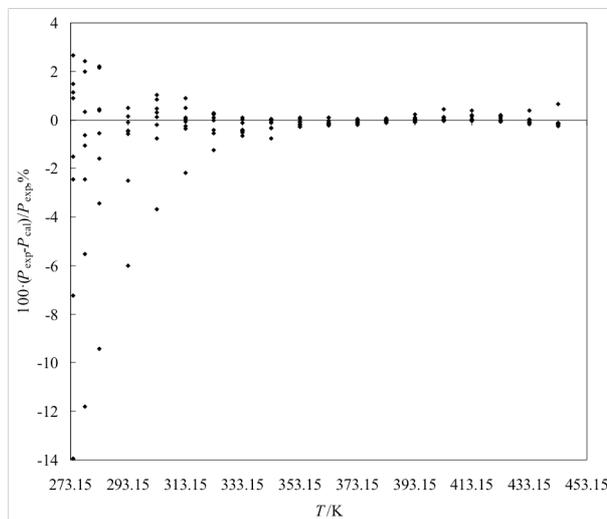


Figure 3. Deviation of experimental P_{exp} and calculated P_{cal} vapour pressure values versus temperature T using Eqs.(3) and (4) at various pressures P and mole fractions.

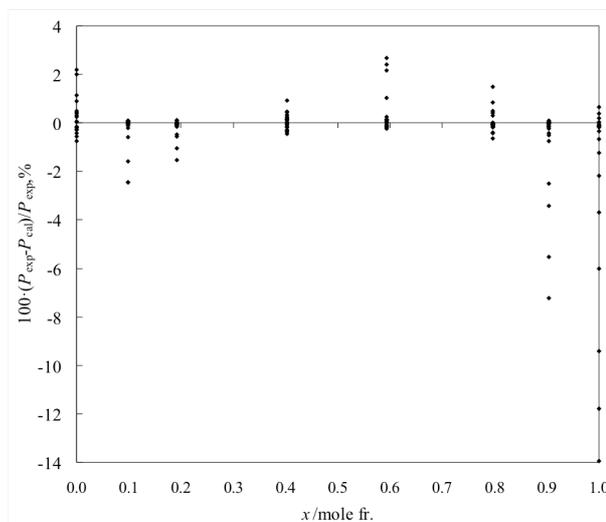


Figure 4. Deviation of experimental P_{exp} and calculated P_{cal} vapour pressure values versus mole fraction x using Eqs.(3) and (4) at various pressures P and temperatures T .

$$A^{\text{CC}} = \sum_{i=0}^3 a_i x^i \quad B^{\text{CC}} = \sum_{i=0}^3 b_i x^i \quad C^{\text{CC}} = \sum_{i=0}^3 c_i x^i \quad D^{\text{CC}} = \sum_{i=0}^3 d_i x^i \quad (4)$$

The coefficients a_i , b_i , c_i , and d_i for the investigated ethanol/1-propanol mixtures are tabulated in Table 4. The uncertainty of fitting was approximately $u_r(\Delta P/P) = 0.7678$. The plots of deviation of experimental P_{exp} and calculated P_{cal} vapour pressure values as a function of

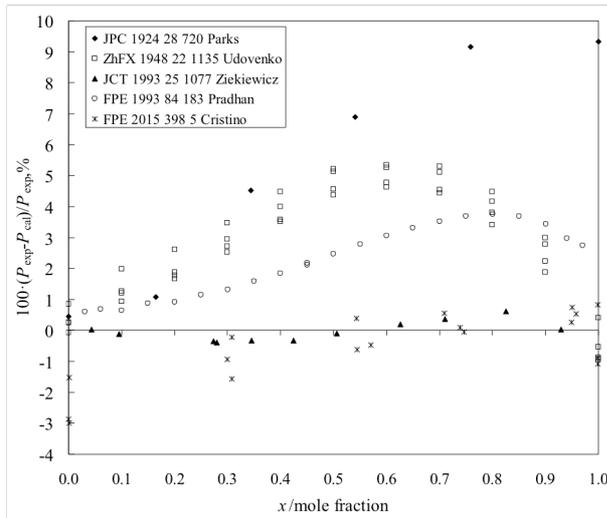


Figure 5. Deviation of experimental P_{exp} and literature P_{lit} vapour pressure values for the ethanol/1-propanol mixture versus 1-propanol mole fraction using Eqs.(3) and (4) at various pressures P and temperatures T .

pressure, temperature, and mole fraction using Eqs.(3) and (4) are shown in Figs.2-4, respectively.

