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Vapour pressure of 1-butanol and Diesel B0 binary fuel blends

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Abstract: The vapour pressure of 1-butanol and Diesel B0 binary fuel blends were investigated at temperatures ranging from 274.15 to 468.67 K, using the two different setups with static method. The measured values were fitted to the Antoine, polynomial and Clausius–Clapeyron type equations. The heat of evaporation of mixture have been determined from the vapour–liquid equilibria data.

Keywords: vapor pressure; Diesel fuel; 1-butanol; static method; Antoine equation; Clausius–Clapeyron-type equation.

INTRODUCTION

Internal combustion engines driven by Diesel fuel are attractive in comparison to gasoline driven engines, because of the relative low CO_2 emissions, high power and reliable functionality. The stringent emission standards require the advanced Diesel engines technology with the improvement of the primary injection and combustion processes within the engine combustion chamber. One important development, successfully applied to meet the stringent legal requirements for emissions of Diesel engine, is the use of a common rail for fuel injection into the primary combustion chamber. Another current development is the use of alternative fuels instead of fossil Diesel, or mixtures of alternative fuels and fossil Diesel, so-called blends. In general, considering the dependence on the fuel properties, the injection rate and the combustion process for burning the alternative fuel in Diesel engines have to be properly adjusted and sometimes engines can run without modification.[1]

Alcohols have been used as alternative fuels or in blends in internal combustion engines for a long time. Typically, alcohol tends to decrease the internal combustion engine emissions.[2] Alcohol fuels often produces higher evaporative

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emissions than Diesel fuels due to higher vapour pressures and the relative low energy density of alcohol fuels, low heating value, cause a drop in engine performance if the engine management system does no compensation.[3] Alcohol fuels have a higher octane number than traditional fossil fuels and can be used as an octane booster for fuels. Nevertheless, alcohols have a lower cetane number, which limits the usage of neat alcohols in Diesel engine as an alternative fuel. Use of cetane enhancers can improve potential of alcohol fuel blends as a promising fuel for Diesel engine.[1] Using alcohol as addition to Diesel fuel changes some thermo-chemical properties of Diesel fuel, particularly reductions in cetane number, density, viscosity, vapour pressure, speed of sound, heat capacity etc., are necessary for the simulation of injection and combustion procedures. With the increase of the amount of alcohol in Diesel fuel mixture, there is an increase in both the ignition delay and the rate of initial heat release, premixed combustion, while there is a decrease in the diffusion combustion, the total combustion duration and the combustion temperature.[4] The increase of the alcohol percentage in Diesel mixtures also improves volatility and viscosity at low temperatures of blend.[5]

1-Butanol is also one of the primary alcohols, which has more advantages than ethanol and methanol as an alternative fuel. Like ethanol, 1-butanol can be a biomass based renewable fuel that can be produced by the alcoholic fermentation of the biomass feedstock used for ethanol production. 1-butanol possesses less hydrophilic tendency, higher heating value, higher cetane number, lower vapour pressure, higher viscosity and lubricity than ethanol and has a very good miscibility.[6]

Upon injection of the fuel in a cylinder, the large depressurization of the fuel results in a significant change of the thermophysical properties of the fluid.[1] For the optimal design of Diesel engine combustion and high pressure fuel injection process with the fuel mixtures concerning understanding, modelling and optimizing the spray formation, evaporation and combustion and pollutant formation, an accurate knowledge of the basic fuel thermophysical properties, like density, vapour pressure, viscosity, speed of sound, surface tension, heat capacity, bulk modulus, etc. as a function of pressure, temperature and composition, is required.[7]

This work is a continuation of our previous publications in the field of thermo-physical properties of 1-butanol and Diesel B0 binary fuel blends.[8,9] In this work, the vapour pressure of these binary fuel blends at T from 274.15 to 468.67 K were investigated. Vapour–liquid equilibria (VLE) play a crucial role in designing, modelling and control of process equipment.[10]

The results of the literature analysis show that there are no vapour pressure values of these fuel blends published earlier. In this case, the vapour pressure of 1-butanol and Diesel B0 binary fuel blends in all concentrations range were investigated in this work, using the high-accuracy fully automatic static experimental conditions, ultra-pure Merck and Shell quality substances at the first time.

