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Density of Diethyl Adipate using a New Vibrating Tube Densimeter from (293.15 to 403.15) K and up to 140 MPa. Calibration and Measurements

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New density data for diethyl adipate (DEA) over 12 isotherms [(293.15 ≤ T ≤ 403.15) K] and 15 isobars [(0.1 ≤ p ≤ 140) MPa] are reported. This paper presents also the calibration procedure we propose for a new experimental equipment. Data reliability has been verified over the pressure and temperature experimental intervals by comparing our experimental results for toluene and 1-butanol with previous literature data. A total of 732 experimental data points have been measured in the framework of this work. The experimental uncertainty is estimated to be ± 0.5 kg·m⁻³ (around 0.05 %). The pressure and temperature dependencies of diethyl adipate densities were accurately represented by the Tammann–Tait equation with standard deviations of 0.3 kg·m⁻³. These data were used to analyze the isothermal compressibility and the isobaric thermal expansivity for this fluid.

Introduction

Modern lubricants are complex formulations, which are continually developed to meet increasing requirements for performance and durability.¹ In the scope of investigating more benign lubricants as potential replacements of oils presently used, we have focused our attention on the diesters (adipates), which have excellent properties for industrial applications. Diesters are obtained by reacting a linear diacid, usually a branched one, with a monofunctional alcohol. They have very good viscosity index (VI) and pour points.¹ The linear diacid portion of the diester contributes to the good VI, while the branched alcohol ends give the lubricant a good pour point. One disadvantage of diesters is their low molecular weight, and this results in a limited viscosity range (VG) coverage [(7 to 46) mPa·s]. Phthalates and dimerates are special subgroups of diesters which have higher viscosities.¹ The small size of the diesters combined with their high polarities makes them effective solvents (they present an attractive alternative to replace chlorinated solvents). They have, for a long time, been used as the primary base fluid in the formulation of low viscosity and of highly oxidatively stable ISO 46 environmentally acceptable lubricants.² Diesters are often blended with poly- α -olefins (PAOs) to help additive solubility and to act as seal-swelling agents.

Their applications include gear oils, engine oils, compressors oils, grease, metalworking, and biodegradable hydraulic fluids. Some diesters are frequently used as plastiziers especially phthalates and adipates: di-2-ethylhexyl (DEHP) and diisononyl phthalates (DINP) and di-2-ethylhexyl adipate (DEHA).³ Diesters are also used as a base oil or part of a base oil for automotive engine oils and in some low temperature greases. Recently, it was found⁴ that oxygenated additives such as diethyl adipate (DEA) could provide great improvement in fuel proper-

ties and reduce engine emissions, thus diethyl adipate is regarded as a good fuel additive and a potential alternative fuel in the future.

For the industry applications of diesters, thermophysical properties are indispensable. Nevertheless, only a few studies about the density of diesters have been done at atmospheric pressure.^{3,5–11} These studies are limited to a very small temperature interval, and they never study the effect of pressure on the density of these fluids. To get some information about this topic which could be interesting in industrial purposes, in the present work, density, isothermal compressibilities, $\kappa_T = (1/\rho)(\partial\rho/\partial p)_T$, and thermal isobaric expansivities, $\alpha_p = -(1/\rho)(\partial\rho/\partial T)_p$ of diethyl adipate (DEA) have been studied from (293.15 to 403.15) K and up to 140 MPa.

Experimental

Chemicals. Toluene, 1-butanol, and decane were obtained from Riedel de Haen (puriss > 99.9 %, with a certificate of analysis of 99.98 %), from Fluka (puriss 99.5 % with a certificate of analysis of 99.9%), and from Merck (>99 % with a certificate of analysis of 99.8 %), respectively. Diethyl adipate (CAS number 141-28-6, C₁₀H₁₈O₄, molar mass = 202.25 g·mol⁻¹) was obtained from Aldrich (puriss 99 % with a certificate of analysis of 99.9 %), and water was purified using a Milli-Q Plus system.

Experimental Apparatus. The density ρ as a function of pressure and temperature was measured with a high pressure vibrating tube densimeter Anton Paar DMA HPM which is adequate to perform measurements up to 140 MPa. In our case, the apparatus was started up as indicated in Figure 1 to perform measurements up to 403.15 K. It should be pointed out that we have not found any density data available in the literature measured with this apparatus, except some measurements that we made¹² about amines up to 353.15 K.

The DMA HPM has been connected to a mPDS 2000V3 evaluation unit which reports the vibration period with seven significant digits. The temperature of the vibrating tube cell is

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controlled through a thermostatic bath Julabo Polystat 36 and measured inside the cell block with an AOIP PN 5207 thermometer with an uncertainty of ± 0.05 K. The temperature was kept constant within ± 0.01 K. The pressure of the system was applied with a piston pressure intensifier (Top Industries) and measured on an absolute manometer HBM PE 200/2000 with an uncertainty of ± 0.1 % of full scale 0.2 MPa. The pressure was kept constant to show digits do not change during the time of the measurement.

Before loading each sample, the densimeter and all tubing were washed with a volatile liquid, such as petroleum ether, to ensure removal of any residue from previous runs. Then, gaseous nitrogen was passed throughout the system. Once this cleaning process was completed, a vacuum was applied to the system. Then, valve V_1 was opened, introducing the sample. When thermal equilibrium was reached, the vibration period of the cell was determined at different pressures, starting at 0.1 MPa, followed by the higher pressures. Then the temperature of the liquid bath was changed, and a new isotherm was started.

Calibration Procedure. One of the most important limitations in using vibrating tube densimeters is to relate the raw data (period of vibration) to the density ρ , i.e., the calibration curve expressed as the apparatus parameters. The density is related to the vibration period, τ , by the equation

$$\rho(T, p) = A(T, p)\tau^2(T, p) - B(T, p) \quad (1)$$

To determine the density of a sample, it is necessary to measure the oscillation period over the T, p range of interest and to know the two characteristic parameters of the apparatus: $A(T, p)$ and $B(T, p)$. These parameters can be determined by measuring the periods for two substances of “well-known” density, but it is difficult to find precise values for two reference substances over all the T, p sets of the experimental domain (in the case of the present work: $293.15 \leq T \leq 403.15$ K and $0.1 \leq p \leq 140$ MPa).

