1430 *J. Chem. Eng. Data* **2005,** *50,* 1430−1433

Volumetric Properties of 1-Phenyldecane and 1-Phenylundecane at Pressures to 65 MPa and Temperature between 293.15 and 353.15 K

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Density data (196 points) for 1-phenyldecane (decylbenzene) and for 1-phenylundecane (undecylbenzene) at several temperatures between (293.15 and 353.15) K and pressures up to 65 MPa are reported. A vibrating tube densimeter with an uncertainty of $\pm 5 \times 10^{-4}$ g·cm^{-3} was used to perform these measurements. The experimental density data were fitted with the Tait-like equation with standard deviations $\leq 1.5 \times 10^{-4}$ g·cm^{-3}. The isobaric thermal expansivity, α_p, and the isothermal compressibility, κ_T, have been evaluated, calculating these derivative density properties using the Tait-like equation.

Introduction

In a recent work,[1] it has been underlined that there is a lack of information on the volumetric and thermoelastic properties of aromatic compounds with large numbers of carbon atoms. The database for the thermophysical properties of these compounds, at present, is scarce and limited (except for very few cases) to narrow temperature and pressure intervals. Most measurements have been made at atmospheric pressure. Moreover, the study of this chemical family presents both obvious fundamental and industrial interests in engineering applications such as in the oil industry.[1] A thorough review of the literature for data as a function of pressure reveals few data for aromatic compounds with more than 15 carbon atoms. The volumetric properties of 1-phenyltridecane (tridecylbenzene, 19 carbon atoms) and 1-phenylpentadecane[1] (pentadecylbenzene, 21 carbon atoms) have been studied up to 60 MPa and up to 40 MPa for 1-phenyldodecane[2] (dodecylbenzene, 18 carbon atoms). For aromatic compounds with fewer than 10 carbon atoms we mention that for toluene (methylbenzene) a review work[3] presents a correlation for the liquid density versus pressure. For ethyl-, butyl-, hexyl-, octyl-, and nonylbenzene, density measurements versus pressure have already been made.[4] With the aim of completing a database relative to this chemical family, in this paper the densities ρ (196 experimental data) for 1-phenyldecane (or decylbenzene, 16 carbon atoms) and for 1-phenylundecane (or undecylbenzene, 17 carbon atoms) have been measured up to 65 MPa between (293.15 and 353.15) K. The isobaric thermal expansivity, $\alpha_p = -(^1/_\rho)(^{\partial\rho}/_{\partial T})_P$, and isothermal compressibility, $\kappa_T = (^1/_\rho)(^{\partial\rho}/_{\partial p})_T$, have been derived.

Experimental Section

Materials. 1-Phenyldecane ($C_{16}H_{26}$, molar mass 218.38 g·mol^{-1}) and 1-phenylundecane ($C_{17}H_{28}$, molar mass 232.40 g·mol^{-1}) were obtained from Fluka (purity of 98%) and Aldrich (purity of 99%), respectively. These chemicals were subject to no further purification.

Measurement Technique. Densities as a function of pressure and temperature were determined using an Anton Paar DMA 60/512P vibrating tube densimeter. All of the devices used for both the filling of the densimeter cell and the measurements have been previously[5] presented. The temperature of the vibrating tube was controlled with a thermoregulated liquid bath. The temperature was measured inside the cell block with an AOIP 5207 thermometer that was calibrated to within ±0.05 K (ITS-90). The pressure was measured with a Hottinger Baldwin Messtechnik (HBM) manometer with an uncertainty of ±0.05 MPa. The calibration parameters of the densimeter cell were determined using the best of the two calibration methods proposed by Lagourette et al.[6] This procedure was based on water as a reference substance and also required the knowledge of the periods of the cell under vacuum as a function of temperature. For water, the density values reported by Kell and Whalley[7] with an uncertainty of 10^{-5} g·cm^{-3} have been used. The uncertainty of our experimental density is less than 0.05%, which is comparable to that in a recent work[8] with the same device. This density uncertainty has been estimated using the law of propagation of uncertainty taking into account the accuracy of the temperature, the pressure, the period of oscillation measurement for water, vacuum, and the studied liquids, and the water density accuracy.

