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Vapor Pressure of 2-Butanol + Diethyl Carbonate and *tert*-Butanol + Diethyl Carbonate at the Temperature of 303.15–323.15 K

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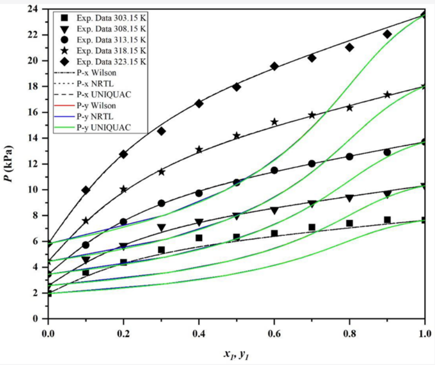
ABSTRACT: In this work, the vapor pressure of binary mixtures of 2-butanol + diethyl carbonate and *tert*-butanol + diethyl carbonate was measured in the temperature range from 303.15 to 323.15 K. The experimental apparatus used in this work was a simple quasi-static ebulliometer developed in our previous works. The reliability of this apparatus was verified by comparing the measured vapor pressures of 2-butanveol, *tert*-butanol, and diethyl carbonate with literature data. The comparisons showed that the vapor pressures of pure 2-butanol, *tert*-butanol, and diethyl carbonate were in good agreement with the literature data with average absolute deviations of 0.6, 0.6, and 0.8%, respectively. The experimental results show that the vapor pressures increased with the alcohol mole fraction for all systems studied. The experimental data were well-correlated with the Wilson, nonrandom two-liquid, and universal quasi-chemical activity coefficient models, giving an average absolute deviation of no more than 1.9%. The binary vapor−liquid equilibrium data obtained in this work showed a positive deviation from Raoult's law.



1. INTRODUCTION

Ethanol is commonly used in gasoline blends because of its high octane number and oxygen number as well as its capability to accelerate flame propagation. However, the addition of ethanol to isooctane at a mole fraction of 0.1 was found to elevate the vapor pressure.[1,2] A higher vapor pressure of gasoline mixtures causes increased emissions and the potential for vapor lock in the engine. On other hand, longer-chain alcohols, such as 2-butanol and *tert*-butanol, have lower vapor pressures and higher heating values, and their properties are closer to gasoline than to ethanol.[3] Butanol has been used as a mixture in engines without any significant engine changes.[4] Butanol may be produced from renewable resources as well, for example, from lignocellulose, the most plentiful renewable material.[5] The combustion of a butanol−gasoline mixture produces a high temperature because of its high heating value and low vaporization rate. Therefore, 2-butanol/*tert*-butanol are expected to be able to be used as gasoline additives that are alternatives to ethanol. The octane number of butanol is lower than that of ethanol, and thus, the addition of an octane booster is required. Diethyl carbonate (DEC) is such an octane booster. The addition of 5 wt % of diethyl carbonate (DEC) into diesel fuel can decrease the emissions by up to 50% due to its higher oxygen content of 40.6% wt.[6] DEC is a nontoxic chemical that is degradable and able to be decomposed gradually into CO_2 and ethanol, which are environmentally friendly.[7,8] Although methyl *tert*-butyl ether (MTBE) has been successfully applied in gasoline blends, as it is nonbiodegradable, it has contributed to groundwater contamination.[9] Thus, DEC is a potential candidate to replace MTBE.

To design the blend of gasoline + 2-butanol/*tert*-butanol + DEC, the vapor pressure data of the binary system of 2-butanol/*tert*-butanol + DEC are required. Studies on the vapor pressure of mixtures including diethyl carbonate have been conducted by many researchers. Rodriguez et al.[10] examined the vapor pressure for the binary systems of diethyl carbonate with five alcohols (methanol, ethanol, 1-propanol, 1-butanol, and 1-pentanol) at a pressure of 101.3 kPa and temperatures of 351.73−396.02 K. Octavian et al.[1] measured the vapor pressure of the ethanol + isooctane and 1-butanol + isooctane systems using a new ebulliometer. Ho et al.[11] examined the vapor pressure of binary mixtures containing diethyl carbonate, phenyl acetate, diphenyl carbonate, or ethyl acetate at 373.2−453.2 K. Jeremy et al.[12] conducted experiments on the vapor liquid equilibrium of a binary mixture of 2-propanone + 2-butanol at 333.15 and 353.15 K and 2-propanone + propanoic acid at 333.15, 353.15, and 373.15 K. Anugraha et al.[13] measured the vapor pressure of binary mixtures of diethyl carbonate + isooctane/*n*-heptane/toluene. To our knowledge,

