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Liquid density of biofuel mixtures: (Dibutyl ether + 1-butanol) system at pressures up to 140 MPa and temperatures from (293.15 to 393.15) K

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ABSTRACT

This work reports new experimental density data (445 points) for binary mixtures of (dibutyl ether + 1-butanol) over the composition range (five compositions; $0.15 \leqslant$ dibutyl ether mole fraction $x \leqslant 0.85$), from (293.15 to 393.15) K (every 20 K), and for 15 pressures from (0.1 to 140) MPa (every 10 MPa).

An Anton Paar vibrating tube densimeter, calibrated with an uncertainty of $\pm 0.5\ \text{kg} \cdot \text{m}^{-3}$ was used to perform these measurements. The experimental density data were fitted with a Tait-like equation with low standard deviations. Excess volumes have been calculated from the experimental data and fitted by the Redlich–Kister equation. In addition, the isobaric thermal expansivity and the isothermal compressibility have been derived from the Tait-like equation.

1. Introduction

Increasing global concern due greenhouse gas emissions has generated much interest in the environmental friendly alternative bio-fuels. Bio-fuels for internal combustion engines as oxygenated compounds are also becoming important because of diminishing petroleum reserves and increasing air pollution [1].

The oxygenated compounds used worldwide like simple alcohols and ethers, are used as gasoline additives to reduce pollutants from vehicle exhaust gases. Proponents of these oxygenates claim several advantages: they are octane enhancers, they have significant anti-knock properties important for unleaded fuels, they can be produced from renewable agricultural and raw materials instead of fossil sources, and they reduce carbon monoxide pollution from vehicle exhaust [2–4].

In this sense, there has been an increasing interest in the thermodynamic behaviour of liquid mixtures of alcohols with ethers. From a thermodynamic point of view, experimental property studies of binary mixtures can provide valuable information about the fluid behaviour under various temperature and pressure conditions. One of these properties is the density or specific molar volume, which is an important property required in a wide range of engineering disciplines as well as in the determination of different fluid properties. This work continues the research on binary mixtures of alcohols and ethers of our group. In previous works [5,6], we have studied high pressure and high temperature of 1-butanol and dibutyl ether (DBE or 1-butoxybutane) as pure compounds. With the aim of studying the intermolecular interactions during the mixing process, this work presents the experimental densities of the system (DBE + 1-butanol) at pressures from (0.1 to 140) MPa and temperatures from (293.15 to 393.15) K over the full composition range. We report densities, excess molar volumes, isobaric thermal expansion coefficient and the isothermal compressibility coefficient for the binary mixtures (dibutyl ether + 1-butanol) at elevated pressures.

2. Experimental

2.1. Materials

Dibutyl ether ($C_8H_{18}O$, molar mass $130.228\ \text{g} \cdot \text{mol}^{-1}$, CAS. 142-96-1) and 1-butanol ($C_4H_{10}O$, molar mass $74.12\ \text{g} \cdot \text{mol}^{-1}$, CAS. 71-36-3) were obtained from Sigma–Aldrich with mole fraction purity of respectively, 0.993 and 0.999 (with certificate of analysis by gas chromatography of 1-butanol 0.9993). These chemicals were subject to no further purification and directly injected into the high pressure cell as soon as the bottles were open.

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TABLE 1
Experimental densities, $\rho/(g \cdot cm^{-3})$, for (x dibutyl ether + (1 − x) 1-butanol) at various temperatures t and pressures p.