Concerning the reference density values of water reported by Kell and Whalley,[7] it was necessary to point out that the temperature scale used by these authors was the IPTS-68. A comparison of the IPTS-68 with the temperature scale that we used (ITS-90) has been performed over the temperature range of our measurements in a previous paper.[9] The differences between both temperature scales, over the experimental temperature range analyzed in the present work, range from 0.005 K at 293.15 K up to 0.019 K at 353.15 K. These differences are lower than the accuracy of the thermometer that was used. Then, the density reference values of water from the data reported by Kell and Whalley have been recalculated, introducing the correction in temperature from IPTS 68 to ITS 90. The differences in the density values for water are lower than 6×10^{-6} g·cm^{-3}. The calibration of the densimeter has been done using the new reference density values for water, and a difference of 6×10^{-6} g·cm^{-3} has been found for the experimental density of 1-phenyldecane and 1-phenylun-

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10.1021/je0500936 CCC: $30.25

Table 1. Experimental Densities, ρ, at Various Temperatures T and Pressures p

	T/K						
p/MPa	293.15	303.15	313.15	323.15	333.15	343.15	353.15
	ρ/g·cm^{-3}						
	1-Phenyldecane						
0.1	0.8558	0.8491	0.8421	0.8351	0.8278	0.8210	0.8140
5	0.8586	0.8522	0.8453	0.8383	0.8313	0.8246	0.8178
10	0.8615	0.8551	0.8484	0.8416	0.8349	0.8282	0.8215
15	0.8642	0.8582	0.8513	0.8446	0.8382	0.8316	0.8252
20	0.8670	0.8609	0.8541	0.8477	0.8413	0.8349	0.8287
25	0.8695	0.8636	0.8569	0.8508	0.8443	0.8381	0.8319
30	0.8720	0.8662	0.8597	0.8536	0.8472	0.8411	0.8350
35	0.8744	0.8687	0.8623	0.8563	0.8501	0.8440	0.8381
40	0.8768	0.8712	0.8649	0.8590	0.8528	0.8468	0.8410
45	0.8790	0.8735	0.8674	0.8615	0.8555	0.8498	0.8439
50	0.8814	0.8759	0.8699	0.8641	0.8581	0.8524	0.8466
55	0.8835	0.8782	0.8722	0.8666	0.8608	0.8551	0.8494
60	0.8857	0.8804	0.8746	0.8689	0.8632	0.8576	0.8520
65	0.8878	0.8826	0.8768	0.8712	0.8657	0.8601	0.8547
	1-Phenylundecane						
0.1	0.8568	0.8495	0.8425	0.8355	0.8286	0.8217	0.8145
5	0.8596	0.8526	0.8457	0.8389	0.8319	0.8252	0.8185
10	0.8624	0.8554	0.8487	0.8421	0.8354	0.8287	0.8222
15	0.8651	0.8585	0.8516	0.8451	0.8386	0.8322	0.8259
20	0.8677	0.8611	0.8544	0.8481	0.8417	0.8354	0.8293
25	0.8703	0.8638	0.8572	0.8511	0.8446	0.8385	0.8325
30	0.8727	0.8663	0.8598	0.8539	0.8475	0.8414	0.8355
35	0.8752	0.8687	0.8624	0.8566	0.8503	0.8440	0.8384
40	0.8775	0.8712	0.8649	0.8592	0.8530	0.8468	0.8413
45	0.8798	0.8735	0.8675	0.8617	0.8557	0.8496	0.8441
50	0.8820	0.8758	0.8699	0.8642	0.8583	0.8523	0.8466
55	0.8842	0.8781	0.8723	0.8666	0.8609	0.8549	0.8494
60	0.8863	0.8803	0.8745	0.8690	0.8633	0.8574	0.8519
65	0.8882	0.8825	0.8768	0.8713	0.8656	0.8598	0.8546

decane. Therefore, the uncertainty due to the correction in the temperature scale is markedly lower than the estimated experimental uncertainty of the measurements.