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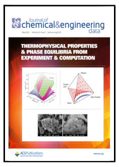

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vapor−liquid equilibrium (VLE) data of 2-butanol/*tert*-butanol + DEC at low temperatures are unavailable in the available literature. Therefore, the aim of this study is to determine the isothermal VLE of 2-butanol/*tert*-butanol + DEC at 303.15−323.15 K. In addition, the experimental data were correlated with the Wilson,[14] nonrandom two-liquid (NRTL),[15] and universal quasi-chemical (UNIQUAC)[16] models.

2. EXPERIMENTAL SECTION

2.1. Materials. The chemicals used in this experiment were 2-butanol, *tert*-butanol, and DEC. All chemicals were used without any additional purification. The compound details are listed in Table 1.

Table 1. Pure Chemicals Description and Properties

component	supplier	CAS reg. no.	MW[a] (g/mol)	purity[b]
2-butanol	Merck, Germany	78−92−2	74.12	0.9900
tert-butanol	Merck, Germany	75−65−0	74.12	0.9950
diethyl carbonate	Wuhan Fortuna Chemical Co., China	105−58−8	118.13	0.9992

[a] MW = molecular weight. [b] Purity from supplier in mass fraction.

2.2. Apparatus and Procedures. The apparatus used in this study is a simple static ebulliometer. The ebulliometer used was an apparatus developed by Oktavian et al.[1] and revalidated by Wibawa et al.,[17] Wiguno et al.,[18] and Anugraha et al.[13] to ensure that the initial composition has not changed significantly when the equilibrium condition is achieved. The detailed apparatus was shown in our previous publication.[1] The equipment's main parts are the ebulliometer cell, condenser, and some auxiliaries, such as a vacuum pump (VALUE VG140) for eliminating the gas impurities from the ebulliometer, magnetic stirrer, temperature controller, and indicator (AUTONICS TC4S), RTD Pt 100 thermocouple with an accuracy of ±0.1 K, pressure gauge (AUTONICS PSAN) with an accuracy of ±0.1 kPa, and ambient pressure gauge (Lutron MHB 382SD).

The experimental procedure was described in detail in the previous work.[19] Initially, each pure component's vapor pressure was measured by charging the pure component into the equilibrium cell, and vacuum conditions were created by turning on the vacuum pump. The pressure at each desired temperature was recorded as the vapor pressure. The vapor pressure data for the two binary systems, i.e., 2-butanol + DEC and *tert*-butanol + DEC, were obtained by the following procedure. The experiment was begun by introducing 225 mL of a mixture having a known composition into the ebulliometer cell. Cooling water was circulated in the condenser, and then the magnetic stirrer was switched on to stir the solution to mix evenly. After that, the vacuum pressure was created in the equilibrium cell by turning on the vacuum pump. The heating system was then lit to heat the solution according to the desired temperature. This heating causes some of the liquid to evaporate. The temperature and pressure in the system are shown by temperature indicator and pressure gauge, respectively. The pressure is recorded when the temperature reaches a constant value. The apparatus was validated by comparing the measured pure vapor pressures of 2-butanol, *tert*-butanol, and DEC with literature data calculated using the Wagner and Antoine equations with parameter constants obtained from Poling et al.[20] and Luo et al.,[21] respectively. Based on this experiment, the data obtained are the mole fractions of component i in the liquid phase (x_i), the equilibrium pressure (P) and the temperature (T). The experimental data are correlated with 3 activity coefficient models, i.e., the Wilson, NRTL, and UNIQUAC equations, and the binary interaction parameter pairs of each equation were optimized using the experimental data obtained in this work.