This is the reason why Lagourette et al.¹³ have proposed a calibration process that is widely used in the literature at pressures up to 70 MPa.^{14–27} The hypotheses of Lagourette et al. are that the first parameter depends only on the temperature, $A(T)$, and the second one on both temperature and pressure, $B(T, p)$. Following these facts, and taking into account that for a vacuum it can be considered $\rho_{\text{vacuum}}(T) = 0 \text{ g}\cdot\text{cm}^{-3}$, eq 1 can be written for a vacuum as follows

$$0 = A(T)\tau_{\text{vacuum}}^2(T) - B(T, 0) \quad (2)$$

where $\tau_{\text{vacuum}}(T)$ is the measured period of the evacuated cell at each temperature. From this equation, $A(T)$ can be obtained as

$$A(T) = \frac{B(T, 0)}{\tau_{\text{vacuum}}^2(T)} \quad (3)$$

and eq 1 can be rewritten as follows

$$\rho(T, p) = \frac{B(T, 0)}{\tau_{\text{vacuum}}^2(T)} \tau^2(T, p) - B(T, p) \quad (4)$$

On the other hand, it is helpful to choose water as a reference substance, for which the density is accurately known at all T, p conditions.^{28,29} Thus, Lagourette et al. have obtained the parameter $B(T, p)$ from eq 4 in the following way

$$B(T, p) = -\rho_{\text{water}}(T, p) + \frac{B(T, 0)}{\tau_{\text{vacuum}}^2(T)} \tau_{\text{water}}^2(T, p) \quad (5)$$

where $\tau_{\text{water}}(T, p)$ is the period of the cell full of water, at each pressure and temperature, and $\rho_{\text{water}}(T, p)$ is the density of water at each temperature and pressure taken from Wagner and Pruss.²⁹ Thus, taking into account the evacuated cell and considering water as the reference substance, i.e., eqs 3 and 5, Lagourette et al. have written eq 1 as follows

$$\rho(T, p) = \rho_{\text{water}}(T, p) + B(T, 0) \left[\frac{\tau^2(T, p) - \tau_{\text{water}}^2(T, p)}{\tau_{\text{vacuum}}^2(T)} \right] \quad (6)$$

The remaining parameter to be determined is $B(T, 0)$. Following Lagourette et al., $B(T, p) = M_0 / V_u(T, p)$ where M_0 is the mass of the empty vibrating cell and V_u the volume of the cell. When the pressure changes, M_0 is constant, but V_u is a function of pressure and temperature. In a first approximation, one can write $V_u(T, p) = V_{u0}(T)(1 + \epsilon p)$ where $V_{u0}(T)$ is the volume of the empty cell at temperature T and ϵ is the expansion pressure coefficient of the cell. As ϵ is very small, it is possible to make the assumption that $V_u(T, 0.1 \text{ MPa}) \approx V_{u0}(T)$ and consequently $B(T, 0) \approx B(T, 0.1 \text{ MPa})$. Thus, using eq 4 written for another reference fluid at 0.1 MPa, the following expression is found

$$\rho_{\text{reference}}(T, 0.1 \text{ MPa}) = B(T, 0.1 \text{ MPa}) \cdot \left[\frac{\tau_{\text{reference}}^2(T, 0.1 \text{ MPa}) - \tau_{\text{vacuum}}^2(T)}{\tau_{\text{vacuum}}^2(T)} \right] \quad (7)$$

So, from this equation, the parameter $B(T, 0) \approx B(T, 0.1 \text{ MPa})$ is obtained

$$B(T, 0) = \rho_{\text{reference}}(T, 0.1 \text{ MPa}) \cdot \left[\frac{\tau_{\text{vacuum}}^2(T)}{\tau_{\text{reference}}^2(T, 0.1 \text{ MPa}) - \tau_{\text{vacuum}}^2(T)} \right] \quad (8)$$

In this equation “reference” denotes a fluid with a “well-known” density over the temperature interval of interest and at 0.1 MPa. Taking into account eq 8, eq 6 can be written as

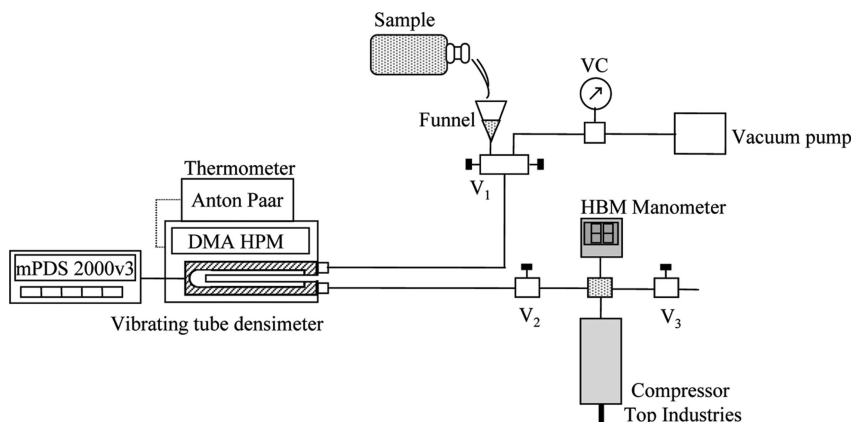


Figure 1. Schematic diagram of the $\rho p T$ apparatus.

$$\rho(T, p) = \rho_{\text{water}}(T, p) + \rho_{\text{reference}}(T, 0.1 \text{ MPa}) \cdot \left[\frac{\tau^2(T, p) - \tau_{\text{water}}^2(T, p)}{\tau_{\text{reference}}^2(T, 0.1 \text{ MPa}) - \tau_{\text{vacuum}}^2(T)} \right] \quad (9)$$

Lagourette et al.¹³ use also water to evaluate eq 8, thus it is only necessary to know the period of vacuum and water and the density of water over all the (T, p) studied ranges. Lagourette et al. write finally eq 9 as

$$\rho(T, p) = \rho_{\text{water}}(T, p) + \rho_{\text{water}}(T, 0.1 \text{ MPa}) \cdot \left[\frac{\tau^2(T, p) - \tau_{\text{water}}^2(T, p)}{\tau_{\text{water}}^2(T, 0.1 \text{ MPa}) - \tau_{\text{vacuum}}^2(T)} \right] \quad (10)$$

As already indicated, this equation is widely used in the literature (see refs 7 to 20 for recent examples). Concerning the present work, we used the following calibration procedures:

(a) At $(0.1 \leq p \leq 140) \text{ MPa}$ and $(293.15 \leq T \leq 353.15) \text{ K}$. The densimeter calibration over these temperature and pressure intervals has been performed by using eq 10. The density values of water have been taken from the equation of state (EoS) reported by Wagner and Pruss.²⁹ 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 K, and 0.003 % at pressures in the interval (10 to 100) MPa and temperatures up to 423 K. The uncertainty is of the order of 0.02 % at 1000 MPa.