Finally, the estimation of our experimental densities using the recent values given for water by Span[10] instead of the values reported by Kell and Whalley[7] shows differences that are markedly lower than our estimated densimeter technique uncertainty.

Results

The measured densities of 1-phenyldecane and 1-phenylundecane are reported in Table 1 along isotherms between 293.15 K and 353.15 K at pressures up to 65 MPa. To correlate our values over the temperature and pressure intervals, the following Tait-like equation has been used as in previous works[8,9,11,12]

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

where $\rho_0(T)$ is the temperature dependence of the density at 0.1 MPa that is expressed as

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

In eq 1, parameter C was assumed to be temperature-independent, whereas $B(T)$ is given by the following polynomial function:

$$B(T) = B_0 + B_1T + B_2T^2 \tag{3}$$

Table 2. Obtained Parameters and Deviations for the Density Correlation by Using Equation 1

coefficients	1-phenyldecane	1-phenylundecane
A_0/g·cm^{-3}	0.97877	0.98602
A_1/g·cm^{-3}·K^{-1}	-7.8964×10^{-7}	-1.7798×10^{-6}
A_2/g·cm^{-3}·K^{-2}	-1.9601×10^{-6}	-2.1428×10^{-6}
A_3/g·cm^{-3}·K^{-3}	1.8159×10^{-9}	2.1878×10^{-9}
C	0.087944	0.077028
B_0/MPa	480.83	324.99
B_1/MPa·K^{-1}	-1.6932	-0.91079
B_2/MPa·K^{-2}	0.0016652	0.0059699
deviation		
σ/g·cm^{-3}	1.0×10^{-4}	1.5×10^{-4}
AAD/%	1.1×10^{-2}	1.3×10^{-2}
DMAX/%	5.2×10^{-2}	4.1×10^{-2}
bias/%	2.8×10^{-3}	1.3×10^{-3}

To compare the experimental density values with those obtained with the Tait correlation, we have used the absolute average deviation (AAD), the maximum deviation (DMAX), the average deviation (bias), and the standard deviation σ, which are defined as follows

$$\mathrm{AAD} = \frac{100}{N}\sum_{i=1}^{N}\left|\frac{\rho_i^{\mathrm{exptl}} - \rho_i^{\mathrm{calcd}}}{\rho_i^{\mathrm{exptl}}}\right| \tag{4}$$

$$\mathrm{DMAX} = \max\left(100\left|\frac{\rho_i^{\mathrm{exptl}} - \rho_i^{\mathrm{calcd}}}{\rho_i^{\mathrm{exptl}}}\right|\right) \tag{5}$$

$$\mathrm{bias} = \frac{100}{N}\sum_{i=1}^{N}\frac{\rho_i^{\mathrm{exptl}} - \rho_i^{\mathrm{calcd}}}{\rho_i^{\mathrm{exptl}}} \tag{6}$$

$$\sigma = \sqrt{\frac{\sum_{i=1}^{N}(\rho_i^{\mathrm{exptl}} - \rho_i^{\mathrm{calcd}})^2}{N - m}} \tag{7}$$

where N is the number of experimental data points (N = 98 for each compound) and m is the number of parameters (m = 8 in eq 1).

The Tait-correlation parameters, along with the AAD, DMAX, bias, and standard deviation, σ, obtained with this correlation are shown in Table 2 for both compounds. Notice that the standard deviation is of the same order of magnitude as the experimental uncertainty. Consequently, eqs 1 to 3 make it possible to interpolate the density under any pT conditions.