EXPERIMENTAL

Materials. The ultra pure 1-butanol (99.995 %, absolute for analysis EMPLURA®, CAS No. 71-36-3, Art. No. 8.22262.2500) was purchased from Merck Schuchardt OHG, Germany, and was thoroughly degassed in glass flasks with special vacuum leak-proof valves before measurements. The water content in alcohols is determined by the Karl Fischer titration and was lower than a mass fraction of 20 ppm. The Shell Global Solution DK5037 Diesel B0 sample taken in 2015 was used during the preparation of 1-butanol and Diesel B0 binary fuel blends.

Experimental procedure. The vapour pressure measurements of 1-butanol and Diesel B0 binary fuel blends were measured using the two high-accuracy static experimental set ups.[11-13] The glass cells are used for vapour pressures lower than ambient pressure at temperatures from 274.15 to 323.15 K and the metal cell for vapour pressures at the temperatures from 323.15 to 468.67 K.

The glass cell method consists of the absolute and the differential parts (if the vapour pressure is lower than the uncertainty of absolute cell – 30 Pa). The vapour pressure of the 1-butanol and Diesel B0 binary fuel blends was each time higher than 30 Pa (uncertainty of measurements at T from 274.15 to 323.15 K). In this case, the measurements in this temperature interval were carried out using only the absolute cell of installation. 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 is 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. The experimental uncertainty of the pressure (Δp) in the absolute vapour pressure measurement using the glass cell is from ±10 to ±30 Pa. The temperature inside the cell is measured by a platinum resistance thermometer PT-100, connected to a signal conditioner Omega PT-104A, with an accuracy of ±0.001 K. Experiments were carried out starting from the low temperature ($T = 274.15$ K) to the high temperature $T = 323.15$ K at $\Delta T = 10$ K intervals using LabView computer programme control. The equilibration of the cells is a rapid process and a constant pressure in the stationary regime is reached within 15 min. The equilibrium pressure readings are performed 3 times approximately in 10–20 min intervals.

The experiments to determine the vapour pressure of liquids at the temperatures from 323.15 to 468.67 K are performed in a metal cell using the static method.[11-13] The internal volume of the measuring cell is approximately $V = 0.140\times10^{-3}$ m^3. The temperature of the measuring cell is controlled using a thermostat with an accuracy of $\Delta T = \pm0.01$ K. The temperatures are measured using two different platinum resistance thermometers, PT-100. The second platinum resistance thermometer, PT-100, transfers the measured temperature to the computer *via* an Omega PT-104A channel RTD input data acquisition module (Omega Engineering, Inc., USA) for the measuring of temperature, with the accuracy of $\Delta T = \pm0.001$ K. The vapour pressure is measured using the various Keller-Omega pressure transmitters: the maximum pressure of 300000 Pa, with uncertainty from ±400 to ±1500 Pa and the maximum pressure of 1000000 Pa with uncertainty from ±1000 to ±5000 Pa.

Before the both experiments the measuring cells were washed with water, methanol and acetone. All residual fluids were removed from the cells. This procedure requires approximately 2–3 h or more to reach the desired minimal pressure. At this point, all measuring cells were sufficiently dried and are ready for the experiments. The measuring cell was dried to the minimal vacuum pressure of 2–10 Pa. The experiments were carried out starting from the low temperature ($T = 333.15$ K) to the high temperature $T = 468.67$ K at $\Delta T = 10$ K intervals using LabView computer programme control.

The 1-butanol and Diesel fuel blends were prepared in full vacuum conditions, the specific quantities of alcohols and Diesel fuel were slowly evacuated, degassed in two separate flasks and mixed using an adapter.[12] All air and soluble gases were removed from the samples. Alcohol flowed into the flask with Diesel B0 sample and the concentration of the solution was defined using the weight of the flask with the solution on an electronic scale (Sartorius ED224S, Germany) with 0.0001 g uncertainty. The volume concentrations were calculated using the density of alcohols and Diesel fuel at room temperature, in which this process was carried out. The samples of 1-butanol and Diesel fuel blend were stable for 2 years (Fig. 1). The vapour pressure of the water[14] (APD in $\Delta p/p = \pm 0.106$ %), methanol[12,15] (APD in $\Delta p/p = \pm 0.11$ %), ethanol[16,17] (APD in $\Delta p/p = \pm 0.37$ %), 1-propanol[16] (APD in $\Delta p/p = \pm 0.38$ %), 1-butanol[18] (APD in $\Delta p/p = \pm 0.107$ %), toluene[19] (APD in $\Delta p/p = \pm 0.079$ %), *etc.* were measured as the reference substances for the testing of both setups.