(b) At $p = 0.1 \text{ MPa}$ and $T \geq 373.15 \text{ K}$. At atmospheric pressure and at temperatures greater or equal to the boiling point of water, the limitation of this procedure appears as water is in the gaseous state. The terms $\rho_{\text{water}}(T, 0.1 \text{ MPa})$ and $\tau_{\text{water}}^2(T, 0.1 \text{ MPa})$ do not correspond to the dense water status anymore. So, in this work, another reference substance has been used at the conditions $p = 0.1 \text{ MPa}$ and $T \geq 363.15 \text{ K}$. Decane has been selected because its density is well-known at atmospheric pressure over wide temperature intervals.³⁰ In this case, eqs 5 to 7 are obtained by using decane instead of water, so eq 10 is replaced by the following expression

$$\rho(T, 0.1 \text{ MPa}) = \rho_{\text{decane}}(T, 0.1 \text{ MPa}) \cdot \left[1 + \frac{\tau^2(T, 0.1 \text{ MPa}) - \tau_{\text{decane}}^2(T, 0.1 \text{ MPa})}{\tau_{\text{decane}}^2(T, 0.1 \text{ MPa}) - \tau_{\text{vacuum}}^2(T)} \right] \quad (11)$$

(c) At $p > 0.1 \text{ MPa}$ and $T \geq 373.15 \text{ K}$. Over these T, p conditions, it is also not possible to use eq 10 for the same reason as in the previous paragraph; in this work, another equation must be considered. There are two possibilities.

The first one is to use decane to calculate $B(T, 0)$ in eq 8. In this case, eq 10 is replaced by

$$\rho(T, p) = \rho_{\text{water}}(T, p) + \rho_{\text{decane}}(T, 0.1 \text{ MPa}) \cdot \left[\frac{\tau^2(T, p) - \tau_{\text{water}}^2(T, p)}{\tau_{\text{decane}}^2(T, 0.1 \text{ MPa}) - \tau_{\text{vacuum}}^2(T)} \right] \quad (12)$$

So, in this case, it is necessary to know at each $p > 0.1 \text{ MPa}$ and for each temperature the period of the cell full of water and the density of water. It is also necessary to know the oscillation period of the evacuated cell for each temperature. Finally, it is necessary to know at atmospheric pressure, for each temperature, the period of the cell full of decane and the density of decane.

The second possibility is to assume the approximation $B(T, 0) \approx B(T, 1 \text{ MPa})$, instead of $B(T, 0) \approx B(T, 0.1 \text{ MPa})$, in a similar way to Lagourette et al.¹³ We 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. In this case, the following equation is obtained instead of eq 10

$$\rho(T, p) = \rho_{\text{water}}(T, p) + \rho_{\text{water}}(T, 1 \text{ MPa}) \cdot \left[\frac{\tau^2(T, p) - \tau_{\text{water}}^2(T, p)}{\tau_{\text{water}}^2(T, 1 \text{ MPa}) - \tau_{\text{vacuum}}^2(T)} \right] \quad (13)$$

Following this procedure, 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 $p > 0.1 \text{ MPa}$ and for $T \geq 373.15 \text{ K}$). By using eq 13 over the above T, p intervals ($p > 0.1 \text{ MPa}$ and $T \geq 373.15 \text{ K}$), only one reference fluid is necessary (water) together with the period of the evacuated cell.

In this work, both procedures have been used to perform the calibration of the densimeter. The measurements of the period of vacuum as a function of the temperature have been performed after having created an adequate vacuum. The differences generated by using eq 12 or eq 13 are discussed in the next section. The experimental device (mPDS 2000V3 evaluation unit) displays the oscillating period with seven digits, which, according to the specification of Anton-Paar, corresponds to an uncertainty of the order of $10^{-5} \text{ kg} \cdot \text{m}^{-3}$ related only to the measured oscillating period. Nevertheless, taking into account the uncertainties of the temperature, the pressure, the period of oscillation measurements for water, vacuum, and the studied liquid (seven-digit frequency counter) and density accuracy of the reference fluids (water and/or decane), the overall experimental uncertainty in the reported density values, by applying the law of propagating errors, is estimated to be $\pm 0.5 \text{ kg m}^{-3}$, i.e., 0.05 %. This uncertainty is similar to that reported by other authors^{31–34} with other Anton Paar densimeters.

It should be mentioned that under certain circumstances (viscous fluids) the viscosity of the fluid can affect the density measured by a vibrating tube densimeter, and a correction of the measured density value is required.^{17,35,36} Several authors^{37,38} obtained the relative density increase, named correction factor, $\Delta\rho$, due to the sample viscosity for the vibrating tube densimeter, but only for equipment at atmospheric pressure. From their results, it is concluded that the correction factor depends not only on the sample viscosity but also on the specific design of the vibrating tube. In the case of our apparatus (DMA HPM), Anton Paar³⁹ provided us a correction curve. From this curve, it can be observed that for samples with a dynamic viscosity of $25 \text{ mPa} \cdot \text{s}$ and a density around $1000 \text{ kg} \cdot \text{m}^{-3}$ the density correction would be around $0.19 \text{ kg} \cdot \text{m}^{-3}$. For the considered fluids in this work (water, toluene, decane, 1-butanol, and DEA), the maximum viscosity data are reached at 293.15 K and 140 MPa. At these T, p conditions, the viscosity of toluene is $1.7 \text{ mPa} \cdot \text{s}$.⁴⁰ For 1-butanol, we have found in the literature that the viscosity value at 298.15 K and 50 MPa is $3.9 \text{ mPa} \cdot \text{s}$.⁴¹ In the case of diethyl adipate (DEA), there are no dynamic viscosity data published in the literature at pressures higher than the atmospheric one, and the only viscosity is known at 293.15 K and 0.1 MPa ($\eta \approx 3.4 \text{ mPa} \cdot \text{s}$). This value is similar to the one of 1-butanol (at 0.1 MPa and 293.15 K, we have $2.97 \text{ mPa} \cdot \text{s}$ ⁴² for 1-butanol). Hence, for the three fluids considered in this work, it can be accepted that the viscosity would be lower than $25 \text{ mPa} \cdot \text{s}$ over the entire T, p interval studied in the present work. So, the density correction will be much lower than $0.19 \text{ kg} \cdot \text{m}^{-3}$. So, since the correction is lower than the uncertainty consequently, correction on the measured densities, due to viscosity, was not considered.

