Density and speed of sound measurements of hexadecane ☆

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ARTICLE INFO

ABSTRACT

The density and speed of sound of hexadecane have been measured with two instruments. Both instruments use the vibrating-tube method for measuring density. Ambient pressure (83 kPa) density and speed of sound were measured with a commercial instrument from $T = (290.65$ to $343.15)$ K. Adiabatic compressibilities are derived from the density and speed of sound data at ambient pressure. Compressed liquid density was measured in a second instrument and ranged from $T = (310$ to $470)$ K with pressures from $(1$ to $50)$ MPa. The overall relative expanded uncertainty of the compressed liquid density measurements is 0.10–0.13% $(k = 2)$. The overall relative expanded uncertainty $(k = 2.3)$ of the speed of sound measurements is 0.2% and that of the ambient pressure density measurements is approximately 0.04% $(k = 2.3)$. The ambient pressure and compressed liquid density measurements are correlated within 0.1% with a modified Tait equation.

Published by Elsevier Ltd.

1. Introduction

Hexadecane ($C_{16}H_{34}$, CAS No. 544-76-3), also known as cetane, is the namesake of the cetane number that characterizes the combustion propensity of a liquid during compression ignition. Hexadecane was given a cetane number of 100, while α-methyl naphthalene was assigned a cetane number of 0. Diesel fuels can be classified (given a cetane number) based on this reference system per ASTM Standard D975 [1]. Hence, even though hexadecane is a slightly larger molecule than what is thought of as the average for petrodiesel, it is commonly used as a surrogate to represent the heavier fractions for the purpose of modeling diesel fuel [2].

Diesel fuel produced from petroleum (petrodiesel) is obtained from the fractional distillation of crude oil at temperatures from $T = (473$ to $623)$ K at atmospheric pressure [3]. Saturated hydrocarbons (primarily paraffins including *n*, *iso*, and cycloparaffins) make up the majority of the composition of petrodiesel, with lesser amounts of aromatic hydrocarbons (including naphthalenes and alkylbenzenes) commonly present as well. Petrodiesel is considered to consist predominantly of hydrocarbons ranging from $C_{10}H_{20}$ to $C_{15}H_{28}$; however, it is a complex mixture in which component fractions can vary considerably, while the fuel is still within specifications.

Results of density and speed of sound measurements for hexadecane are reported in this work. Adiabatic compressibilities have been derived from the ambient pressure density and speed of sound data and are also included in the tables. Compressed liquid density data have been extrapolated to 83 kPa and combined with ambient pressure density data for correlation by a Rackett equation. Additionally, the compressed liquid density data have been correlated with a Tait equation and compared to the existing literature data.

2. Experimental

The hexadecane measured in this work was obtained from Sigma–Aldrich Chemicals[1] with a stated minimum purity of 0.99. Prior to measurements of compressed liquid density, the sample was transferred to a stainless steel cylinder and degassed as described by Outcalt and McLinden [4]. Otherwise, the sample was used as received.

A density and speed of sound analyzer, the DSA 5000 from Anton Paar Company, was used for measurements at ambient pressure (~83 kPa in Boulder, CO, USA). Details of the instrument and experimental procedures have been reported in [5] and only a brief description will be given here. The instrument contains a sound speed cell and a vibrating quartz tube densimeter connected in series. Temperature is measured with an integrated Pt-100 thermometer with an estimated uncertainty of 0.01 K. The instrument was calibrated with air and deionized water at $T = (293.15, 313.15,$

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[1] In order to describe materials and experimental procedures adequately, it is occasionally necessary to identify commercial products by manufacturers' names or labels. In no instance does such identification imply endorsement by the National Institute of Standards and Technology, nor does it imply that the particular product or equipment is necessarily the best available for the purpose.

and 333.15) K. In addition, the instrument performance is checked before and after measurements to ensure that the density and speed of sound of water at 293.15 K are reproduced within 0.01%. For the hexadecane measurements, temperature scans were programmed from $T = (343.15$ to 290.65) K. In an attempt to sub-cool the liquid below its melting temperature ($T = 291$ K), data were collected every 5 K from $T = (343.15$ to 293.15) K and in 0.5 K increments thereafter. This approach made it possible to obtain some data on sub-cooled hexadecane. Fresh samples of test liquid were injected for each temperature scan. Four temperature scans were performed for hexadecane. The relative standard deviation (reproducibility) of these repeated sound speed measurements was no greater than 0.02%. Based on repeated measurements of the speed of sound of water, we conservatively estimate the relative expanded uncertainty ($k = 2.3$) of these measurements to be 0.2%.