Figure 1 shows the variation of density as a function of temperature for 1-phenyldecane at P = (0.1, 30, and 60) MPa, and Figure 2 shows the variation of density as a function of pressure for 1-phenylundecane at T = (293.15, 323.15, and 353.15) K. 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. Furthermore, it should be noted that the variations of the density with temperature, in the interval considered in this investigation (293.15 to 353.15 K), are practically linear, so in eq 2, it is possible to use a simpler linear expression of the form $\rho_0(T) = A'_0 + A'_1T$.

As already indicated, no study of density at high pressure has been found in the literature for these substances. However, some comparison with the literature data at atmospheric pressure can be made for 1-phenyldecane. For this compound, our experimental values at atmospheric pressure show relative differences of 0.03 % and 0.09 %

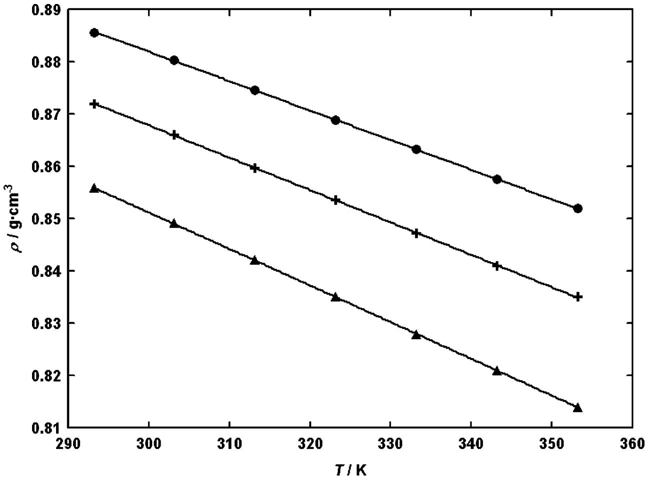

Figure 1. Variation of 1-phenyldecane density as a function of temperature: ▲, P = 0.1 MPa; +, P = 30 MPa; ●, P = 60 MPa.

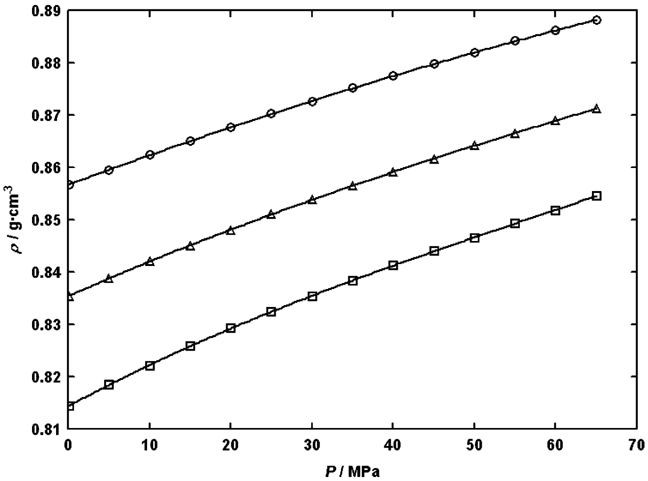

Figure 2. Variation of the 1-phenylundecane density as a function of pressure: ○, T = 293.15 K; △, T = 323.15 K; □, T = 353.15 K.

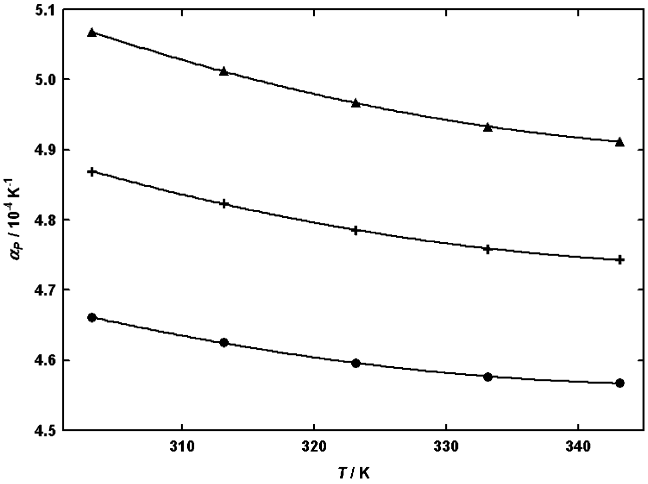

Figure 3. Variation of the 1-phenyldecane α_p as a function of temperature: ▲, P = 5 MPa; +, P = 30 MPa; ●, P = 60 MPa.