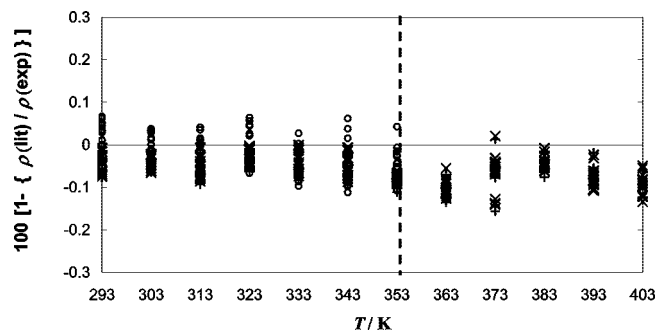


Figure 2. Comparison between experimental data (obtained using eqs 10, 11, and 13) and those previously reported for 1-butanol and for toluene against temperature: ○, 1-butanol vs Cibulka and Zikova;^{43,48} ×, toluene vs Cibulka and Takagi;⁴⁴ +, toluene vs Lemmon and Span⁴⁵ (NIST).

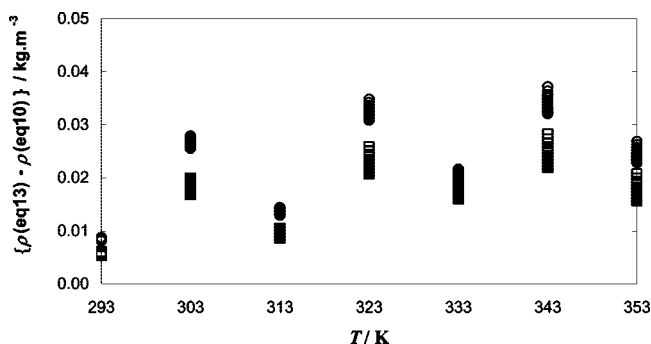


Figure 3. Comparison between experimental data obtained using eqs 10 or 13 over the temperature interval (293.15 ≤ T ≤ 353.15) K against temperature: ○, toluene; □, 1-butanol.

Results and Discussion

Checking of the Calibration Procedure. With the aim to verify the calibration procedure detailed above, the densities of 1-butanol and toluene have been measured over the temperature and pressure intervals (293.15 ≤ T ≤ 403.15) K and (0.1 ≤ p ≤ 140) MPa. Both fluids have been chosen due to the fact that there are reference correlations in the literature^{43–45} that allow us to establish comparisons with the data experimentally obtained in the present work. As in previous works,^{46,47} these comparisons have been performed based on the absolute average percentage deviation (AAD), the percentage maximum deviation (MD), the average percentage deviation (Bias), and the standard deviation which are defined as follows

$$\text{AAD} = \frac{100}{N} \sum_{i=1}^N \left| \frac{\rho_i^{\text{exptl}} - \rho_i^{\text{calcd}}}{\rho_i^{\text{exptl}}} \right| \quad (14)$$

$$\text{MD} = \text{Max} \left(100 \left| \frac{\rho_i^{\text{exptl}} - \rho_i^{\text{calcd}}}{\rho_i^{\text{exptl}}} \right| \right) \quad (15)$$

$$\text{Bias} = \frac{100}{N} \sum_{i=1}^N \frac{\rho_i^{\text{exptl}} - \rho_i^{\text{calcd}}}{\rho_i^{\text{exptl}}} \quad (16)$$

$$\sigma = \sqrt{\frac{\sum_{i=1}^N (\rho_i^{\text{exptl}} - \rho_i^{\text{calcd}})^2}{N - m}} \quad (17)$$

where N is the number of experimental data; ρ_i^{exptl} is the experimental density; ρ_i^{calcd} is the density values obtained from the correlation equations; and m is the number of parameters involved in the correlation equation.

Comparison with 1-Butanol. In the case of 1-butanol, the data experimentally determined in the present work have been compared with the correlation equation proposed by Cibulka and Zikova.^{43,48} The equation proposed by these authors has been obtained by using literature data of 1-butanol from different sources over the temperature interval (194.61 to 523.88) K and for pressures between (0.2 and 1863.3) MPa. The equation reproduces the literature data with a root-mean-square deviation of 0.5 kg·m⁻³ which corresponds to 0.05 %. The data we obtained in the ranges (293.15 ≤ T ≤ 353.15) K and (0.1 ≤ p ≤ 50) MPa agree with those obtained from the Cibulka and Zikova correlation (at the same T , p conditions) with an AAD of 0.04 %, an MD of 0.11 %, and a Bias of -0.005 %. In the range (293.15 ≤ T ≤ 353.15) K and from (0.1 ≤ p ≤ 140) MPa, the deviations are AAD = 0.04 %, MD = 0.11 %, and Bias = -0.02 %. When the comparison is performed over the entire temperature interval (up to 403.15 K, using eqs 10, 11, and 12 to perform the calibration) the deviations are slightly higher (AAD = 0.08 %, MD = 0.27 %, and Bias = -0.06 %), but it is necessary to take into account that the database used by Cibulka and Zikova contains, for temperatures higher than 360 K, only points up to 50 MPa; furthermore, the number of points is very scarce for high temperatures and high pressures.⁴³ When the calibration is performed with eqs 10, 11, and 13, the results are AAD = 0.07 %, MD = 0.21 %, and Bias = -0.05 %, respectively.

Table 1. Experimental Densities, $\rho/\text{kg}\cdot\text{m}^{-3}$, for Diethyl Adipate at Different Pressures and Temperatures

