



Liquid densities and excess volumes of biofuel mixtures: (2-butanol + di-isopropyl ether) system at pressures up to 140 MPa and temperatures from 293.15 K to 393.28 K



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ABSTRACT

This work reports new experimental density data for binary mixtures of 2-butanol + di-isopropyl ether over the composition range, between 293.15 and 393.28 K, and for 23 pressures from 0.1 MPa up to 140 MPa. A vibrating tube densitometer, calibrated with an uncertainty of $\pm 0.7 \text{ kg}\cdot\text{m}^{-3}$ 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. In addition, the isobaric thermal expansivity and the isothermal compressibility have been derived from the Tait-like equation.

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1. Introduction

The scarcity of known petroleum reserves will make renewable energy resources more attractive [1–3]. Today fossil fuels take up 80% of the primary energy consumed in the world, of which 58% is consumed by the transport sector [4]. The most feasible way to meet this growing demand is by utilizing alternative fuels [5,6]. Biofuels containing oxygenated compounds are the best candidate for new fuels in reciprocating engines [7]. The biggest advantage that biofuel has over petroleum gasoline and diesel is its environmental friendliness [8].

Oxygenated compounds are generally added to gasoline to increase combustion efficiency and to reduce air pollution. Usage of oxygenated additives have been recognized as safe, efficient, and cost-effective way to reduce the levels of mainly soot and particulate emissions and to improve combustion. During the last two decades, various ethers, either alone or in mixture with other ethers or alcohols, are widely used to increase the amount of oxygen in fuel and enhance the octane rating, improving combustion

and reducing emissions and the contaminant agents of automobile catalysts [9–13].

The thermodynamic properties of binary liquid mixtures containing oxygenated compounds, such as ethers and alcohols have been the subject of interest as a basis for checking the possibility of predicting the behaviour of complex mixtures containing polar compounds. 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 [14,15]. In previous works [16,17], we have studied high pressure and high temperature of di-isopropyl ether (DIPE) and 2-butanol as pure compounds. With the aim of studying the intermolecular interactions during the mixing process, this work presents the experimental densities of the system (2-butanol + DIPE) at pressures from (0.1 to 140) MPa and temperatures from (293.15 to 393.28) K over the full composition range (1269 points). We report densities, excess molar volumes, isobaric thermal expansion coefficient and the isothermal compressibility coefficient for the binary mixtures (2-butanol + DIPE) at high pressures.

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Nomenclature

List of symbols

AAD	Absolute Average Deviation
a_i	coefficients of isobaric thermal expansivity correlation
A_i, B_i, C	coefficients of density correlation
Bias	Average Deviation
calc	calculated
exp	experimental
i	constituent identification
lit	literature
MD	Maximum Deviation
N	number of experimental data points which are in our p, T ranges
p	pressure

p_0	reference pressure
T	temperature
V	volume
V^E	excess molar volumes
x	molar fraction

Greek letters

σ	standard deviation
α_p	isobaric thermal expansivity
ρ	density
ρ_0	density at a reference pressure p_0
κ_T	isothermal compressibility

2. Experimental

2.1. Materials

2-butanol and di-isopropyl ether were obtained from Sigma-Aldrich with mole fraction purity of 0.995, as shown in Table 1. Both liquids were stored over a molecular sieve to avoid any moisture and were used without any further purification except careful degassing before the use.

2.2. Measurement technique. Experimental procedure

An Anton Paar DMA HPM high pressure vibrating tube densitometer was used to measure the experimental densities. This type of densitometer can be used to perform measurements in a broad range of temperature and pressure. The experimental setup was similar to the one described in a previous paper [18]. The calibration of the densitometer was performed according to the procedure described by Comuñas et al. [19] which is a modification of the procedure previously proposed by Lagourette et al. [20]. Two reference fluids were used to perform the calibrations (vacuum and water). The density values of water were taken from the equation of state (EoS) reported by Wagner and Pruss [21]. After filling the densitometer with the sample, it was brought to the desired temperature and pressure of interest and measured when the system reached both thermal and mechanical equilibrium.

The estimated expanded uncertainty of the measured temperature was ± 0.03 K (Pt 100 calibrated probe). The estimated expanded uncertainty of the measured pressure was ± 0.04 MPa (pressure transducer WIKA CPH 6000). The DMA HPM measuring cell is connected to the Anton Paar mPDS 2000V3 evaluation unit, which evaluates the oscillation period from the measuring cell filled with the sample. 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 estimated expanded density uncertainty ($k = 2$) is

$\pm 0.7 \text{ kg}\cdot\text{m}^{-3}$ (i.e., around $\pm 0.07\%$ for density close to water density), following the EA-4/02 document [22].

Pure fluids used in binary mixtures were degassed prior to the preparation of the samples. Degassing is carried out using an ultrasonic bath PSelecta, model Ultrason-H. Each mixture was prepared immediately before measuring, by weighing in glass vials sealed to prevent evaporation. For weighing, a Sartorius balance model BP 221S has been used, with resolution of 10^{-4} g, uncertainty ± 0.0001 g. The estimated expanded uncertainty in the composition of the mixture is $\pm 4 \cdot 10^{-5}$ in mole fraction. Therefore, the excess molar volume should be accurate to within $\pm 0.004 \text{ cm}^3\cdot\text{mol}^{-1}$.

3. Results and discussion

3.1. Density

The measured densities of (2-butanol + di-isopropyl ether) (2-butanol mole fractions x : 0.0000, 0.2036, 0.3227, 0.4464, 0.4973, 0.6595, 0.8482, 1.0000) are reported in Table 2 along the seven isotherms from (293.15 to 393.28) K at pressures up to 140 MPa (23 isobars). The density of pure DIPE and pure 2-butanol were previously reported in [16,17] and have been measured again in this work as recalibration of the experimental technique was performed between both sets of measurements. Comparison of the two sets of data gives an absolute average deviation of $2.5 \cdot 10^{-5} \text{ g}\cdot\text{cm}^{-3}$ for 2-butanol and $8.5 \cdot 10^{-5} \text{ g}\cdot\text{cm}^{-3}$ for DIPE, showing a very good repeatability of the technique as these values are much lower than density uncertainty. No measurements were made at $p = 0.1$ MPa and at 353.19 K, at 373.24 K and at 393.28 K for pure DIPE because its boiling point is 341.5 K. For pure 2-butanol, as its boiling point is 371.15 K, no measurements were performed at $p = 0.1$ MPa and at 373.24 K and at 393.28 K.

Several fitting parameters were used for the purpose of comparing the experimental density values with those obtained with the two correlations considered in this work. The Absolute Average Deviation (AAD), the Maximum Deviation (MD), the Average Deviation (Bias), and the standard deviation (σ) are defined as follows:

Table 1
Purity and related data of chemicals.

Compound	Molecular formula	Molar mass ($\text{g}\cdot\text{mol}^{-1}$)	Stated purity ^a (mol%)	CAS number
2-butanol ^b	$\text{C}_4\text{H}_{10}\text{O}$	74.12	99.5	78-92-2
DIPE ^b	$\text{C}_6\text{H}_{14}\text{O}$	102.17	99.5 ^c	108-20-3

^a Determined by gas chromatography (GC).

^b Supplied by Sigma Aldrich.

^c The water content was checked to be less than 0.01% by titration method.

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