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Liquid Density of Biofuel Additives: 1-Butoxybutane at Pressures up to 140 MPa and from (293.15 to 393.15) K[†]

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ABSTRACT: This work reports new density data (90 points) of 1-butoxybutane (also known as dibutyl ether, DBE) at six temperatures between (293.15 and 393.15) K (every 20 K) and 15 pressures from (0.1 to 140) MPa (every 10 MPa). An Anton Paar vibrating tube densimeter, calibrated with an uncertainty of ± 0.5 $\text{kg}\cdot\text{m}^{-3}$, was used to perform these measurements. The experimental density data were fitted with the Tait-like equation with low standard deviations. In addition, the isobaric thermal expansivity and the isothermal compressibility have been derived from the Tait-like equation.

INTRODUCTION

The increasing worldwide use of biofuels constitutes one of the measures considered to reduce greenhouse gas emissions. Biofuels also have an important part to play in promoting the security of energy supply and promoting technological development and innovation. 1-Butoxybutane (DBE) is used as a blending agent in reformulated gasoline and has been included in recent international regulations on the promotion of the use of energy from renewable sources for transport.[1] DBE acts as a nonpolluting, high octane number blending agent. DBE could be also used as a cetane enhancer in biodiesel fuel and can be obtained as an added valued additive to second-generation biofuels.[2]

$p\rho T$ data have great importance from a theoretical point of view. Reliability of molecular-based equations of state are often analyzed by comparing their predictions with $p\rho T$ experimental values, as mentioned by Zuñiga-Moreno et al.[3,4] Moreover, for the optimized design of several industrial processes (storage, transport, separation, and mixing processes), reliable experimental data are needed. For these reasons, this work aims to provide experimental information on the volumetric properties which should be useful for modeling purposes.

This work concerns DBE. The density values for DBE obtained in the present work (90 experimental data) will be compared with previous literature data, which are rather scarce. To the extent we know, liquid densities for DBE at high pressure have been measured by Meng et al.,[5] which provide 70 data points between $T = (243.15 \text{ and } 373.15)$ K and between $p = (0.1 \text{ to } 21.1)$ MPa with a claimed uncertainty of ± 0.2 % (i.e., around 2 $\text{kg}\cdot\text{m}^{-3}$ for density close to water density). As we can see, the review of the currently available literature sources reveals that there is still a lack of information on the density of DBE, particularly at high pressures. Therefore, the database for thermophysical properties of this compound, at present, is very limited. Moreover, the study of this compound presents obvious industrial interest in engineering applications, and it presents also fundamental aspects as DBE is a very associative compound. We can expect that these experimental data will be valuable for testing various theoretical models attempting to take into account the polar and associating effects between molecules. Previous works[6,7] on alternative oxygenate additives to biofuels have been carried out.

EXPERIMENTAL SECTION

Materials. DBE ($C_8H_{18}O$, molar mass 130.228 $\text{g}\cdot\text{mol}^{-1}$, CAS. 142-96-1) was obtained from Sigma-Aldrich with mole fraction purity of 0.993. This chemical was subject to no further purification and directly injected into the high-pressure cell as soon as the bottle was open.

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 (between (293.15 and 393.15) K). The experimental setup was similar to the one described in a previous paper.[8] The calibration of the densimeter was performed according to the new procedure described by Comuñas et al.[9] which is a modification of the procedure previously proposed by Lagourette et al.[10]

(a) At $(0.1 \le p \le 140)$ MPa and $(293.15 \le T \le 363.15)$ K. The densimeter calibration over these temperature and pressure intervals has been performed by using the procedure previously proposed by Lagourette et al.[10] The density values of water have been taken from the equation of state (EoS) reported by Wagner and Pruss.[11] The uncertainty in density of this EoS is ± 0.0001 % at 0.1 MPa in the liquid phase, ± 0.001 % at other liquid states at pressures up to 10 MPa and temperatures to 423

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K, and ± 0.003 % at pressures in the interval (10 to 100) MPa and temperature up to 423 K. The uncertainty is of the order of ± 0.02 % at 1000 MPa.