The enthalpies of vaporisation, ΔH_{vap} in J mol^{-1} , for the $(1-x)\text{C}_2\text{H}_5\text{OH} + x\text{C}_3\text{H}_7\text{OH}$ mixture at the four middle temperatures (293.15, 333.15, 373.15, and 423.15 within temperature ranges of 274.15–313.15 K, 313.15–353.15 K, 353.15–393.15 K, and 373.15–443.15 K, respectively) were defined using Eq.(5) from Ref. [12]:

$$\frac{d \ln P}{d\left(\frac{1}{T}\right)} = -\frac{\Delta H_{\text{v}}}{R} \quad (5)$$

If we plot $\ln(P)$ as a function of $1/T$, we can define ΔH_{v} from the gradient of the line:

$$\Delta H_{\text{v}} = -R \cdot \frac{d \ln P}{d\left(\frac{1}{T}\right)} \quad (6)$$

After the integration of Eq.(6) we can find

$$\ln p = \left(-\frac{\Delta H_{\text{v}}}{R}\right)\left(\frac{1}{T}\right) + \text{intercept} \quad (7)$$

$$\Delta H_{\text{v}} = RT(\text{intercept} - \ln P) \quad (8)$$

The calculated enthalpy of vaporisations ΔH_{v} in J mol^{-1} for the $(1-x)\text{C}_2\text{H}_5\text{OH} + x\text{C}_3\text{H}_7\text{OH}$ mixture within the temperature range of 274.15–443.15 K are listed in Table 5 and compared to the available literature results [7–11] shown in Fig.5.

When the measured values by Parks [7] at $T = 298.15$ K are compared to our values, we obtain $\Delta P = \pm 242$ Pa or $\Delta P/P = \pm 5.24\%$ deviations. The maximum deviation is $\Delta P = 394$ Pa at $x = 0.759$ mole fractions of

Table 5. Enthalpy of vaporisation, ΔH_{v} in kJ mol^{-1} for a $(1-x)\text{C}_2\text{H}_5\text{OH} + x\text{C}_3\text{H}_7\text{OH}$ mixture at various temperatures.

x	293.15 K	333.15 K	373.15 K	423.15 K
0.0000	43.245	41.210	39.579	37.457
0.1574	43.905	41.300	39.542	37.576
0.2876	44.499	41.416	39.425	37.553
0.5351	45.768	41.989	39.707	37.718
0.7130	46.768	42.811	40.157	38.002
0.8699	47.809	43.815	40.965	38.613
0.9411	47.887	44.441	41.546	39.125
1.0000	47.908	45.052	42.330	39.963

1-propanol. The Parks' values [7] are higher than our results and the vapour pressures of ethanol exhibit small deviations compared to ours and all other literature values presented in Ref. [15]. The vapour pressure of 1-propanol published in Ref. [7] exhibits a large deviation from ours and all other literature values presented in Ref. [16]. We hypothesise that the vapour pressure values of 1-propanol with high deviation from the literature were used during the analysis of concentration dependence in Ref. [7].

The 44 data points of Udovenko and Frid [8] measured within the range of 323.15 – 353.15 K are mostly higher than our values. The average deviations of both sources are $\Delta P = \pm 242$ Pa and $\Delta P/P = \pm 5.24\%$ with maximum deviations of $\Delta P = 2952$ Pa at $T = 343.15$ K and $x = 0.5$ mole fractions of 1-propanol.

The 11 data points of Zielkiewicz [9] at $T = 313.15$ K exhibit small deviations from our results with $\Delta P = \pm 33$ Pa and $\Delta P/P = \pm 0.2631\%$ mean deviation. The maximum obtained deviation in $\Delta P = -59$ Pa at $x = 0.2793$ mole fractions of 1-propanol.

The next 22 data points of Pradhan *et al.* [10] are mostly higher than our values. The average mean deviation of this comparison is $\Delta P = \pm 143$ Pa and $\Delta P/P = \pm 2.2378\%$. The maximum obtained deviation in $\Delta P = 209$ Pa and $\Delta P/P = \pm 3.5182\%$ at $T = 303.15$ K and $x = 0.7002$ mole fractions of 1-propanol.

The last 18 experimental values from the recent work of Cristino *et al.* [11] measured at high vapour pressure intervals of 304.2–967.4 kPa also exhibit small differences from our values as the mean deviation between two experimental sources is $\Delta P = \pm 5698$ Pa and $\Delta P/P = \pm 0.9227\%$. The maximum deviation of this comparison is $\Delta P = -21134$ Pa at $T = 413.2$ K and $x = 0.0002$ mole fractions of 1-propanol.

4. Conclusion

Vapour pressure measurements for the binary mixture of ethanol and 1-propanol over a wide range of temperatures from 274.15 K to 468.15 K were studied. The Antoine and Clausius–Clapeyron equations were used to fit the experimental results. The enthalpies of vaporisation at four various temperatures were calculated. The available literature values were compared with measured values and small deviations were observed.

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