with the data reported by Camin et al.[13] at 293.15 K (ρ = 0.85553) and 303.15 K (ρ = 0.84833) made with a density balance and 1-phenyldecane with 99.9% (mole) purity. The uncertainty claimed by these authors was 1 × 10^{-4} g·cm^{-3}.

Finally, it should be mentioned that the variation of density with the number of carbon atoms of the alkyl chain, for this chemical family, is very weak. For instance, at P = 30 MPa and T = 333.15 K, the densities are 0.8472 g·cm^{-3} for 1-phenyldecane (this work), 0.8475 g·cm^{-3} for 1-phenylundecane (this work), 0.8471 g·cm^{-3} for 1-phenyldodecane[2], 0.8459 g·cm^{-3} for 1-phenyltridecane,[1] and 0.8469 g·cm^{-3} for 1-phenylpentadecane.[1] An examination of the data as a function of temperature and pressure for alkylbenzenes ranging from toluene to 1-phenyltridecane[1–4] does not reveal a general trend in the density behavior versus the number of carbon atoms.

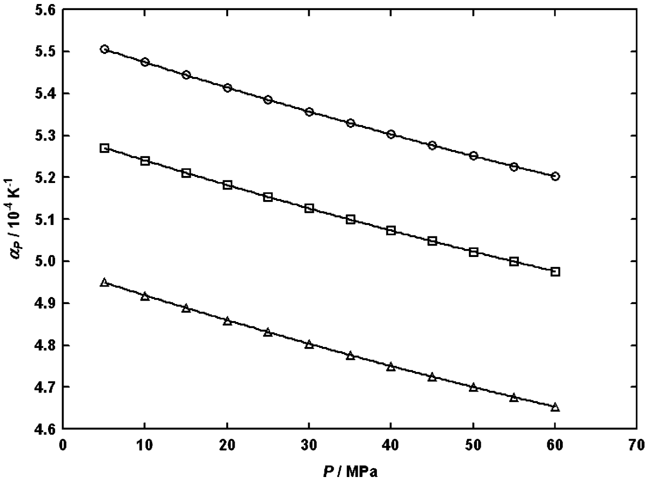

Figure 4. Variation of the 1-phenylundecane α_p as a function of pressure: ○, T = 303.15 K; □, T = 323.15 K; △, T = 343.15 K.

Derived Thermodynamic Properties

The temperature and pressure dependences of the isobaric thermal expansivity, α_p, and the isothermal compressibility, κ_T, are obtained from eq 1 and are expressed as follows:

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

$$\alpha_p = -\left(\frac{1}{\rho}\right)\left(\frac{\partial \rho}{\partial T}\right)_p = -\frac{1}{\rho}\left(\frac{\rho_0'(T)\,F(T,p) - \rho_0(T)\,F'(T,p)}{F(T,p)^2}\right) \tag{9}$$

where $F(T, P)$ is the denominator of the right side of eq 1 and $F'(T, p)$ is its temperature derivative given by

$$F'(T,p) = \left(\frac{\partial F(T,p)}{\partial T}\right)_p = CB'(T)\left(\frac{1}{B(T)+0.1\ \mathrm{MPa}} - \frac{1}{B(T)+p}\right) \tag{10}$$

where $\rho_0'(T) = \mathrm{d}\rho_0(T)/\mathrm{d}T$ and $B'(T) = \mathrm{d}B/\mathrm{d}T$ are easily obtained from eqs 2 and 3, respectively, and $\rho_0'(T) = A_1 + 2A_2T + 3A_3T^2$ and $B'(T) = B_1 + 2B_2T$.