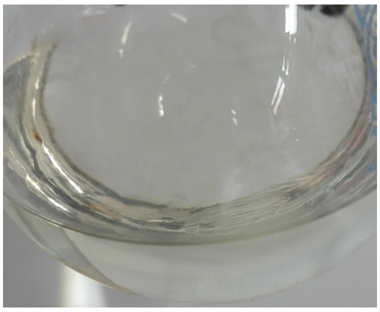

Fig. 1. Stable 1-butanol and Diesel fuel blend during the two years of control.

RESULTS

The measured experimental vapour pressures, p, of 1-butanol and Diesel B0 binary fuel blends at $T = 274.15$ to 468.67 K are listed in Table I. The vapour pressure of pure 1-butanol[18] and Diesel B0 fuel (sample from 2015)[8] were taken from our previous publications.

The experimental vapour pressure, p, results of investigated 1-butanol and Diesel B0 binary fuel blends were analysed by the Antoine equation:[20–22]

$$\ln p = A - \frac{B}{T + C} \tag{1}$$

where p is vapour pressure, Pa; T is the absolute temperature in K; A, B and C are the coefficients of Eq. (1) and presented in Table II, and depending on the mole fraction x of 1-butanol, in the following form:

$$A = \sum_{i=0}^{5} a_i x^i,\ B = \sum_{i=0}^{5} b_i x^i,\ C = \sum_{i=0}^{5} c_i x^i \tag{2}$$

where: a_i, b_i and c_i are the coefficients of Eq. (2) and are given in Table II. The average percent deviation (*APD*) between the experimental and the fitted vapour pressure values of 1-butanol and Diesel B0 blend is $u_r(\Delta p/p) = \pm 0.62$ %.

TABLE I. Experimental vapour pressure of 1-butanol and Diesel B0 binary fuel blends (p / Pa) at various temperatures and mole fractions x of 1-butanol; standard uncertainties, u, are $u(T) = 0.01$ K, $u(x) = 0.0001$ mole fraction 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, $U_c(P) = 5000$ Pa for $p < 10$ MPa (level of confidence = 0.95)

T / K	$x_{\text{1-Butanol}}$						
	0.0000[a]	0.1773	0.3493	0.5514	0.8112	0.9200	1.0000[b]
274.15	40	64	86	106	125	130	133
278.15	52	89	121	151	178	185	188
283.15	71	132	182	229	270	281	285
293.15	129	275	392	500	592	613	624
303.15	222	539	788	1017	1209	1257	1281
313.15	367	1000	1495	1946	2327	2428	2483
323.15	587	1770	2692	3530	4249	4452	4571
333.15	907	2999	4630	6109	7400	7788	8030
343.15	1363	4890	7646	10142	12360	13061	13517
353.15	1995	7701	12179	16227	19885	21084	21890
363.15	2851	11754	18777	25123	30930	32883	34224
373.15	3986	17441	28118	37764	46675	49713	51825
383.15	5465	25224	41010	55277	68528	73064	76225
393.15	7358	35638	58403	78989	98145	104659	109178
403.15	9745	49293	81387	110443	137422	146451	152632
413.15	12711	66868	111200	151393	188497	200599	208708
423.15	16348	89111	149216	203809	253738	269452	279656
433.15	20758	116829	196948	269872	335729	355517	367818
443.15	26043	150887	256033	351967	437246	461424	475581
453.15	32314	192194	328229	452673	561238	589895	605332
463.15	39684	241701	415397	574750	710801	743704	759418
468.67	44267	272898	470637	652350	805479	840476	855726

[a]The vapour pressure values of Diesel B0 were taken from ref. 8; [b]the vapour pressure values of 1-butanol were taken from [18]

The vapour pressure values of 1-butanol and Diesel B0 binary fuel blends were fitted also into the polynomial type equation:

$$\ln p = \sum_{i=0}^{4} x^i \sum_{j=0}^{4} a_{ij} \left(\frac{100}{T}\right)^j \tag{3}$$

where a_{ij} are the coefficients of Eq. (3) and shown in Table III. APD between experimental and fitted values using the polynomial equation is $u_r(\Delta p/p) = \pm 2.02$ %.