(b) At $p = 0.1$ MPa and $T \geq 373.15$ K. The limitation of this procedure[10] appears when the measurements are performed at 0.1 MPa and at temperatures greater than or equal to the boiling point of water (373.15 K). This is the case of the present work because the study is done up to temperatures above 373.15 K. So, in this work decane has been selected as a reference substance at the conditions $p = 0.1$ MPa and $T \geq 373.15$ K because its density is well-known at atmospheric pressure over wide temperature intervals.[12]

(c) At $p > 0.1$ MPa and $T \geq 373.15$ K. One can think that in the interval (0 to 1) MPa, as the pressure expansion coefficient of the cell is very low, the volume of the cell does not significantly change with pressure. Following the new procedure proposed by Watson et al.,[8] the reference pressure for water density is 1 MPa instead of 0.1 MPa. It is necessary to know the oscillation period of the evacuated cell (over the entire temperature interval) and the period of the cell full of water and the density of water (for $p > 0.1$ MPa and for $T \geq 373.15$ K). Over the above T, p intervals, only one reference fluid is necessary (water) together with the period of the evacuated cell.

After the densimeter was filled with the sample to be studied as described in Watson et al.,[8] 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 between (293.15 and 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).

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 ± 0.5 $\mathrm{kg \cdot m^{-3}}$ (i.e., around ± 0.05 % for density close to water density). This uncertainty is similar to that reported in several studies.[8,9,13−16] However, for measurements at atmospheric pressure at $T = (373.15$ and $393.15)$ K, we used decane as the reference fluid. The uncertainty of the decane density data reported by TRC[12] (of the order of ± 0.0001 $\mathrm{g \cdot cm^{-3}}$) is greater than that of water and consequently for the two data points at (373.15 and 393.15) K at $p = 0.1$ MPa the overall uncertainty is estimated to be less than ± 0.5 %.

■ RESULTS

The measured densities of DBE are reported in Table 1 along the six isotherms between (293.15 and 393.15) K at pressures up to 140 MPa (15 isobars).

Table 1. DBE Density, ρ, versus Temperature and Pressure

	T/K					
p/MPa	293.15	313.15	333.15	353.15	373.15	393.15
	$\rho/\mathrm{g \cdot cm^{-3}}$					
0.1	0.7680	0.7505	0.7335	0.7151	0.6964	0.6773
10	0.7764	0.7598	0.7441	0.7272	0.7103	0.6939
20	0.7839	0.7681	0.7535	0.7378	0.7223	0.7073
30	0.7908	0.7759	0.7619	0.7471	0.7328	0.7184
40	0.7972	0.7828	0.7695	0.7555	0.7418	0.7287
50	0.8032	0.7894	0.7766	0.7632	0.7501	0.7372
60	0.8087	0.7954	0.7831	0.7702	0.7576	0.7453
70	0.8140	0.8015	0.7892	0.7764	0.7647	0.7528
80	0.8191	0.8068	0.7950	0.7827	0.7711	0.7599
90	0.8238	0.8120	0.8007	0.7884	0.7772	0.7662
100	0.8286	0.8167	0.8057	0.7940	0.7830	0.7722
110	0.8328	0.8214	0.8105	0.7992	0.7886	0.7781
120	0.8370	0.8259	0.8147	0.8042	0.7935	0.7834
130	0.8410	0.8302	0.8194	0.8086	0.7985	0.7886
140	0.8449	0.8343	0.8237	0.8133	0.8031	0.7935