| p | T/K | | | | | | | | | | | |
|-----------------|--------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|
| MPa | 293.15 | 303.15 | 313.15 | 323.15 | 333.15 | 343.15 | 353.15 | 363.15 | 373.15 | 383.15 | 393.15 | 403.15 |
| Diethyl Adipate | | | | | | | | | | | | |
| 0.1 | 1006.6 | 997.0 | 987.7 | 978.3 | 969.0 | 959.8 | 949.7 | 940.2 | 930.9 | 921.3 | 911.6 | 902.0 |
| 10 | 1013.1 | 1003.9 | 994.9 | 986.2 | 977.1 | 968.2 | 958.7 | 949.9 | 940.9 | 932.0 | 922.1 | 912.7 |
| 20 | 1019.4 | 1010.4 | 1001.8 | 993.4 | 984.8 | 976.1 | 967.0 | 958.6 | 950.1 | 941.7 | 932.2 | 923.3 |
| 30 | 1025.3 | 1016.6 | 1008.3 | 1000.1 | 991.7 | 983.3 | 974.8 | 966.6 | 958.8 | 950.2 | 941.4 | 932.2 |
| 40 | 1030.9 | 1022.5 | 1014.3 | 1006.4 | 998.3 | 990.3 | 982.1 | 974.2 | 966.7 | 958.5 | 949.8 | 941.5 |
| 50 | 1036.2 | 1028.0 | 1020.1 | 1012.4 | 1004.6 | 996.7 | 989.0 | 981.3 | 973.6 | 966.0 | 957.1 | 949.8 |
| 60 | 1041.2 | 1033.3 | 1025.6 | 1018.0 | 1010.4 | 1002.9 | 995.4 | 987.7 | 980.3 | 973.0 | 964.3 | 957.2 |
| 70 | 1046.0 | 1038.3 | 1030.8 | 1023.4 | 1016.0 | 1008.7 | 1001.2 | 994.0 | 986.7 | 979.6 | 971.5 | 964.4 |
| 80 | 1050.8 | 1043.2 | 1035.9 | 1028.6 | 1021.3 | 1014.3 | 1006.9 | 999.9 | 992.8 | 986.0 | 978.0 | 971.0 |
| 90 | 1055.2 | 1047.8 | 1040.7 | 1033.6 | 1026.4 | 1019.5 | 1012.4 | 1005.4 | 998.7 | 991.9 | 984.2 | 977.3 |
| 100 | 1059.6 | 1052.3 | 1045.1 | 1038.3 | 1031.3 | 1024.6 | 1017.4 | 1010.8 | 1004.1 | 997.6 | 990.1 | 983.3 |
| 110 | 1063.8 | 1056.5 | 1049.5 | 1042.8 | 1035.9 | 1029.5 | 1022.3 | 1015.9 | 1009.3 | 1003.0 | 995.7 | 989.0 |
| 120 | 1067.9 | 1060.9 | 1053.9 | 1047.4 | 1040.4 | 1034.2 | 1026.7 | 1021.0 | 1014.3 | 1008.3 | 1000.8 | 994.6 |
| 130 | 1071.8 | 1064.8 | 1058.1 | 1051.6 | 1045.0 | 1038.5 | 1031.4 | 1025.8 | 1019.3 | 1013.1 | 1006.1 | 999.7 |
| 140 | 1075.7 | 1068.9 | 1062.1 | 1055.8 | 1049.3 | 1042.9 | 1036.2 | 1030.4 | 1023.9 | 1017.8 | 1011.0 | 1004.7 |

Table 2. Coefficients and Results Obtained for the Tamman–Tait Equation Correlation (Equation 18) for Diethyl Adipate

| parameter values | |
|--|----------|
| A_0 , $\text{kg} \cdot \text{m}^{-3}$ | 1268.554 |
| A_1 , $\text{kg} \cdot \text{m}^{-3} \cdot \text{K}^{-1}$ | −0.8531 |
| $10^3 A_2$, $\text{kg} \cdot \text{m}^{-3} \cdot \text{K}^{-2}$ | −0.13911 |
| B_0 , MPa | 551.817 |
| B_1 , $\text{MPa} \cdot \text{K}^{-1}$ | −2.02917 |
| $10^3 B_2$, $\text{MPa} \cdot \text{K}^{-2}$ | 2.03826 |
| C | 0.08884 |
| AAD, % | 0.02 |
| DM, % | 0.09 |
| Bias, % | −0.001 |
| σ , $\text{kg} \cdot \text{m}^{-3}$ | 0.3 |

Comparison with Toluene. The data experimentally determined in the present work for toluene have been compared with the correlation equation proposed by Cibulka and Takagi⁴⁴ and with the equation of state used by the NIST (Lemmon and Span⁴⁵). The equation proposed by Cibulka and Takagi⁴⁴ has been obtained by using literature data of toluene from different sources over the temperature interval (179.02 to 583.18) K and pressures from (0.13 to 379.20) MPa. The equation reproduces the literature data with a root-mean-square deviation of $0.4 \text{ kg} \cdot \text{m}^{-3}$ which corresponds to 0.05 %. The data we obtained during this work in the range ($293.15 \leq T \leq 353.15$) K and from ($0.1 \leq p \leq 50$) MPa agree with those obtained from the Cibulka and Takagi correlation (at the same T , p conditions) with an AAD of 0.04 %, an MD of 0.09 %, and a Bias of −0.04 %. In the same temperature but up to 140 MPa, the deviations are AAD = 0.05 %, MD = 0.09 %, and Bias = −0.05 %. When the comparison is performed over the entire temperature interval (up to 403.15 K, using eqs 10, 11, and 12 to perform the calibration), the deviations are slightly higher (AAD = 0.06 %, MD = 0.14 %, and Bias = −0.06 %). When the calibration is performed with eqs 10, 11, and 12, these values are AAD = 0.07 %, MD = 0.18 %, and Bias = −0.07 %, respectively.

The National Institute of Standards and Technology (NIST) utilizes, for toluene, the equation of state recently proposed by Lemmon and Span.⁴⁵ This equation represents the densities of this fluid with uncertainties of 0.05 % in the liquid phase up to 540 K and 100 MPa, of 0.5 % at pressures from (100 to 500) MPa, and approaches 0.01 % near the atmospheric pressure and around 300 K. The data reported in this work over the temperature and pressure intervals ($293.15 \leq T \leq 403.15$) K and ($0.1 \leq p \leq 140$) MPa agree with those obtained from this equation with an AAD of 0.07 %, an MD of 0.16 %, and a Bias of −0.07 % when eqs 10, 11, and 12 are used. The same results are found when eq 13 was utilized instead of eq 12. When the comparison is performed only up to 353.15 K but over the whole pressure interval, these deviations are much lower, 0.05 %, 0.09 %, and −0.05 %, respectively.

As the deviations found for 1-butanol and for toluene using eqs 10, 11, and 13 are slightly lower than those obtained with eqs 10, 11, and 12, the first group of equations has been used to perform the calibration of the densimeter to study other pure fluids and mixtures. The comparisons realized for 1-butanol and toluene can also be observed in Figure 2 where the percentage relative deviations, $\Delta\rho = 100(\rho_{\text{exptl}} - \rho_{\text{lit}})/\rho_{\text{exptl}}$, between the experimental data (using eqs 10, 11, and 13) of this work and those obtained from the reference correlations^{43–45} have been plotted against temperature. As can be seen in this figure, the agreement is good.