Densities of the hexadecane were measured during the same temperature scans that were carried out to obtain the speeds of sound. ASTM D4052 Standard Test Method [6] was taken into account during these measurements. The instrument uses a proprietary method to correct the measured densities for the viscosity of the test liquid if the viscosity exceeds 700 mPa·s^{-1}. The viscosity of hexadecane in the measurements presented here did not exceed this limit and thus no correction was made. The relative standard deviation (reproducibility) of repeated density measurements was no greater than 2 ppm. Repeat measurements of the density of Standard Reference Material (SRM) 211d (toluene) and comparison with the expression of McLinden and Splett [7], resulted in deviations of less than 0.02%. In contrast, repeat measurements of the density of SRM 2214 (2,2,4-trimethylpentane (isooctane)) and comparison with certified values resulted in deviations of less than 0.003%. The density of SRM 211d is (819.5 to 880.8) kg·m^{-3} over the temperature range of this study, while the density of SRM 2214 is (687.8 to 696.0) kg·m^{-3} over the temperature range $T = (288.15$ to 298.15) K. The density of hexadecane falls between these two limits. In the absence of additional standard reference materials with intermediate densities, it is not possible to predict the instrument's behavior between the limits of toluene and isooctane. To be conservative, we have assumed 0.02% to be representative of a systematic uncertainty when calculating an overall uncertainty for the density measurements. The resulting overall relative expanded uncertainty ($k = 2.3$) of the density measurements is 0.04%.

TABLE 1

Density (ρ), speed of sound (w), and derived values of isentropic compressibility (κ_s) of hexadecane measured in the density and sound speed analyzer. The ambient pressure during the measurements was approximately 0.083 MPa.

T/K	ρ/(kg·m^{-3})	w/(m·s^{-1})	κ_s/(TPa^{-1})
290.65[a]	775.0	1367.2	690.3
291.15	774.7	1365.3	692.5
291.65	774.4	1363.4	694.8
292.15	774.0	1361.5	697.0
292.65	773.6	1359.6	699.2
293.15	773.3	1357.7	701.6
298.15	769.8	1338.8	724.7
303.15	766.4	1320.2	748.7
308.15	762.9	1301.7	773.6
313.15	759.4	1283.4	799.4
318.15	756.0	1265.3	826.2
323.15	752.5	1247.4	854.0
328.15	749.1	1229.8	882.7
333.15	745.6	1212.3	912.6
338.15	742.2	1194.9	943.6
343.15	738.7	1177.9	975.8

[a] Sub-cooled liquid.

Densities of the compressed liquid were measured with the automated densimeter described by Outcalt and McLinden [4]. Central to the apparatus is a vibrating-tube densimeter DMA-HPM from Anton Paar Company. In order to minimize the uncertainty in the measurements, several physical and procedural improvements have been implemented beyond that of the commercial instrument operated in a stand-alone mode. Some of these improvements include more accurate temperature and pressure measurements, better temperature control, and complete automation of the instrument control and data acquisition. The temperature range of the instrument is $T = (270$ to 470) K, with pressures up to 50 MPa. In this work, we measured hexadecane along nine isotherms over the range (1 to 50) MPa. The instrument was calibrated with propane and toluene over the entire temperature and pressure range. Uncertainty in the temperature measurement = 0.03 K and the pressure uncertainty = 5 kPa, where $k = 1$ for both values. The overall expanded uncertainty ($k = 2$) in density is (0.64 to 0.81) kg·m^{-3} corresponding to a relative uncertainty in density of 0.10–0.13%.