For the purpose of comparing the experimental density values with those obtained with the correlation considered in this work, we have used the Absolute Average Deviation, AAD, the Maximum Deviation (MD), the Average Deviation (Bias), and the standard deviation σ which are defined as follows

$$\mathrm{AAD} = \frac{100}{N}\sum_{i=1}^{N}\left|\frac{\rho_i^{\mathrm{exp}} - \rho_i^{\mathrm{calc}}}{\rho_i^{\mathrm{exp}}}\right| \tag{1}$$

$$\mathrm{MD} = \mathrm{Max}\left(100\left|\frac{\rho_i^{\mathrm{exp}} - \rho_i^{\mathrm{calc}}}{\rho_i^{\mathrm{exp}}}\right|\right) \tag{2}$$

$$\mathrm{Bias} = \frac{100}{N}\sum_{i=1}^{N}\frac{\rho_i^{\mathrm{exp}} - \rho_i^{\mathrm{calc}}}{\rho_i^{\mathrm{exp}}} \tag{3}$$

$$\sigma = \sqrt{\frac{\sum_{i=1}^{N}(\rho_i^{\mathrm{exp}} - \rho_i^{\mathrm{calc}})^2}{N - m}} \tag{4}$$

where N is the number of experimental data ($N = 90$ for all our data) and m is the number of parameters (we will see below that with our correlation $m = 8$).

Tait Representation of DBE Density. To correlate accurately our values over our entire temperature and pressure ranges (and consequently to obtain a correct evaluation of the derived properties), we use the following Tait-like equation, which has been used in these previous works.[6,7,13−19]

$$\rho(T, p) = \frac{\rho_0(T)}{1 - C\ln\left(\dfrac{B(T) + p}{B(T) + 0.1\ \mathrm{MPa}}\right)} \tag{5}$$

with

$$\rho_0(T) = A_0 + A_1T + A_2T^2 + A_3T^3 \tag{6}$$

$$B(T) = B_0 + B_1T + B_2T^2 \tag{7}$$

We mention here that the A_i, B_i, and C parameter values were determined by correlating simultaneously all the experimental density values versus pressure and temperature. The Tait-correlation parameters, along with the AAD (0.0194 %), MD (0.0620 %), Bias ($0.837 \cdot 10^{-4}$ %), and standard deviation, σ ($2.014 \cdot 10^{-4}$ $g \cdot cm^{-3}$), obtained with this correlation are given in Table 2. The maximum deviation occurs at T = 333.15 K and p = 90 MPa. Notice that the AAD and the standard deviation are lower than the experimental uncertainty. Consequently these equations make it possible to interpolate the density at any T, p conditions. Note that if using $\rho_0(T) = A_0 + A_1T + A_2T^2$ instead of eq 6, then AAD = 0.032 %, MD = 0.074 %, Bias = −0.024 %, and σ = $3.05 \cdot 10^{-4}$ $g \cdot cm^{-3}$. So, as the AAD and the standard deviation are a little better with eq 6, we have used in the following this three-degree polynomial representation. Figure 1 shows the deviation plots between the measured and calculated densities with eqs 5 to 7 and also a comparison with some literature data.[5] Figure 2 shows the variation of density as a function of temperature at p = (0.1, 70, and 140) MPa. As usual, density decreases when temperature increases. This figure shows that as the temperature interval considered here is sufficiently large the density versus temperature is nonlinear (more particularly at low pressure) which justifies the use of eq 6. To get a good estimation of some derivative properties (see paragraph below), density at atmospheric pressure has been represented using eq 6 which is nonlinear because the derivative properties are sensitive to the mathematical expression used to evaluate the atmospheric pressure density versus temperature. Figure 3 shows the variation of density as a function of pressure at T = (293.15, 333.15, and 393.15) K. As usual, density increases when pressure increases. At constant temperature the curves are concave, which is associated with a negative second-order derivative. The shape of the isothermal curves of the density versus pressure is compatible with the logarithmic relationship used in the Tait-type density relation used to model the influence of pressure on density.