The isothermal compressibility, κ_T, and the isobaric thermal expansivity, α_p, were calculated from eqs 8 and 9 derived from the Tait-like correlation $\rho(T, p)$ using the coefficients given in Table 2. As recently indicated in a similar high-pressure density study[8] with the same technique, the estimated uncertainty is 1 % for the isothermal compressibility, and 0.5% for the isobaric thermal expansivity. Figure 3 shows the variation of α_p as a function of temperature for 1-phenyldecane, and Figure 4 shows the variation of α_p as a function of pressure for 1-phenylundecane. Figure 5 shows the variation of κ_T as a function of temperature for 1-phenyldecane, and Figure 6 shows the variation of κ_T as a function of pressure for 1-phenylunde-

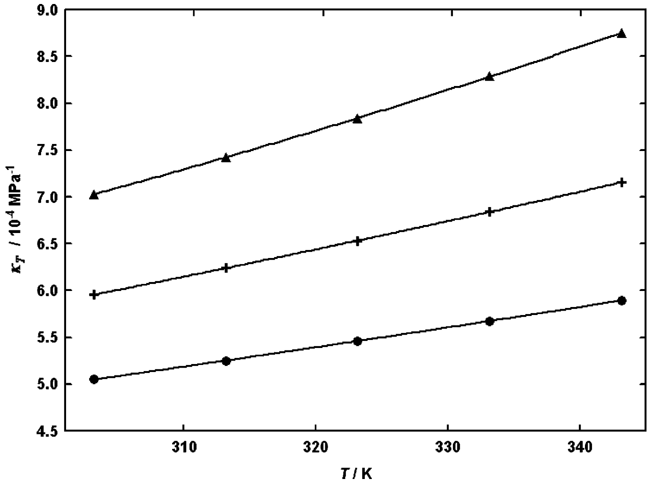

Figure 5. Variation of the 1-phenyldecane κ_T as a function of temperature: ▲, P = 5 MPa; +, P = 30 MPa; ●, P = 60 MPa.

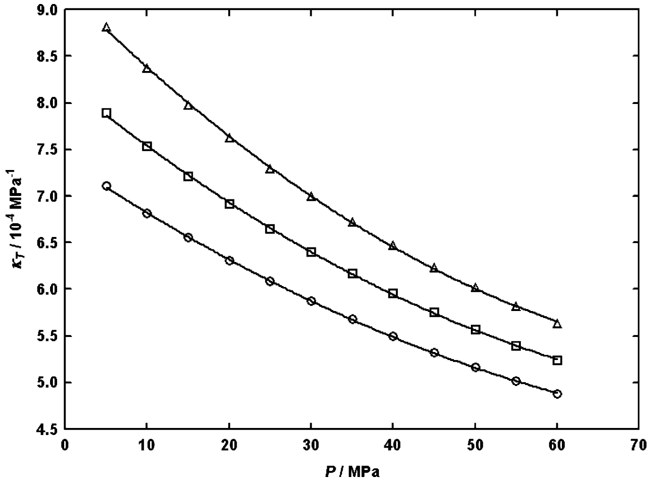

Figure 6. Variation of the 1-phenylundecane κ_T as a function of pressure: ○, T = 303.15 K; □, T = 323.15 K; △, T = 343.15 K.

cane. We notice that in the temperature interval studied the variations of α_p for 1-phenyldecane are quite small.

Conclusions

The pressure and temperature dependences of the density of 1-phenyldecane and 1-phenylundecane have been determined over a temperature range of (293.15 to 353.15) K and at pressure from (0.1 to 65) MPa. The density data were fitted to a Tait-like equation within the experimental uncertainty, and the isobaric thermal expansivity and the isothermal compressibility were derived. These data are a part of a program of systematic investigation of hydrocarbons with high molecular weight undertaken in our laboratory. These data will be of use for the thermodynamic characterization of this chemical family, which is important in petroleum engineering because of the presence of these compounds in heavy oils.

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Received for review March 10, 2005. Accepted May 22, 2005.

JE0500936