3. Results

Table 1 lists measured values of density, speed of sound, and derived adiabatic compressibilities for hexadecane from (290.65 to 343.15) K at ambient atmospheric pressure. Speed of sound and adiabatic compressibility results are also illustrated in figure. 1. Isentropic compressibilities were calculated from the measured densities and speeds of sound via the thermodynamic relation

$$\kappa_s = -(\partial V/\partial \rho)_s/V = 1/(\rho w^2), \qquad (1)$$

where V denotes volume, p is pressure, ρ is the density, and w the speed of sound. The subscript s indicates that the derivative is taken at constant entropy.
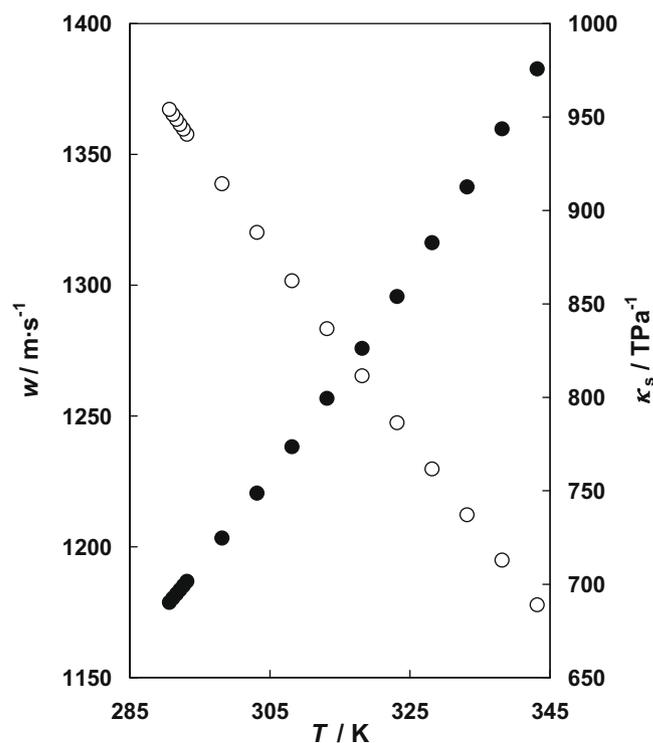

FIGURE 1. ○, Ambient pressure speed of sound, and ●, derived adiabatic compressibility, as a function of temperature.

Table 2 lists measured density values of compressed liquid hexadecane from T = (310 to 470) K with pressures to 50 MPa. Also listed are density values extrapolated to 83 kPa (the approximate atmospheric pressure in Boulder, CO) for each temperature. These were obtained by fitting a second-order polynomial to the isothermal data at pressures less than or equal to 10 MPa and extrapolating to 83 kPa. This extrapolation was performed to examine the consistency of the compressed liquid data with the measurement results at ambient pressure from the density and sound speed analyzer. Figure 2 depicts measured compressed liquid and ambient pressure densities as a function of temperature and pressure.

3.1. Correlation of data

The speed of sound data at atmospheric pressure were correlated as a function of temperature with the second-order polynomial,

$$(w/\{m \cdot s^{-1}\}) = A_1 + A_2(T/K) + A_3(T/K)^2, \tag{2}$$

where w is the speed of sound in $m \cdot s^{-1}$ and T is the absolute temperature. This simple correlation, with the parameter values A_1 = 2804.158, A_2 = −6.077077, and A_3 = 0.0038984, fits our speed of sound data with an average absolute deviation (AAD) of 0.001% and serves as a baseline against which we can compare our data to those found in the literature [8–19]. Figure 3 shows that for data in the same temperature range as ours and at approximately atmospheric pressure, the majority agree with our data within the relative expanded uncertainty of 0.2%. There are an additional three points of Rolling and Vogt [17] not shown in the figure, as their deviations were greater than ±0.4%.Density measurements at ambient pressure and values extrapolated to 83 kPa from measured compressed liquid data were correlated with the Rackett equation to check the repeatability of the instruments and the consistency of our data in the combined temperature range T = (290.65 to 470) K. The Rackett equation is written as

$$(\rho/\{kg \cdot m^{-3}\}) = \beta_1 \cdot \beta_2^{-\left(1 + \left(1 - \frac{(T/K)}{\beta_3}\right)^{\beta_4}\right)}. \tag{3}$$