Table 2. Obtained Parameters and Deviations for Density Correlation by Using Equation 5

$A_0/g \cdot cm^{-3}$	0.9395
$A_1/g \cdot cm^{-3} \cdot K^{-1}$	$-2.794 \cdot 10^{-4}$
$A_2/g \cdot cm^{-3} \cdot K^{-2}$	$-1.221 \cdot 10^{-6}$
$A_3/g \cdot .cm^{-3} \cdot K^{-3}$	$6.054 \cdot 10^{-10}$
B_0/MPa	348.7
$B_1/MPa \cdot K^{-1}$	−1.282
$B_2/MPa \cdot K^{-2}$	$1.221 \cdot 10^{-3}$
C	0.08861
$\sigma/g \cdot cm^{-3}$	$2.014 \cdot 10^{-4}$
AAD/%	0.0194
MD/%	0.0620
Bias/%	$0.837 \cdot 10^{-4}$

Comparison with Literature Data. Certain numbers of authors[20−33] have reported experimental density data at atmospheric pressure and at low temperatures. No data were reported at high temperatures and at high pressures, except the data reported by Meng et al.[5] It is difficult to compare our measurements with the results given in the literature mainly because the experimental p, T sets are generally not exactly the same. We have made a comparison with the values generated by our correlation (eqs 5, 6, and 7 and Table 2) at exactly the same experimental p, T sets given for several references for comparable pressure and temperature intervals. For data reported by Meng et al.[5] (70 experimental values; $(0.1 \le p \le 21.12)$ MPa and $(243.15 \le T \le 373)$ K), we get AAD = 0.177 %, MD = 0.44 %, and Bias = 0.177 %. The uncertainty claimed by Meng et al.[5] is ± 0.2 % (in our work the uncertainty is around ± 0.05 %). The deviations of the literature data respect the present results at high pressure and high temperatures and at atmospheric pressure are shown in Figures 1 and 4, respectively. As can be seen from the Figure 4, most of the reported data at 0.1 MPa agree with the present values, except some data by Meng et al.[5] at high temperatures. It can be seen that the majority of deviations is lower than 0.1 %. Excellent agreement within 0.01 % and 0.07 % was found between the present results and the data reported by some authors.[20−24,26−33] Good agreement within 0.1 % is found