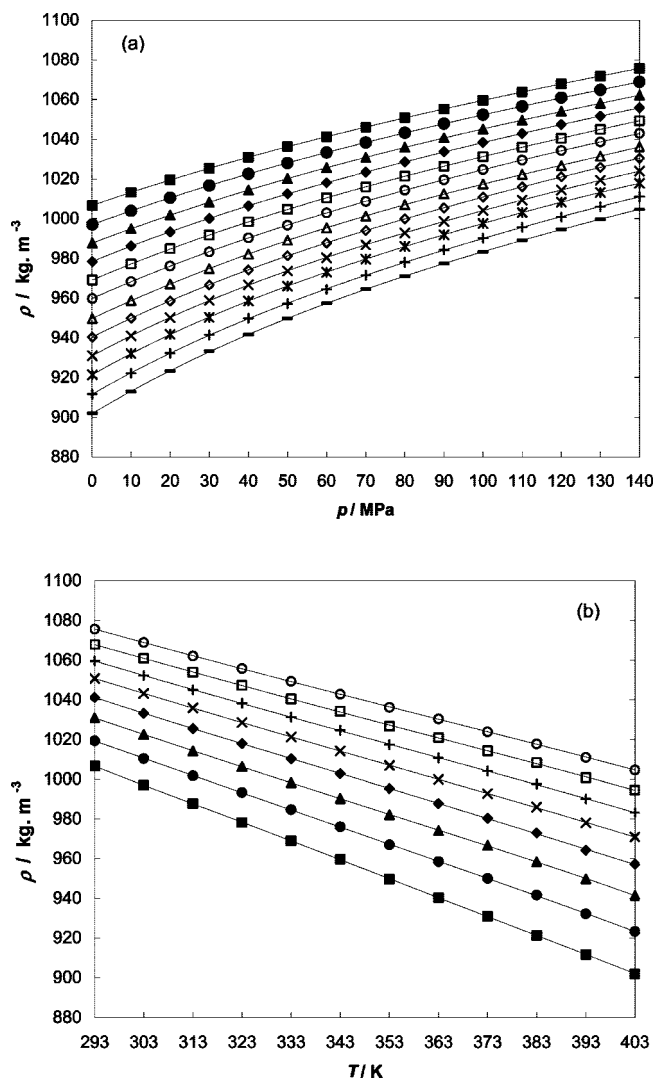


Figure 4. Density for diethyl adipate (a) at different temperatures against pressure: ■, $T = 293.15$ K; ●, $T = 303.15$ K; ▲, $T = 313.15$ K; ◆, $T = 323.15$ K; □, $T = 333.15$ K; ○, $T = 343.15$ K; △, $T = 353.15$ K; ◇, $T = 363.15$ K; ×, $T = 373.15$ K; *, $T = 383.15$ K; +, $T = 393.15$ K; −, $T = 403.15$ K. (b) Density at several pressures vs temperature: ■, $p = 0.1$ MPa; ●, $p = 20$ MPa; ▲, $p = 40$ MPa; ◆, $p = 60$ MPa; ×, $p = 80$ MPa; +, $p = 100$ MPa; □, $p = 120$ MPa; ○, $p = 140$ MPa. —, Correlation line obtained with eq 18.

On the other hand, to see how the approximation $B(T,0) \approx B(T, 1 \text{ MPa})$, instead of $B(T,0) \approx B(T, 0.1 \text{ MPa})$, affects the density values, some comparisons have been established over the intervals ($293.15 \leq T \leq 353.15$) K and ($0.1 \leq p \leq 140$) MPa between the density values obtained for 1-butanol and for toluene with both above approximations (i.e., with eqs 10 and 13). The results are presented in Figure 3. As expected, the density values found with eq 13 are always slightly higher than those found with eq 10. The data obtained with both equations agree within standard deviations of $0.01 \text{ kg} \cdot \text{m}^{-3}$ for 1-butanol and for toluene. As can be seen in Figure 3, the deviations are much lower than the estimated experimental uncertainty ($\pm 0.5 \text{ kg} \cdot \text{m}^{-3}$). Results presented in Figures 2 and 3 confirm the correct operation of the equipment as well as the calibration procedure.

Volumetric Study of Diethyl Adipate. Once the calibration procedure was defined, we studied the volumetric behavior of diethyl adipate (DEA) over the temperature and pressure intervals ($293.15 \leq T \leq 403.15$) K and (0.1 to 140) MPa. The

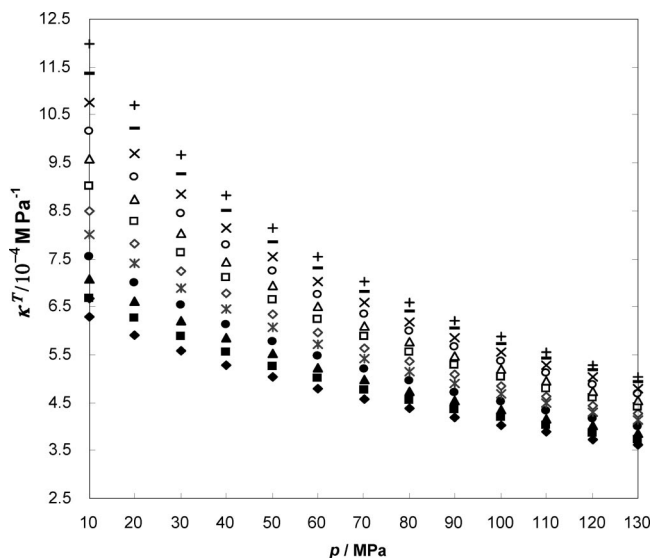


Figure 5. Isothermal compressibilities for diethyl adipate vs pressure at different temperatures: ♦, $T = 293.15$ K; ■, $T = 303.15$ K; ▲, $T = 313.15$ K; ●, $T = 323.15$ K; *, $T = 333.15$ K; ◇, $T = 343.15$ K; □, $T = 353.15$ K; △, $T = 363.15$ K; ○, $T = 373.15$ K; ×, $T = 383.15$ K; −, $T = 393.15$ K; +, $T = 403.15$ K.

density values as a function of temperature and pressure are reported in Table 1. In order to correlate these data over the entire temperature and pressure ranges, the following Tammann–Tait equation⁴⁹ has been used

$$\rho(p, T) = \frac{\rho_0(T)}{1 - C \ln\left(\frac{B(T) + p}{B(T) + 0.1 \text{ MPa}}\right)} \quad (18)$$

where

$$\rho_0(T) = A_0 + A_1 T + A_2 T^2 \quad (19)$$

$$B(T) = B_0 + B_1 T + B_2 T^2 \quad (20)$$

Coefficients A_i ($i = 0, 1, 2$) have been determined in a preliminary fit of the density at 0.1 MPa as a function of the temperature. Coefficients B_j ($j = 0, 1, 2$) and C have been further fitted to the density measurements, at pressures different than 0.1 MPa, using the Levenberg–Marquardt algorithm. The parameter values are presented in Table 2. The comparisons between the experimental points and those obtained from the corresponding equation (eq 18) yield an

AAD of 0.02 %, a Bias of -0.001 %, an MD of 0.09 %, and a standard deviation (σ) of $0.3 \text{ kg} \cdot \text{m}^{-3}$, which is a measure of the dispersion of the data. It must be taken into account that σ is lower than the estimated experimental uncertainty ($\pm 0.5 \text{ kg} \cdot \text{m}^{-3}$). It is interesting to have the parameters of the Tammann–Tait equation because it permits us to interpolate the density of diethyl adipate at temperatures and pressures not exactly the same as those studied here, so it is a useful tool for the industry applications of this diester where thermophysical properties are indispensable.