Table 3 lists the Rackett correlation parameters; also listed are the standard deviations of the parameters. The 1996 survey of critical properties of normal alkanes by Ambrose and Tsonopoulos [20] lists a range of critical temperatures for hexadecane from T = (717 to 723) K, with a recommended value of T = 723 ± 2 K. In the correlation of our data, we constrained the β_3 parameter to 722.1 to reflect the critical temperature of hexadecane as given by Lemmon and Goodwin [21]. The Rackett equation is most commonly used to fit saturated liquid densities, but it works well for our purposes as vapor pressures of hexadecane over the temperature range of our data are only slightly less than atmospheric pressure. Our Rackett correlation serves as the baseline (in figure 4) to compare our ambient pressure and extrapolated density values as well as correlations for ambient pressure and saturated liquid densities found in the American Institute for Chemical Engineers (AIChE) Design Institute for Physical Properties (DIPPR) [22] and ThermoData Engine (TDE) [23] databases. Figure 4 also includes 25 sets (from the literature) [8,24–47] of ambient pressure liquid density data. These literature data were found in the DIPPR and TDE databases and comprise all of the ambient pressure liquid density data found in those two databases with the exception of sources that listed only a single point. Figure 4 illustrates that both our measured and extrapolated data agree within 0.1%, and that there is good agreement between our correlation and that of the TDE correlation. For both the TDE and DIPPR correlations, the larger deviations at higher temperatures are not surprising, because at those temperatures our Rackett correlation is based solely on our extrapolated

TABLE 2
Compressed liquid densities of hexadecane measured in the high pressure vibrating-tube densimeter along isotherms from T = (310 to 470) K. Values extrapolated to 0.083 MPa are indicated in *italics*.