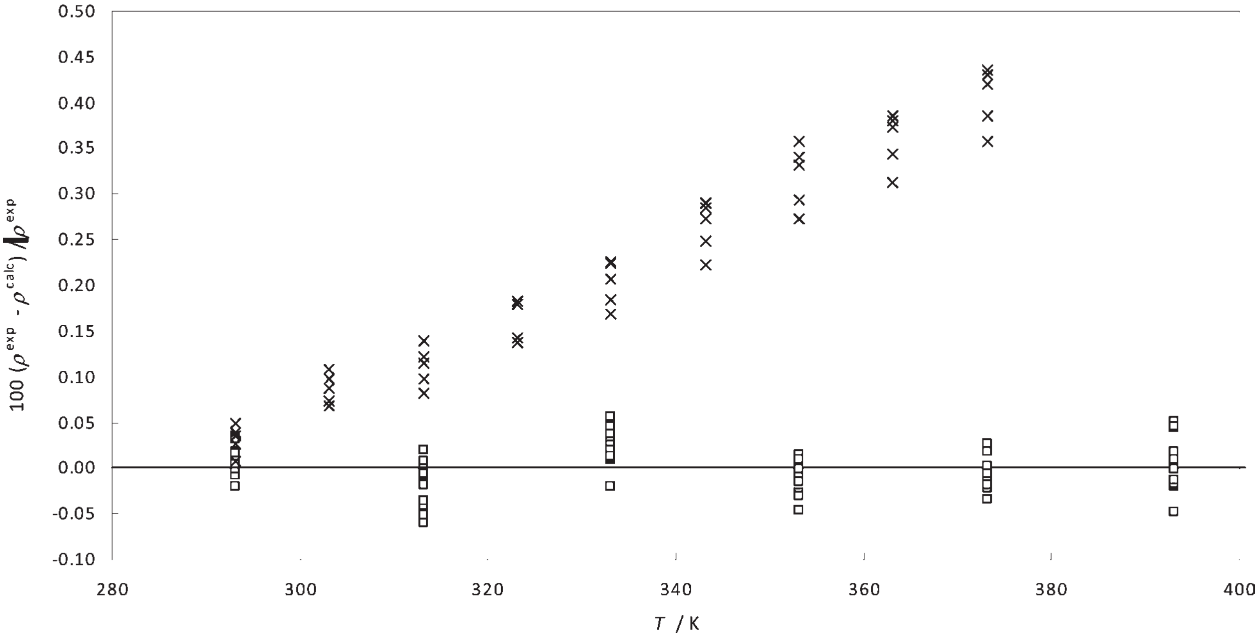

Figure 1. Deviation between the measured and calculated densities with eqs 5 to 7: □, this work; ×, Meng et al.[5]

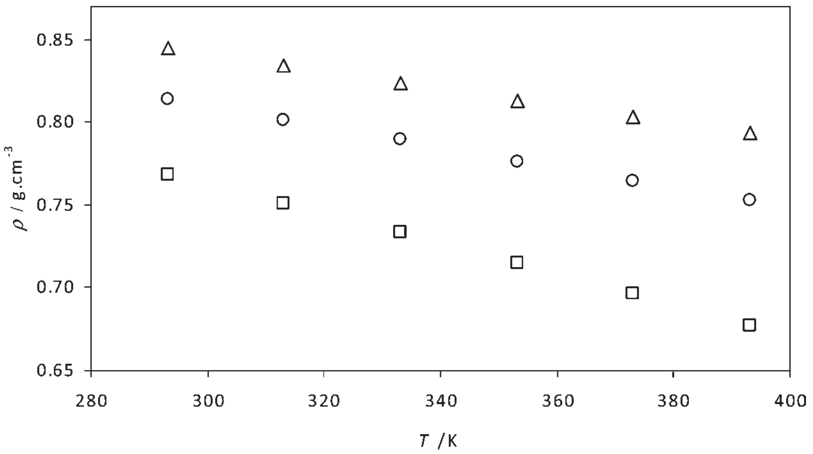

Figure 2. Variation of DBE density as a function of temperature at □, p = 0.1 MPa; ○, p = 70 MPa; and Δ, p = 140 MPa.

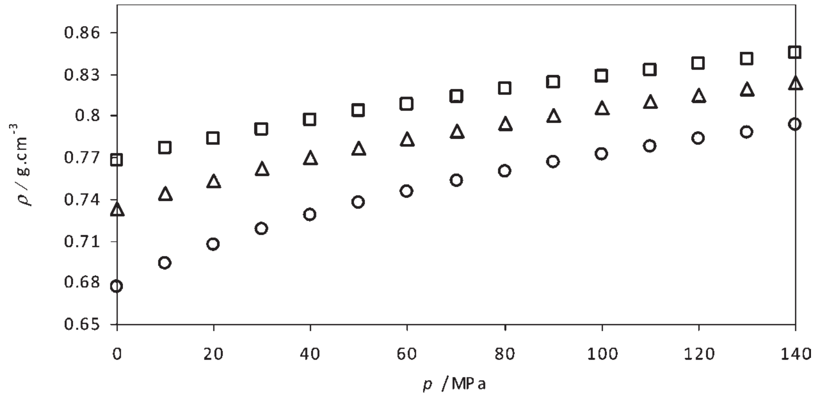

Figure 3. Variation of DBE density as a function of pressure at □, T = 293.15 K; Δ, T = 333.15 K; and ○, T = 393.15 K.