3. RESULTS AND DISCUSSION

3.1. Reliability of Apparatus. The validity of the experimental apparatus was checked by comparing the experimental pure vapor pressures of 2-butanol, *tert*-butanol, and DEC with the literature pure vapor pressures of 2-butanol and *tert*-butanol calculated from the Wagner equation (equation 1) and that of DEC calculated from the Antoine equation (equation 2). The parameter constants of the Wagner and Antoine equations are listed in Tables 2 and 3, respectively.

$$\ln P(\mathrm{kPa}) = \frac{a\tau + b\tau^{1.5} + c\tau^{2.5} + d\tau^{5}}{T_\mathrm{r}} \qquad (1)$$

$$\log_{10} P(\mathrm{kPa}) = A - \frac{B}{T(\mathrm{K}) + C} \qquad (2)$$

where: $\tau = 1 - T_\mathrm{r}$ and $T_\mathrm{r} = T/T_\mathrm{c}$.

Table 2. Wagner Parameter of 2-Butanol and *tert*-Butanol[20]

component	*a*	*b*	*c*	*d*
2-butanol	−8.0982	1.64406	−7.49	−5.2735
tert-butanol	−8.4792	2.47845	−9.279	−2.5399

Table 3. Antoine Parameter of DEC[21]

component	*A*	*B*	*C*
DEC	5.883	1223.77	−84.304

A comparison between the experimental data and literature data obtained from the Wagner or Antoine equations are presented in Table 4, and the average absolute deviation

Table 4. Vapor Pressures of 2-Butanol, *tert*-Butanol, and DEC[a]

	2-butanol		*tert*-butanol		DEC	
T/K	P_exp	P_lit	P_exp	P_lit	P_exp	P_lit
			(kPa)			
303.15	3.22	3.20	7.64	7.66	1.96	1.95
305.65	3.74	3.76	8.84	8.92	2.27	2.26
308.15	4.43	4.41	10.32	10.34	2.57	2.61
310.65	5.13	5.16	11.82	11.96	2.96	2.99
313.15	6.04	6.00	13.71	13.78	3.46	3.43
315.65	7.05	6.97	15.71	15.83	3.96	3.92
318.15	8.04	8.06	18.03	18.13	4.45	4.46
320.65	9.34	9.30	20.55	20.71	5.04	5.07
323.15	10.81	10.69	23.56	23.59	5.82	5.75
AAD	**0.6%**		**0.6%**		**0.8%**	

[a] The standard uncertainty of measurements are $u(T)$ = 0.1 K and $u(P)$ = 0.3 kPa, where $u(T)$ is the uncertainty in temperature and $u(P)$ is the uncertainty in pressure.

(AAD) between the experimental data and calculated vapor pressures were obtained by the following equation:

$$\mathrm{AAD} = \frac{1}{n}\sum_{i=1}^{n}\left|\frac{P_{\mathrm{exp}} - P_{\mathrm{lit}}}{P_{\mathrm{lit}}}\right| \times 100\% \tag{3}$$

where P_{exp} is the vapor pressure obtained from the experiment, P_{lit} is the literature value, and n is the number of data points. As presented in Table 4, good agreement was shown by all pure components with a 0.6% AAD for 2-butanol, 0.6% AAD for *tert*-butanol, and 0.8% AAD for DEC. The result indicates that the ebulliometer used in this experiment is reliable.

3.2. Vapor Pressure Data Measurement and Correlation. The vapor pressure data obtained in this work for the 2-butanol (1) + DEC (2) and *tert*-butanol (1) + DEC (2) systems are presented in Tables 5 and 6, respectively. The experimental equilibrium temperatures were set in the range of 303.15−323.15 K to accommodate common fuel storage conditions in tropical countries. This is done in the absence of changes in the pressure, temperature, and composition of the system.

Table 5. Experimental VLE data for 2-Butanol (1) + DEC (2)[a]

	P_{exp}/kPa				
x_1	303.15 K	308.15 K	313.15 K	318.15 K	323.15 K
0	1.96	2.57	3.46	4.45	5.82
0.1	2.72	3.73	4.74	6.16	7.89
0.2	2.93	4.02	5.42	7.01	8.62
0.3	2.98	4.28	5.78	7.49	9.41
0.4	3.17	4.31	5.61	7.42	9.61
0.5	3.23	4.52	5.93	7.92	9.93
0.6	3.4	4.49	5.88	7.75	10.22
0.7	3.05	4.44	6.01	8	10.67
0.8	3.41	4.61	6.03	7.96	10.56
0.9	3.27	4.67	6.26	8.07	10.76
1	3.22	4.43	6.04	8.04	10.81

[a] The standard uncertainty of measurements are $u(x_i) = 0.0002$, $u(T) = 0.1$ K, and $u(P) = 0.3$ kPa, where $u(x_i)$ is the uncertainty of component i of the liquid phase mole fractions, $u(T)$ is the uncertainty in temperature, and $u(P)$ is the uncertainty in pressure.