| 310.00 K		330.00 K		350.00 K		370.00 K		390.00 K		410.00 K		430.00 K		450.00 K		470.00 K	
p/MPa	$\rho/(kg \cdot m^{-3})$	p/MPa	$\rho/(kg \cdot m^{-3})$	p/MPa	$\rho/(kg \cdot m^{-3})$	p/MPa	$\rho/(kg \cdot m^{-3})$	p/MPa	$\rho/(kg \cdot m^{-3})$	p/MPa	$\rho/(kg \cdot m^{-3})$	p/MPa	$\rho/(kg \cdot m^{-3})$	p/MPa	$\rho/(kg \cdot m^{-3})$	p/MPa	$\rho/(kg \cdot m^{-3})$
36.73	783.55	45.96	777.44	40.13	762.70	41.34	752.26	46.90	744.61	50.70	736.75	48.49	724.39	45.31	711.32	47.79	703.60
34.70	782.46	38.24	773.11	36.14	760.24	34.01	747.41	44.32	742.94	44.82	732.63	44.32	721.28	43.40	709.80	42.79	699.27
25.60	777.30	36.16	771.90	25.56	753.33	25.58	741.44	39.90	739.91	37.22	726.98	39.59	717.56	39.15	706.20	38.87	695.70
20.65	774.41	25.98	765.74	20.60	749.88	20.61	737.69	35.16	736.52	30.42	721.61	30.42	709.84	30.80	698.66	30.47	687.53
15.70	771.38	20.68	762.35	15.65	746.27	15.66	733.76	25.55	729.18	25.57	717.52	25.53	705.37	25.53	693.48	25.53	682.28
10.77	768.25	15.74	759.04	10.71	742.54	10.71	729.65	20.60	725.12	20.63	713.12	20.56	700.57	20.56	688.27	20.64	676.69
5.87	765.01	10.79	755.61	5.79	738.58	5.78	725.30	15.65	720.83	15.70	708.47	15.59	695.44	15.59	682.65	15.69	670.57
2.71	762.85	5.89	752.03	2.63	735.94	2.62	722.35	10.69	716.31	10.75	703.47	10.65	689.93	10.62	676.53	10.73	663.85
1.11	761.74	2.73	749.66	1.02	734.55	1.03	720.83	5.79	711.50	5.83	698.12	5.77	684.04	5.75	669.98	5.77	656.40
0.083	*761.02*	1.13	748.39	*0.083*	*733.74*	*0.083*	*719.90*	2.63	708.23	2.68	694.47	2.59	679.90	2.56	665.22	2.66	651.24
		0.083	*747.58*					1.04	706.50	1.08	692.52	0.97	677.73	0.96	662.64	1.07	648.42
								0.083	*705.46*	*0.083*	*691.30*	*0.083*	*676.50*	*0.083*	*661.21*	*0.083*	*646.65*
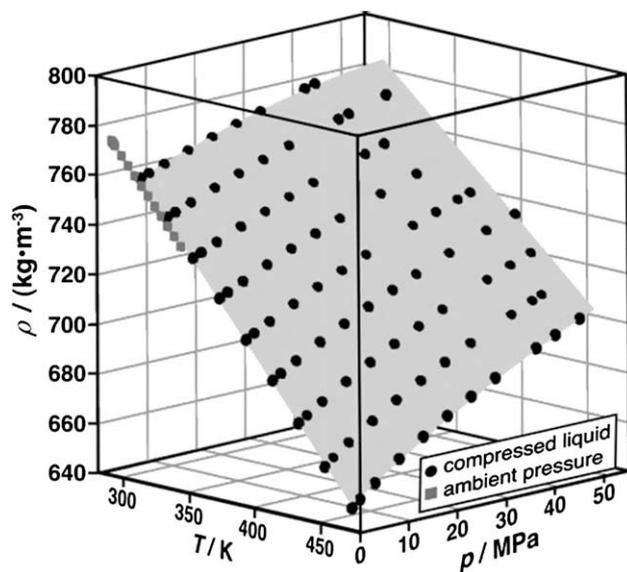
FIGURE 2. Measured compressed liquid and ambient pressure densities as a function of temperature and pressure.

data, and those data have a greater uncertainty than our measured data. The TDE correlation is based on saturated liquid density data, and the good agreement with our data confirms that the difference between saturation pressures and atmospheric pressure has negligible affect on liquid densities in the temperature range of our data. The deviations of the DIPPR correlation from ours are generally higher than those of TDE; however, the majority of the data used to formulate the DIPPR correlation agree with our measured and extrapolated data within ±0.2%.

To make the present results immediately usable for engineering and design purposes the compressed liquid density data were correlated with a Tait equation similar to that of Dymond and Malhotra [48], with the exception that the parameter C was treated as a

temperature independent constant. The equation used to fit the compressed liquid density data reads

$$\rho(T, \rho) = \frac{\rho_{ref}(T, p_{ref})}{1 - C \ln \left(\frac{p + B(T)}{p_{ref} + B(T)} \right)}, \tag{4}$$

where $\rho_{ref}(T)$ is the temperature-dependent density at the reference pressure $p_{ref} = 0.083$ MPa from equation (3). The temperature dependence of the Tait parameter $B(T)$ was expressed by a quadratic polynomial,

$$(B(T)/\{MPa\}) = D_1 + D_2 T_r + D_3 T_r^2, \tag{5}$$

where $T_r = T/273.15$ K. Parameters for the Tait correlations are given in table 4. The correlation represents our data with an average absolute deviation (AAD) of 0.033% which is well within the estimated uncertainty. Figure 5 shows deviations of measured compressed liquid densities from the reported Tait correlation.