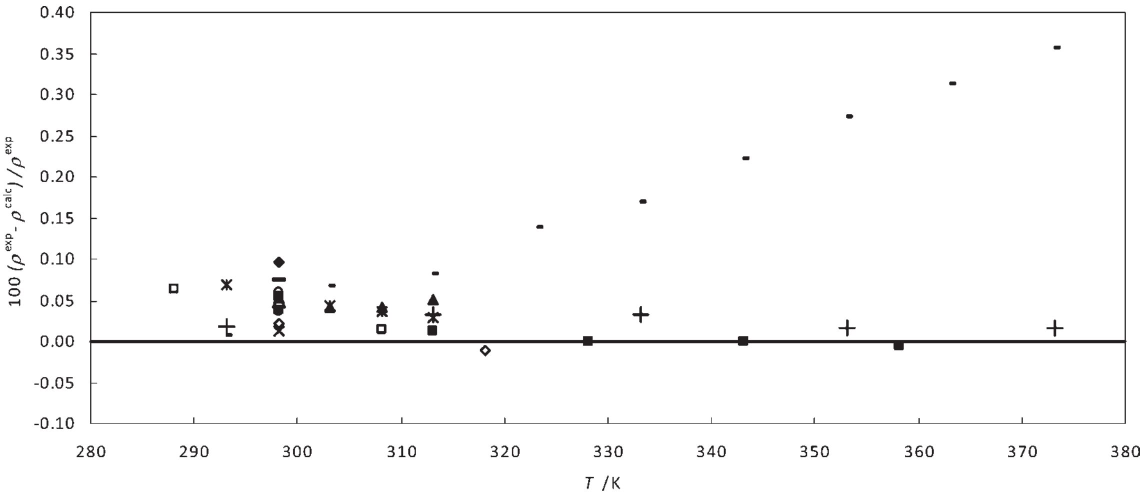

Figure 4. Percentage deviations at 0.1 MPa of the present experimental densities for DBE with respect to the values calculated with eqs 5 to 7: +, this work; -, Meng et al.;[5] *, Monge et al.;[30] ◇, Villa et al.;[20] Δ, Lladosa et al.;[21] ●, Arce et al.;[23] ○, Serna et al.;[24] ♦, Domanska et al.;[25] ×, Tanaka et al.;[27] −, Park et al.;[28] ■, Venkatesu et al.;[31] □, Tovar et al.;[32] ▲, George et al.[33]

between the present data and the values reported by Domańska et al.,[25] while the data by Meng et al.[5] show large deviations between 0.01 % and 0.36 % at atmospheric pressure (Figure 4) and between 0.02 % and 0.44 % at high-pressure levels (Figure 1). Most of the data show systematic positive deviations at atmospheric pressure, except for results published by Villa et al.[20] at 318.15 K and Venkatesu et al.[31] at 358.15 K.

Table 3. Isobaric Thermal Expansivity, α_p, at Various Temperatures and Pressures

	T/K					
p/MPa	293.15	313.15	333.15	353.15	373.15	393.15
			$\alpha_p/10^{-4}\cdot\mathrm{K}^{-1}$			
0.1	11.01	11.56	12.16	12.83	13.58	14.41
10	10.28	10.70	11.09	11.44	11.75	12.01
20	9.80	9.99	10.19	10.39	10.58	10.78
30	9.31	9.36	9.47	9.63	9.85	10.13
40	8.85	8.92	8.97	9.02	9.05	9.08
50	8.48	8.47	8.50	8.57	8.68	8.85
60	8.11	8.11	8.12	8.16	8.22	8.30
70	7.65	7.82	7.91	7.90	7.79	7.58
80	7.34	7.53	7.62	7.60	7.45	7.18
90	6.90	7.20	7.37	7.39	7.26	6.96
100	7.03	7.08	7.09	7.07	7.01	6.91
110	6.73	6.82	6.86	6.83	6.74	6.57
120	6.70	6.72	6.70	6.63	6.53	6.37
130	6.44	6.54	6.55	6.48	6.32	6.06
140	6.23	6.37	6.41	6.35	6.17	5.87