Table 6. Experimental VLE Data for *tert*-Butanol (1) + DEC (2)[a]

	P_{exp}/kPa				
x_1	303.15 K	308.15 K	313.15 K	318.15 K	323.15 K
0	1.96	2.57	3.46	4.45	5.82
0.1	3.62	4.61	5.71	7.6	9.97
0.2	4.37	5.67	7.51	10.05	12.75
0.3	5.34	7.14	8.95	11.38	14.54
0.4	6.26	7.54	9.72	13.12	16.68
0.5	6.33	8.02	10.55	14.2	17.96
0.6	6.62	8.46	11.5	15.26	19.57
0.7	7.1	8.97	12.02	15.78	20.21
0.8	7.4	9.4	12.57	16.35	21.03
0.9	7.67	9.69	12.91	17.36	22.06
1	7.64	10.32	13.71	18.03	23.56

[a] The standard uncertainty of measurements are $u(x_i) = 0.0002$, $u(T) = 0.1$ K, and $u(P) = 0.3$ kPa, where $u(x_i)$ is the uncertainty of component i of the liquid phase mole fractions, $u(T)$ is the uncertainty in temperature, and $u(P)$ is the uncertainty in pressure.

In vapor−liquid equilibrium conditions, the liquid-component fugacity is equal to the vapor-component fugacity. Due to low vapor pressure of the mixture, the ideal gas law is applied. Considering the differences between the alcohol and DEC molecules, the liquid becomes a nonideal mixture. Therefore, an activity coefficient of component i, γ_i, is used as a nonideal factor for the liquid phase in solution. Accordingly, the calculation of the vapor pressure of the mixture is based on the following equation.

$$P = \sum_{i=1}^{m} x_i \gamma_i P_i^{\mathrm{sat}} \tag{4}$$

where m is the number of components in the mixture, P is the vapor pressure in the equilibrium condition, x_i is the component i mole fraction in the liquid phase, γ_i is the activity coefficient of component i, and P_i^{sat} is the vapor pressure of component i. Data correlation is carried out by using the Wilson, NRTL, and UNIQUAC equations. The binary interaction parameters are determined using Barker's method[22] by minimizing the following objective function (OF):

$$\mathrm{OF} = \sum_{i=1}^{n} (\mathrm{P}_{i,\mathrm{cal}} - \mathrm{P}_{i,\mathrm{exp}})^2 \tag{5}$$

where n is the number of data points and the subscripts cal and exp indicate the calculated and experimental values, respectively. The values of the AAD are presented in Table 7.

Table 7. Parameter and AADs of Correlation Results

2-Butanol(1) + DEC(2) System						
Wilson		NRTL			UNIQUAC	
a_{12} (J/mol)	a_{21} (J/mol)	α (−)	b_{12} (J/mol)	b_{21} (J/mol)	Δu_{12} (J/mol)	Δu_{21} (J/mol)
5142.8	−977.7	0.3	−1050.6	4917.6	−1223.9	2590.1
AAD = 1.9%		AAD = 1.8%			AAD = 1.9%	
tert-butanol(1) + DEC(2) system						
Wilson		NRTL			UNIQUAC	
a_{12} (J/mol)	a_{21} (J/mol)	α (−)	b_{12} (J/mol)	b_{21} (J/mol)	Δu_{12} (J/mol)	Δu_{21} (J/mol)
1634.2	971.3	0.3	1466.9	995.1	108.1	460.3
AAD = 1.9%		AAD = 1.9%			AAD = 1.9%	

The experimental data were well-correlated with the Wilson, NRTL, and UNIQUAC activity coefficient models, giving average absolute deviations in the range of 1.8−1.9%. The correlation results were plotted in Figure 1 and Figure 2.

Figure 1 shows that the increasing temperature causes a rise in the vapor pressure of the 2-butanol + DEC mixture. At constant temperature, increasing the content of 2-butanol (x_1) leads to an increase of equilibrium pressure of the mixture to the peak point and then a decrease in the 2-butanol vapor pressure.