A search of the literature found several sets of compressed liquid density data [31,35,44,49–56] on hexadecane at lower temperatures, but only three sets that included data at temperatures above $T = 393$ K: Matthews et al. [55], Chang et al. [31], and Amorim et al. [49] The data set of Matthews et al. [55] consists of 10 points ranging from $T = (323$ to $564)$ K, with a maximum pressure of 3.5 MPa. The Chang et al. [31] data range from $T = (333$ to $413)$ K to pressures of 30 MPa, and the Amorim et al. [49] set includes data from $T = (318$ to $413)$ K at pressures from (6.9 to 62.1) MPa. Figure 6 shows deviations of literature data from a

Table 3
Parameters (and their standard deviation, σ) of the Rackett correlation (equation (3)), for the density of hexadecane at an ambient pressure of 83 kPa and temperatures from (290 to 470) K.

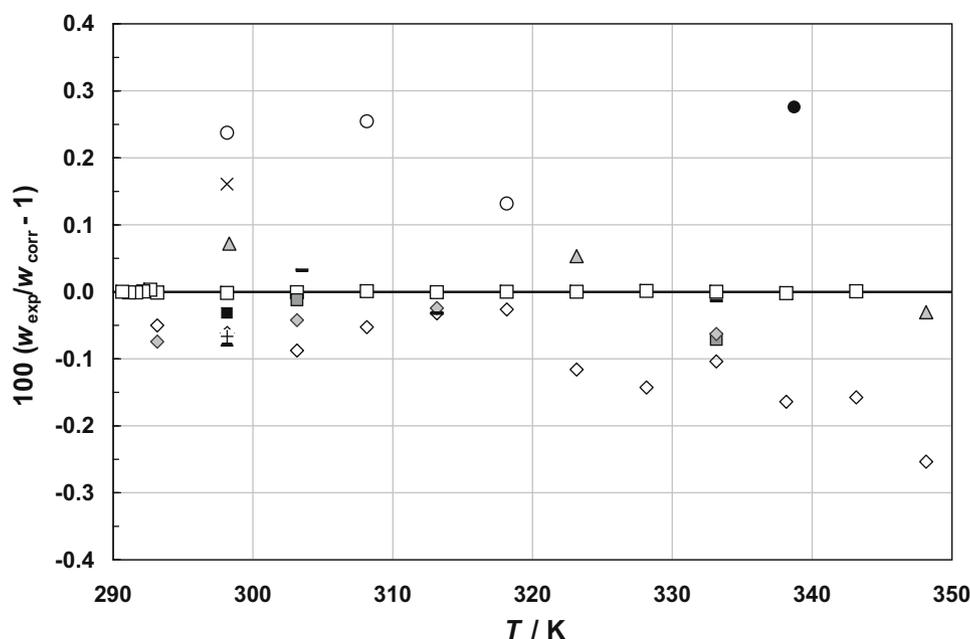| Parameter	Value	σ
β_1	139.7	3.0
β_2	0.3788	4.1×10^{-3}
β_3	722.1	2.9
β_4	0.5190	4.1×10^{-3}