x	p/MPa	T/K					
		293.15	313.15	333.15	353.15	373.15	393.15
		$\rho/(g \cdot cm^{-3})$					
0.1553	0.1	0.8000	0.7839	0.7677	0.7495	0.7305	
	10	0.8076	0.7920	0.7766	0.7587	0.7421	0.7242
	20	0.8144	0.7993	0.7847	0.7689	0.7526	0.7359
	30	0.8206	0.8063	0.7922	0.7771	0.7620	0.7458
	40	0.8264	0.8125	0.7990	0.7846	0.7700	0.7552
	50	0.8319	0.8185	0.8054	0.7916	0.7775	0.7630
	60	0.8370	0.8240	0.8113	0.7979	0.7845	0.7706
	70	0.8419	0.8294	0.8168	0.8039	0.7909	0.7776
	80	0.8467	0.8343	0.8222	0.8097	0.7969	0.7842
	90	0.8510	0.8390	0.8275	0.8150	0.8027	0.7901
	100	0.8555	0.8434	0.8322	0.8200	0.8081	0.7958
	110	0.8594	0.8478	0.8367	0.8248	0.8133	0.8014
	120	0.8634	0.8520	0.8406	0.8295	0.8179	0.8065
	130	0.8672	0.8560	0.8451	0.8339	0.8226	0.8114
	140	0.8709	0.8599	0.8490	0.8382	0.8270	0.8162
0.3249	0.1	0.7915	0.7750	0.7582	0.7397	0.7204	
	10	0.7990	0.7832	0.7676	0.7505	0.7327	0.7145
	20	0.8058	0.7908	0.7761	0.7601	0.7437	0.7268
	30	0.8126	0.7979	0.7838	0.7687	0.7534	0.7371
	40	0.8182	0.8043	0.7909	0.7765	0.7618	0.7469
	50	0.8238	0.8104	0.7975	0.7836	0.7695	0.7549
	60	0.8290	0.8161	0.8036	0.7902	0.7767	0.7626
	70	0.8339	0.8216	0.8092	0.7963	0.7834	0.7698
	80	0.8388	0.8266	0.8147	0.8020	0.7895	0.7766
	90	0.8432	0.8315	0.8201	0.8075	0.7954	0.7827
	100	0.8478	0.8360	0.8249	0.8125	0.8010	0.7886
	110	0.8518	0.8405	0.8295	0.8175	0.8064	0.7943
	120	0.8558	0.8448	0.8334	0.8223	0.8110	0.7994
	130	0.8597	0.8488	0.8380	0.8268	0.8158	0.8044
	140	0.8634	0.8529	0.8420	0.8311	0.8202	0.8092
0.4979	0.1	0.7841	0.7671	0.7499	0.7312	0.7119	
	10	0.7919	0.7758	0.7598	0.7424	0.7250	0.7069
	20	0.7991	0.7837	0.7685	0.7524	0.7363	0.7197
	30	0.8055	0.7910	0.7765	0.7612	0.7461	0.7303
	40	0.8117	0.7976	0.7838	0.7692	0.7546	0.7403
	50	0.8174	0.8039	0.7905	0.7766	0.7626	0.7486
	60	0.8226	0.8097	0.7967	0.7832	0.7699	0.7566
	70	0.8278	0.8154	0.8025	0.7895	0.7767	0.7638
	80	0.8326	0.8205	0.8082	0.7956	0.7830	0.7707
	90	0.8371	0.8254	0.8137	0.8012	0.7890	0.7769
	100	0.8418	0.8300	0.8186	0.8064	0.7946	0.7825
	110	0.8459	0.8345	0.8231	0.8114	0.8000	0.7883
	120	0.8500	0.8390	0.8272	0.8162	0.8048	0.7935
	130	0.8540	0.8430	0.8318	0.8193	0.8097	0.7986
	140	0.8577	0.8471	0.8359	0.8252	0.8143	0.8035
0.6748	0.1	0.7778	0.7606	0.7432	0.7245	0.7049	
	10	0.7858	0.7695	0.7534	0.7361	0.7184	0.7007
	20	0.7931	0.7775	0.7625	0.7463	0.7301	0.7138
	30	0.7997	0.7850	0.7706	0.7553	0.7403	0.7247
	40	0.8060	0.7918	0.7780	0.7635	0.7491	0.7349
	50	0.8117	0.7982	0.7849	0.7710	0.7572	0.7433
	60	0.8172	0.8041	0.7913	0.7778	0.7647	0.7513
	70	0.8223	0.8098	0.7972	0.7842	0.7716	0.7587
	80	0.8273	0.8150	0.8029	0.7904	0.7779	0.7657
	90	0.8319	0.8200	0.8085	0.7961	0.7841	0.7720
	100	0.8366	0.8247	0.8135	0.8013	0.7897	0.7780
	110	0.8407	0.8293	0.8181	0.8064	0.7953	0.7838
	120	0.8448	0.8337	0.8222	0.8113	0.8001	0.7891
	130	0.8488	0.8379	0.8268	0.8159	0.8051	0.7942
	140	0.8526	0.8420	0.8310	0.8204	0.8096	0.7991
0.8501	0.1	0.7724	0.7550	0.7374	0.7187	0.6996	
	10	0.7806	0.7642	0.7478	0.7305	0.7134	0.6962
	20	0.7880	0.7724	0.7571	0.7410	0.7253	0.7095
	30	0.7948	0.7800	0.7654	0.7502	0.7357	0.7206
	40	0.8011	0.7869	0.7730	0.7585	0.7446	0.7309
	50	0.8070	0.7934	0.7800	0.7661	0.7528	0.7393
	60	0.8124	0.7994	0.7864	0.7730	0.7604	0.7475
	70	0.8177	0.8052	0.7924	0.7795	0.7674	0.7549
	80	0.8227	0.8105	0.7982	0.7857	0.7738	0.7620