In Figure 4, the density has been plotted against pressure and against temperature for diethyl adipate. This figure shows also the good agreement between the experimental density data and those obtained from eq 18. Density data for diethyl adipate are available in the literature^{5,6,10,11} but only at 0.1 MPa and for few temperatures. Our data at 293.15 K and 0.1 MPa agree with that reported by Kirbaslar et al.⁶ within a relative absolute deviation of 0.10 %. Ren et al.⁵ have measured the density value of DEA at 301.15 K and 0.1 MPa, and this value presents a relative absolute deviation of 0.003 % with that obtained in this work with eq 18. Chiurdoglu¹⁰ reported density values for this diester at 0.1 MPa and at (273.15, 288.15, 293.15, and 303.15) K, and Serwy¹¹ reported values at the same temperatures except 273.15 K. The data reported in the present work present AADs of 0.11 % and 0.03 % with these authors, respectively.

The temperature and pressure dependence of the isothermal compressibility was calculated according to its definition, $\kappa_T = (1/\rho)(\partial\rho/\partial p)_T$, from analytical differentiation of the Tammann–Tait equation with respect to pressure. From eq 18, the following expression is obtained

$$\kappa_T(T, p) = \frac{C}{(B(T) + p) \left[1 - C \ln\left(\frac{B(T) + p}{B(T) + 0.1 \text{ MPa}}\right) \right]} \quad (21)$$

The values of parameter C and of B_i coefficients for the temperature function $B(T)$ are taken from Table 2. We have estimated by applying the law of propagating errors an uncertainty of 1 % in the isothermal compressibility. This value has also been checked, establishing comparisons over the entire temperature and pressure intervals between the κ_T reported by the NIST⁴⁵ and those obtained if eq 21 is applied to the data of toluene measured in the background of the present work. Thus, an average deviation of 0.6 % is obtained. The isothermal compressibility (κ_T) of diethyl adipate ranges from $3.74 \cdot 10^{-4} \text{ MPa}^{-1}$ to $12.0 \cdot 10^{-4} \text{ MPa}^{-1}$ over the (T, p) interval analyzed in

Table 3. Isobaric Thermal Expansivity and Isothermal Compressibility for Diethyl Adipate Calculated from Equation 22

| p MPa | T/K | | | | | | | | | |
|-------------------------------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|
| | 303.15 | 313.15 | 323.15 | 333.15 | 343.15 | 353.15 | 363.15 | 373.15 | 383.15 | 393.15 |
| $10^4 \alpha_p / (\text{K}^{-1})$ | | | | | | | | | | |
| 10 | 8.98 | 9.06 | 9.15 | 9.24 | 9.33 | 9.43 | 9.53 | 9.63 | 9.74 | 9.86 |
| 30 | 8.28 | 8.32 | 8.36 | 8.41 | 8.46 | 8.51 | 8.57 | 8.65 | 8.74 | 8.85 |
| 50 | 7.72 | 7.74 | 7.75 | 7.78 | 7.80 | 7.84 | 7.88 | 7.95 | 8.03 | 8.15 |
| 70 | 7.27 | 7.27 | 7.27 | 7.27 | 7.29 | 7.31 | 7.35 | 7.41 | 7.50 | 7.62 |
| 90 | 6.89 | 6.88 | 6.87 | 6.87 | 6.87 | 6.89 | 6.93 | 6.99 | 7.08 | 7.22 |
| 110 | 6.57 | 6.55 | 6.54 | 6.53 | 6.53 | 6.54 | 6.58 | 6.65 | 6.75 | 6.89 |
| 130 | 6.30 | 6.27 | 6.25 | 6.24 | 6.24 | 6.25 | 6.29 | 6.36 | 6.46 | 6.62 |
| $10^4 \kappa_T / (\text{MPa}^{-1})$ | | | | | | | | | | |
| 10 | 6.68 | 7.09 | 7.53 | 8.00 | 8.49 | 9.02 | 9.57 | 10.15 | 10.75 | 11.37 |
| 30 | 5.88 | 6.20 | 6.53 | 6.89 | 7.25 | 7.63 | 8.03 | 8.43 | 8.85 | 9.26 |
| 50 | 5.26 | 5.52 | 5.78 | 6.06 | 6.34 | 6.63 | 6.93 | 7.23 | 7.54 | 7.84 |
| 70 | 4.77 | 4.98 | 5.19 | 5.42 | 5.64 | 5.87 | 6.11 | 6.34 | 6.58 | 6.81 |
| 90 | 4.36 | 4.54 | 4.72 | 4.90 | 5.09 | 5.28 | 5.47 | 5.66 | 5.85 | 6.03 |
| 110 | 4.02 | 4.17 | 4.33 | 4.48 | 4.64 | 4.80 | 4.95 | 5.11 | 5.27 | 5.42 |
| 130 | 3.74 | 3.86 | 4.00 | 4.13 | 4.26 | 4.40 | 4.53 | 4.67 | 4.80 | 4.92 |

this work, increasing when the temperature rises and when the pressure decreases. The temperature and pressure dependencies of this property can be observed in Figure 5.

Although several authors⁵⁰ experimentally determine the isobaric thermal expansivity, α_p , this property is very often obtained from $\rho(T)$ data.^{51,52} According to its definition, $\alpha_p = -(1/\rho)(\partial\rho/\partial T)_p$, the isobaric thermal expansivity data can be obtained analytically by differentiating eq 18, taking into account the temperature dependence of $\rho_0(T)$ and $B(T)$. Considering eqs 19 and 20 for the temperature dependencies of $\rho(T, 0.1 \text{ MPa})$ and of $B(T)$, this derived property can be expressed as follows

$$\alpha_p(T, p) = -\frac{A_1 + 2A_2T}{\rho(T, 0.1 \text{ MPa})} - \frac{C(0.1 \text{ MPa} - p)}{(B(T) + p)(B(T) + 0.1 \text{ MPa})} \left[\frac{B_1 + 2B_2T}{1 - C \ln\left(\frac{B(T) + p}{B(T) + 0.1 \text{ MPa}}\right)} \right] \quad (22)$$

where the density is expressed in $\text{kg}\cdot\text{m}^{-3}$, the pressure in MPa, and the temperature in K. The values of C , A_1 , B_1 , and B_2 parameters are taken from Table 2.