FIGURE 3. Comparison of our speed of sound data and literature data to a baseline of the polynomial fit of our data. □, Our data; ○, Aminabhavi [8]; ×, Awwad [9]; △, Ball [10]; ▲, Benson [11]; ◈, Belhouwer [12]. ◇ Bolotnikov [13]; ◉, Dubey [14]; +, Janquera [15]; ▦, Khasanshin [16]; ●, Rolling [17]; ■, Tardajos [18]; –, Ye [19].
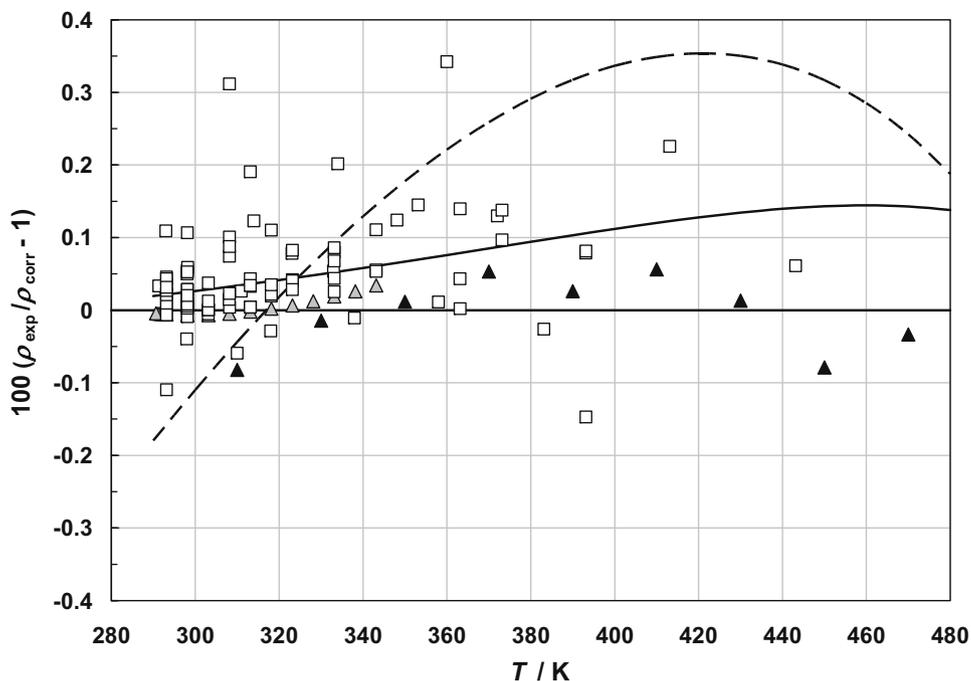
FIGURE 4. Comparison of measured and extrapolated ambient pressure density data, the DIPPR and TDE correlations, and literature data. The baseline is the reported Rackett equation (equation (3)) correlated to the present data. △, our ambient pressure data; ▲, our extrapolated data; □, literature data [8,24–47]; – –, DIPPR correlation [22]; —, TDE correlation [23].

TABLE 4
Parameters (and their standard deviation, σ) of the Tait correlation (equations (4) and (5)) for the density of hexadecane.

Parameter	Value	σ
C	0.08447	2.0×10^{-4}
D_1	361.7	1.2
D_2	−327.0	1.4
D_3	78.04	0.43

baseline of the Tait correlation fitted to our data as a function of temperature. The figure illustrates that the literature data are relatively evenly scattered about the baseline, and with the exceptions of the data of Amorim *et al.* [49], Chang *et al.* [31], and Glaser *et al.* [51] the majority of the data deviate less than ±0.2% and thus agree well with our data.
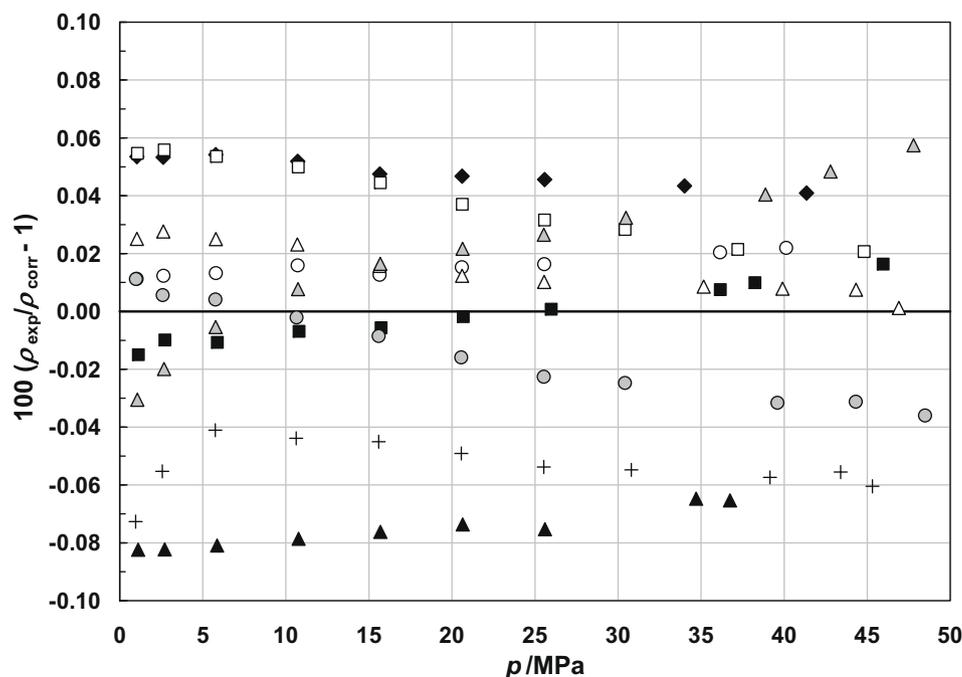