TABLE 1 (*continued*)

x	p/MPa	T/K					
		293.15	313.15	333.15	353.15	373.15	393.15
	90	0.8274	0.8156	0.8038	0.7915	0.7800	0.7683
	100	0.8321	0.8203	0.8089	0.7968	0.7857	0.7743
	110	0.8363	0.8249	0.8137	0.8019	0.7913	0.7802
	120	0.8405	0.8294	0.8178	0.8069	0.7961	0.7854
	130	0.8445	0.8336	0.8225	0.8115	0.8011	0.7906
	140	0.8483	0.8378	0.8267	0.8160	0.8057	0.7956

2.2. Measurement technique. Experimental procedure

An Anton-Paar DMA HPM high-pressure vibrating-tube densimeter was used to measure the density ρ as a function of pressure p (up to 140 MPa) and temperature T from (293.15 to 393.15) K. The experimental setup was similar to the one described in a previous paper [7]. The calibration of the densimeter was performed according to the new procedure described by Comuñas *et al.* [8] which is modification of the procedure previously proposed by Lagourette *et al.* [9].

After the densimeter was filled with the sample to be studied as described in reference [7], the sample was brought to the desired temperature and pressure of interest and measured when thermal and mechanical equilibrium were reached. The temperature of the high-pressure vibrating-tube cell of the densimeter was controlled by an external circulating temperature-controlled fluid and was measured inside the high-pressure cell. With an AOIP PHP602 thermometer with an uncertainty of ±0.05 K above 353.15 K, and with an Anton-Paar CKT100 with an uncertainty of ±0.01 K from (293.15 to 353.15) K. Above atmospheric pressure, the pressure was measured with a digital pressure transmitter (Presens Precise Gold Plus) with an uncertainty of ±0.015 MPa (1/10,000 of the full scale).

For calibration of the densimeter at high pressure and $293.15\ K \leqslant T \leqslant 363.15\ K$, we used water and vacuum as references measurement. Taking into account the accuracy of the temperature, the pressure, the period of oscillation measurement for water, vacuum and the studied systems, and the water density accuracy, the overall experimental uncertainty in the reported density values is estimated to be $\pm 0.0005\ g \cdot cm^{-3}$ (i.e. around ± 0.05% for density close to water density). This uncertainty is similar to that reported in several studies [7,8,10–13]. However, for measurements at atmospheric pressure at T = (373.15 and 393.15) K we used decane instead of water as reference fluid. The uncertainty of the decane density data reported by TRC studies [14] (of the order of $\pm 0.0001\ g \cdot cm^{-3}$) is greater than that of water and consequently for the two data points at 373.15 K and 393.15 K, at p = 0.1 MPa the overall uncertainty is estimated to be less than ±0.5%. No measurements have been done at p = 0.1 MPa and T = 393.15 K because the boiling temperature of 1-butanol is 390.6 K.

The binary (DBE + 1-butanol) mixtures (x is the DBE molar fraction) were prepared immediately before use by weighing at atmospheric pressure using a high-precision Sartorius balance with an uncertainty of ±0.001 g. For each mixture, a sample of 50 g was prepared, which, taking into account the uncertainty of the balance, resulted in an uncertainty in the mole fraction of less than $6 \cdot 10^{-5}$.

3. Results and discussion

3.1. Density

The measured densities of (dibutyl ether + 1-butanol) (five DBE molar ratio composition x: 0.1553, 0.3249, 0.4979, 0.6748, 0.8501)