The vapour pressure values of alcohol and Diesel B0 fuel blends were also fitted to the extended version of Clausius–Clapeyron type equation:[23,24]

$$\ln p(T,x) = D(x) + \frac{E(x)T}{100} + F(x)\ln\frac{100}{T} + G(x)\frac{100}{T} + H(x)\left(\frac{100}{T}\right)^2 \qquad (4)$$

where p is vapour pressure*,* Pa; T is the absolute temperature in K; D, E, F, G and H are the coefficients of equation depending on the mole fraction x of 1-butanol.

TABLE II. The Antoine equation fitting parameters of 1-butanol and Diesel B0 binary fuel blends at various mole fractions x of 1-butanol; the vapour pressure values of Diesel B0 were taken from ref. 8 and 1-butanol from ref. 18

<table>
<tr><th colspan="4">Values of coefficients A, B and C in the Eq. (1)</th></tr>
<tr><th>x</th><th>A</th><th>B</th><th>C</th></tr>
<tr><td>0.0000</td><td>18.9970</td><td>3526.20</td><td>–43.7896</td></tr>
<tr><td>0.1773</td><td>21.2283</td><td>3455.04</td><td>–71.7903</td></tr>
<tr><td>0.3493</td><td>21.8295</td><td>3441.30</td><td>–76.1288</td></tr>
<tr><td>0.5514</td><td>22.1741</td><td>3432.77</td><td>–78.0762</td></tr>
<tr><td>0.8112</td><td>22.3787</td><td>3411.91</td><td>–79.7994</td></tr>
<tr><td>0.9200</td><td>22.3390</td><td>3354.50</td><td>–82.2984</td></tr>
<tr><td>1.0000</td><td>22.2600</td><td>3289.71</td><td>–85.0376</td></tr>
</table>

<table>
<tr><th colspan="3">Values of coefficients a_i, b_i and c_i in Eq. (2)</th></tr>
<tr><th>a_i</th><th>b_i</th><th>c_i</th></tr>
<tr><td>a_0 = 18.9974627779018</td><td>b_0 = 3526.00940253748</td><td>c_0 = –43.8089717786916</td></tr>
<tr><td>a_1 = 21.6820888203802</td><td>b_1 = –678.885447449982</td><td>c_1 = –294.486051019513</td></tr>
<tr><td>a_2 = –69.4251935840584</td><td>b_2 = 2169.2924286127</td><td>c_2 = 1061.94943505711</td></tr>
<tr><td>a_3 = 117.904750982299</td><td>b_3 = –3778.27123236656</td><td>c_3 = –1894.4982034415</td></tr>
<tr><td>a_4 = –97.2775978986173</td><td>b_4 = 3693.63104820251</td><td>c_4 = 1634.51284447312</td></tr>
<tr><td>a_5 = 30.3817404555156</td><td>b_5 = –1643.40536153316</td><td>c_5 = –548.842762004584</td></tr>
</table>

TABLE III. Polynomial type equation fitting parameters a_{ij} of 1-butanol and Diesel B0 binary fuel blends

a_{00} = 18.22893139	a_{14} =–708.3647551	a_{33} = 11455.76457
a_{01} = 6.17311432	a_{20} = –79.11707914	a_{34} = –6418.381103
a_{02} = 37.12846739	a_{21} = –450.0651401	a_{40} = –141.7215492
a_{03} = –78.647629	a_{22} = 3888.993667	a_{41} = 284.2822515
a_{04} =36.47503754	a_{23} = –6812.568969	a_{42} = 2609.455697
a_{10} = –23.69923978	a_{24} = 3376.310585	a_{43} = –7253.385913
a_{11} = 130.3511055	a_{30} = 147.111127	a_{44} = 4700.479372
a_{12} = –942.6745828	a_{31} = 91.48415756	
a_{13} = 1536.705486	a_{32} = –5380.172054	

$$D = \sum_{i=0}^{3} d_i x^i,\ E = \sum_{i=0}^{3} e_i x^i,\ F = \sum_{i=0}^{3} f_i x^i,\ G = \sum_{i=0}^{3} g_i x^i,\ H = \sum_{i=0}^{3} h_i x^i \qquad (5)$$

where: d_i, e_i, f_i, g_i and h_i are the coefficients of Eq. (5) and are tabulated in Table IV. The APD between experimental and fitted values using Eqs. (4) and (5) is $u_r(\Delta p/p) = \pm 6.69$ %.