FIGURE 5. Deviations of measured compressed liquid densities from the Tait correlation (equations (3) and (4)) as a function of pressure. Temperatures: ▲, 310 K; ■, 330 K; ○, 350 K; ♦, 370 K; △, 390 K; □, 410 K; ◔, 430 K; +, 450 K; △, 470 K.
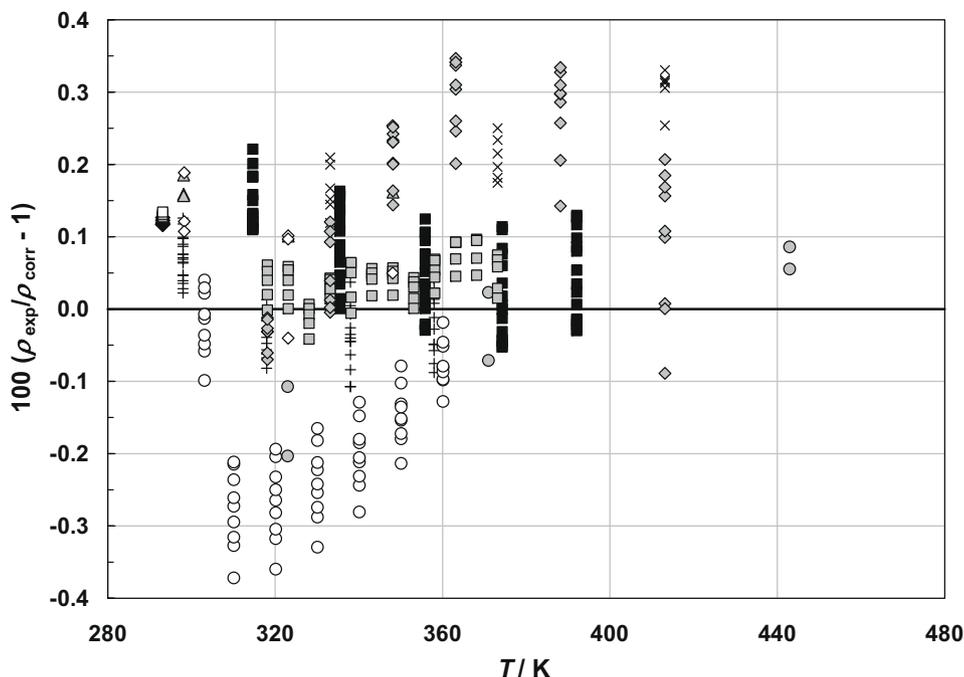
FIGURE 6. Deviations of compressed liquid density data found in the literature from our Tait correlation as a function of temperature: +, Snyder [44]; ■, Gouel [52]; ○, Glaser [51]; ♦, Holzapfel [53]; □, Holzapfel [54]; ◉, Matthews [55]; ▣, Banipal [50]; △, Tanaka [56]; ◇, Dymond [35]; ×, Chang [31]; ◈, Amorim [49].

4. Conclusions

Density and speed of sound at ambient pressure and compressed liquid densities of hexadecane have been measured covering a temperature range of T = (290.65 to 470) K with pressures to 50 MPa. The compressed liquid density data have been correlated with a modified Tait equation within the experimental uncertainty of 0.1%, and the speed of sound data at ambient pressure were correlated with a second-order polynomial in temperature within the experimental uncertainty of 0.2%. Our measurements are consistent with previously published data and extend the range of available compressed liquid density data from T = (413 to 470) K.

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