Figure 2 shows that the increasing temperature causes a rise in the vapor pressure of the mixture. In addition, at constant temperature, the vapor pressure of the mixture increases with the increasing content of *tert*-butanol (x_1). Therefore, the pressure of the mixture is between the pure vapor pressures of each component.

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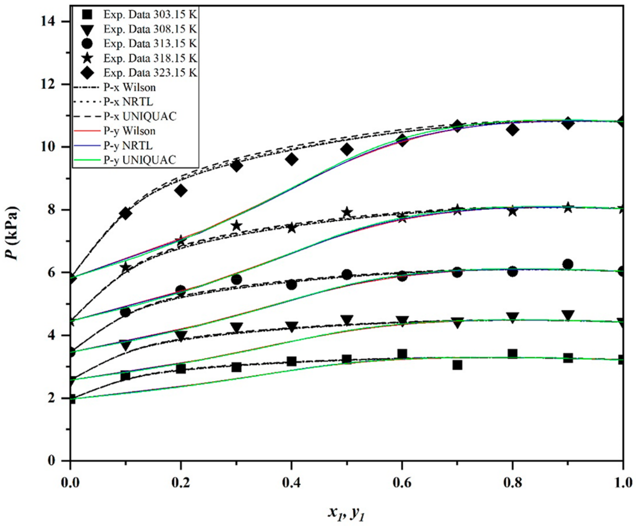

Figure 1. Diagram of P–x_1–y_1 for 2-butanol (1) + DEC (2) system at various temperatures.

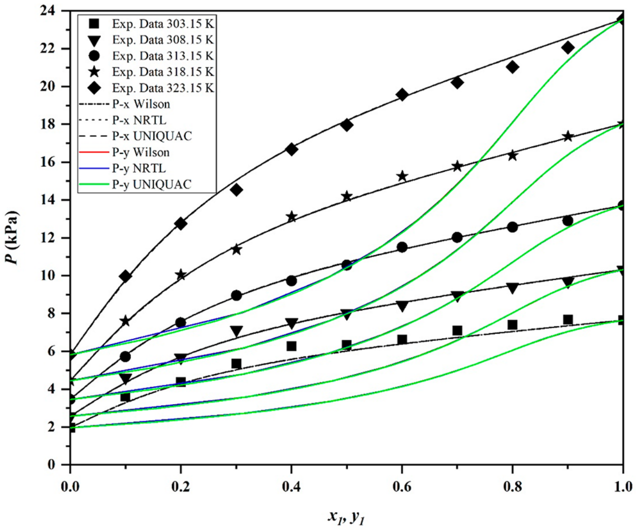

Figure 2. Diagram of P–x_1–y_1 for *tert*-butanol (1) + DEC (2) system at various temperatures.

The composition of the vapor can be obtained based on the concept of equilibrium by using the values of the activity coefficients obtained from the Wilson, NRTL, and UNIQUAC equations. The result of calculated of y_1 is then plotted in Figures 1 and 2.

According to Figure 1, the correlation results for 2-butanol + DEC showing that at the point of x_1 above 0.6, the x_1 has same value of y_1, where it could be suspected as azeotrope point. However, due to the differences between the maximum equilibrium pressures and the vapor pressure of pure 2-butanol are smaller than the uncertainty of pressure measurement (0.3 kPa), the azeotrope behavior still could not be justified for this system. The small vapor pressure differences indicate that the effect of DEC at the point of x_1 above 0.6 is insignificant. The vapor pressure is greatly affected by 2-butanol.

4. CONCLUSIONS

An experiment was successfully conducted to obtain accurate vapor–liquid equilibrium data for binary systems of 2-butanol (1) + DEC (2) and *tert*-butanol (1) + DEC (2) at 303.15–323.15 K. The experimental data for the 2-butanol (1) + DEC (2) system were well-correlated using the Wilson, NRTL and UNIQUAC models with AADs in the vapor pressure of 1.9, 1.8, and 1.9%, respectively. For the *tert*-butanol (1) + DEC (2) system, correlation using the Wilson, NRTL, and UNIQUAC models gave the same AAD of 1.9%. The systems studied show positive deviations from Raoult's law in the temperature range studied.

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Notes

The authors declare no competing financial interest.

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