TABLE IV. Clausius–Clapeyron equation fitting parameters, d_i, e_i, f_i, g_i and h_i of 1-butanol and Diesel B0 binary fuel blends

d_i	e_i	f_i	g_i	h_i
$d_0 = -159.816$	$e_0 = -7.16203$	$f_0 = -86.0816$	$g_0 = 345.969$	$h_0 = -242.395$
$d_1 = -219.883$	$e_1 = -9.84237$	$f_1 = -117.224$	$g_1 = 470.324$	$h_1 = -316.54$
$d_2 = 388.525$	$e_2 = 17.3569$	$f_2 = 206.694$	$g_2 = -834.269$	$h_2 = 569.737$
$d_3 = -160.05$	$e_3 = -7.10404$	$f_3 = -84.786$	$g_3 = 347.882$	$h_3 = -244.562$

The enthalpy change of vaporization, ΔH_v, of 1-butanol and Diesel B0 fuel blends for the four middle temperatures (for temperature interval 274.15–323.15 K in $T = 298.15$ K, for the temperature interval 323.15–373.15 K in $T = 348.15$ K, for the temperature interval 373.15–423.15 K in $T = 398.15$ K and for the temperature interval 423.15–468.67 K in $T = 448.15$ K) was defined using the Eq. (9),[25] which are defined at the result of following relations:

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

If we will plot ln p as function of $1/T$, we can define ΔH_v from the slope of the line:

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

After the integration of Eq. (7) we can find:

$$\ln p = \left(-\frac{\Delta H_\text{v}}{R}\right)\left(\frac{1}{T}\right) + C \tag{8}$$

$$\Delta H_\text{v} = RT(C - \ln p) \tag{9}$$

The calculated enthalpy changes of vaporization, ΔH_v / J mol^{-1}, of 1-butanol and Diesel B0 binary fuel blends from 274.15 to 468.67 K are listed in Table V.

TABLE V. Enthalpy changes of vaporisation, ΔH_v / J mol^{-1}, of 1-butanol and Diesel B0 binary fuel blends

T / K	$x_\text{1-Butanol}$						
	0.0000	0.1773	0.3493	0.5514	0.8112	0.9200	1.0000
298.15	40372	49901	51886	52754	53035	53136	53239
348.15	38420	45876	47038	47513	48049	48380	48691
398.15	37053	42822	43816	44257	44450	44374	44259
448.15	36073	40524	41592	42125	41822	41183	40484

CONCLUSION

The vapour pressure measurements of 1-butanol and Diesel B0 binary fuel blends over a wide range of temperatures (274.15–468.67 K) were studied for the first time. The Antoine, the polynomial and the Lausius–Clapeyron equations were used to fit the experimental results. The enthalpy changes of vaporization, ΔH_v / J mol^{-1}, at various four temperatures were calculated. The available literature values were compared with the measured values and good deviation between of them were obtained. After considering all three equations, it was proved that the best fitting equation is the Antoine equation with the $u_\mathrm{r}(\Delta p/p)$ = ±0.62 % average deviation.

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LIST OF SYMBOLS

p	vapour pressure
T	absolute temperature
w	mass percent
x	mole fraction
APD	average percent deviation

ИЗВОД

НАПОН ПАРЕ БИНАРНИХ СМЕША 1-БУТАНОЛА И ДИЗЕЛ ГОРИВА

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Напон паре бинарних смеша 1-бутанола и дизел горива је испитиван у температурном опсегу од 274,15–468,67 K, применом две различите поставке са статичким методама за експериментална мерења. Измерене вредности су корелисане помоћу различитих модела: Антоановом једначином, једначином полиномског типа и Клаузијус–Клапејроновом једначином. Из експерименталних података за равнотежу пара-течност одређене су топлоте испаравања испитиваних смеша.

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