Table 4. Isothermal Compressibility, κ_T, at Various Temperatures and Pressures

	T/K					
p/MPa	293.15	313.15	333.15	353.15	373.15	393.15
			$\kappa_T/10^{-4}\,\mathrm{MPa}^{-1}$			
0.1	11.37	13.21	15.49	18.33	21.91	26.44
10	10.20	11.65	13.39	15.47	17.95	20.89
20	9.25	10.43	11.80	13.39	15.23	17.31
30	8.46	9.45	10.56	11.83	13.25	14.81
40	7.81	8.64	9.57	10.61	11.74	12.97
50	7.25	7.97	8.76	9.62	10.56	11.56
60	6.77	7.40	8.08	8.82	9.60	10.43
70	6.36	6.91	7.50	8.14	8.82	9.52
80	5.99	6.48	7.00	7.56	8.15	8.76
90	5.67	6.10	6.57	7.07	7.59	8.12
100	5.38	5.77	6.19	6.64	7.10	7.57
110	5.12	5.48	5.86	6.27	6.67	7.09
120	4.88	5.21	5.56	5.92	6.29	6.68
130	4.67	4.97	5.29	5.62	5.96	6.31
140	4.48	4.76	5.05	5.35	5.66	5.98

Derived Thermodynamic Properties. The temperature and pressure dependence of the isothermal compressibility, κ_T, is obtained from eq 5 and is expressed as follows

$$\kappa_T = \left(\frac{1}{\rho}\right)\left(\frac{\partial\rho}{\partial p}\right)_T = \frac{C}{\left(1 - C\ln\left(\frac{B(T)+p}{B(T)+0.1\ \mathrm{MPa}}\right)\right)(B(T)+p)} \qquad (8)$$

Similarly, the isobaric thermal expansivity could also be obtained analytically by differentiating eq 5 taking into account the temperature dependence of $\rho_0(T)$ and $B(T)$. However, as Cerdeiriña et al.[34] and Troncoso et al.[35] mention, the estimated isobaric thermal expansivity depends on the form of functions $B(T)$ and $\rho_0(T)$. This is the reason why they recommend to derive the isobaric thermal expansivity from the isobaric densities. So, at each pressure, we suppose that $\rho_p(T) = a_0 + a_1T + a_2T^2 + a_3T^3$ and consequently $(\partial\rho/\partial T)_p = a_1 + 2a_2T + 3a_3T^2$. For each pressure, we get a set (a_0, a_1, a_2, a_3). In such a way, for density AAD = 0.014 %, Bias = $-3.8\cdot10^{-6}$ %, and MD = 0.047 %. By inserting the differentiated density and the calculated densities $\rho_p(T)$ into $\rho_p = -1/\rho(\partial\rho/\partial T)_p$, the isobaric thermal expansivity at the different T, p conditions has been derived

$$\alpha_p = -\frac{a_1 + 2a_2T^2 + 3a_3T^2}{a_0 + a_1T + a_2T^2 + a_3T^3} \qquad (9)$$

As already pointed out, the method used to evaluate the isobaric thermal expansion coefficient may affect the accuracy of the values. It has to be stated (Jacquemin et al.[36]) that the differences sometimes found for the values of this coefficient from the literature are due not only to differences in density values but also to the fitting equations.

The isobaric thermal expansivity, α_p, and the isothermal compressibility, κ_T, were calculated from the above procedures. The observed average difference between our calculated values for α_p and those calculated directly by differentiating eq 5 is 0.9 %, though there is a maximum absolute deviation of 4.5 %. As recently indicated on similar high-pressure density studies[7,9,13,19,37] with the same methods, the estimated uncertainty is ± 1 % for the isothermal compressibility and around ± 3 % for the isobaric thermal expansivity.

The calculated values of isobaric thermal expansivity, α_p, and the isothermal compressibility, κ_T, are presented in Tables 3 and 4, respectively, for various temperatures and pressures.

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