However, some authors^{15,51,52} mention that the estimated isobaric thermal expansivity depends on the form of functions $B(T)$ and $\rho_0(T)$, and they recommend deriving the isobaric thermal expansivity from the isobaric densities. So, in the present work, another possibility to calculate the isobaric thermal expansivity has been considered. Over each isobar, the density was correlated with the temperature using a simple polynomial function (similar to eq 19). The polynomial correlation $\rho(T)$ is performed at fixed pressure, and then the derivative with respect to temperature, $(\partial\rho/\partial T)_p$, is determined. Thus, at each pressure we suppose that

$$\rho_p(T) = D_0 + D_1T \quad (23)$$

and consequently

$$\alpha_p(T, p) = -\frac{D_1}{D_0 + D_1T} \quad (24)$$

For each pressure, we get a different set of D_0 and D_1 . By using eq 24 and the calculated densities (eq 23) into $\alpha_p = -(1/\rho)(\partial\rho/\partial T)_p$, the isobaric thermal expansivity at the different (T, p) conditions for diethyl adipate has been derived. In this case, a total of 30 parameters are needed over the full temperature and pressure experimental range. The standard deviations, σ , of the $\rho_p(T)$ correlations at fixed pressure are always lower than $0.4 \text{ kg}\cdot\text{m}^{-3}$. The comparison among the α_p values for diethyl adipate calculated from the global correlation, eq 22, with those obtained with the individual correlations at each fixed pressure, eq 24, gives an average absolute deviation of 0.9 % and a maximum deviation of 2.8 % (at 303.15 K and 130 MPa). The α_p values obtained with the global correlation (eq 22) are reported in Table 3 at some temperatures and pressures. The average deviation is clearly lower than the estimated uncertainty of α_p , ± 4 %. As in the case of the compressibility, the uncertainty of α_p has also been checked establishing comparisons with the α_p reported for toluene by the NIST⁴⁵ and those obtained if eq 22 is applied to the data measured in the background of the present work for toluene. Thus, an average deviation of 1.3 % is obtained which is much lower than the estimated uncertainty.

In Figure 6, the isobaric thermal expansivities (α_p) obtained from eq 22 are plotted against pressure for temperatures between

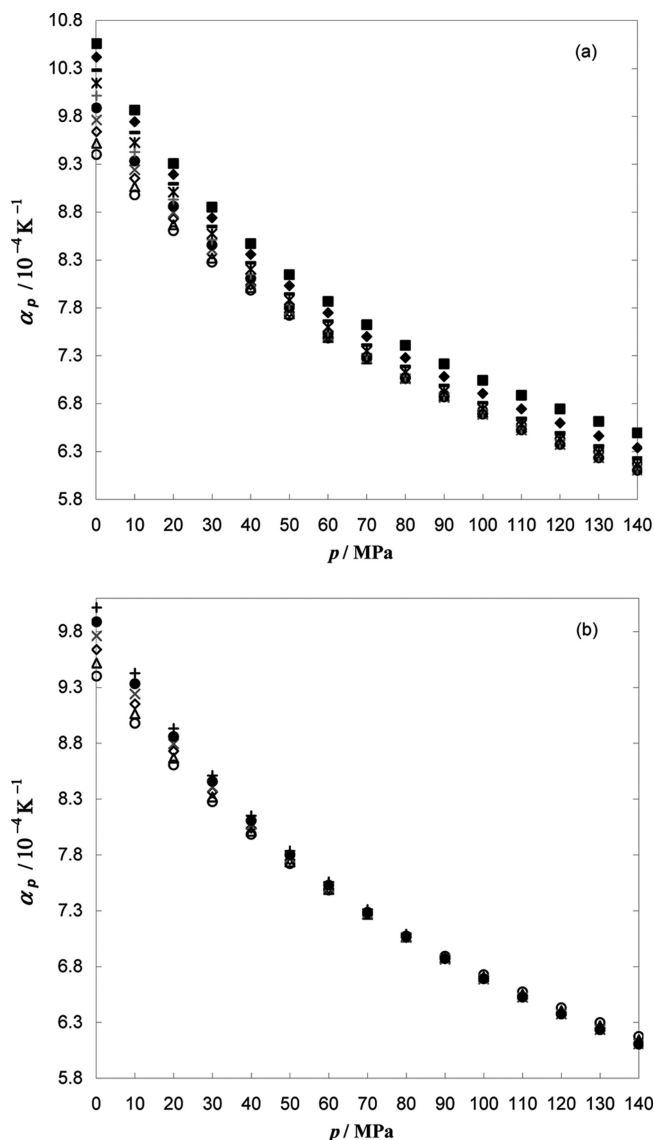


Figure 6. Isobaric thermal expansivities for diethyl adipate vs pressure at different temperatures: \circ , $T = 303.15 \text{ K}$; \triangle , $T = 313.15 \text{ K}$; \diamond , $T = 323.15 \text{ K}$; \times , $T = 333.15 \text{ K}$; \bullet , $T = 343.15 \text{ K}$; $+$, $T = 353.15 \text{ K}$; $*$, $T = 363.15 \text{ K}$; $-$, $T = 373.15 \text{ K}$; \blacklozenge , $T = 383.15 \text{ K}$; \blacksquare , $T = 393.15 \text{ K}$.

(303.15 and 393.15) K (we have not considered (293.15 and 403.15) K isotherms as they are extremes in the temperature derivative). The isobaric thermal expansivity decreases with increasing pressure. With respect to the temperature dependency, α_p reaches the highest values for 393.15 K over the entire pressure interval followed by the values at (383.15, 373.15, and 363.15) K. Nevertheless, for temperatures from (303.15 to 353.15) K, a crossing point of isotherms appears between (60 and 70) MPa. So, over this temperature interval, for pressures lower than this point, α_p rises when the temperature increases, and on the contrary, for pressures higher than this point, the isobaric thermal expansivity decreases when the temperature rises.

Conclusions

A calibration method for the new densimeter DMA HPM is presented for pressures up to 140 MPa and temperatures up to 403.15 K which permits us to measure the density of liquid samples within an uncertainty of $\pm 0.5 \text{ kg}\cdot\text{m}^{-3}$. The validation of this procedure has been checked with toluene

and also with 1-butanol, finding average absolute deviations with previous literature data lower than 0.08 % over all the temperature and pressure intervals. Experimental density data for diethyl adipate were obtained and correlated with the Tammann–Tait equation with a standard deviation of $0.3 \text{ kg}\cdot\text{m}^{-3}$. For diethyl adipate, it can be concluded that the effect of changing the temperature or the pressure has more impact on the values of the isothermal compressibility than on the isobaric thermal expansivity.

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