

Densities of n-Alkanes and Their Mixtures at Elevated Pressures

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Accurate density data for n-alkanes over a wide range of temperature and pressure have been used to test existing correlation and prediction methods. It is found that the most successful representation at temperatures up to 0.66 times the critical temperature and pressures up to 150 MPa is given by the Tait equation in the form $(\rho - \rho_0)/\rho = C \log[(B + P)/(B + P_0)]$, where subscript 0 refers to 0.101 MPa, with C equal to 0.2000, and $[B + (C_n - 6)]$, where C_n is the number of carbon atoms in the alkane chain, is a smooth function of reduced temperature. A simple extension of this method to mixtures gives an excellent prediction of densities at pressures up to 150 MPa over the same reduced temperature range.

KEY WORDS: alkanes; density; high pressure; liquid mixtures; Tait equation.

1. INTRODUCTION

For the correlation of transport properties of liquids over a wide temperature range and up to elevated pressures, it is the molar volume which is the important variable [1-5]. In the case of viscosity, for example, where recent accurate measurements have been made at pressures up to 500 MPa, mainly on n-alkanes and their mixtures, Dymond and collaborators [6, 7] have satisfactorily correlated the data on the basis of a free-volume approach. For this purpose, the molar volumes and hence the liquid densities must be known with an accuracy better than 0.2%.

The availability of new experimental data for the density of a number of liquid normal alkanes and their binary mixtures, over a wide range of

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temperature and pressure, provides the opportunity to examine existing correlation and prediction methods so as to determine a reliable method for the prediction of density of those fluids for which accurate experimental measurements are unavailable.

2. PREVIOUS WORK

Three main correlation methods have been recommended in the literature [8] for the prediction of compressed liquid densities: the graphical Lu Chart method [9] as revised by Ewbank and Harden [10], the Yen-Woods correlation [11], and the method of Chueh and Prausnitz [12, 13].

Rea et al. [14] expressed the revised and expanded Lu Chart [10] by a set of generalized polynomials to make it adaptable for computer calculation. For application of the other methods, they suggested the use of the modified Rackett equation [15] to predict liquid densities at saturated vapor pressures.

For these three methods, the temperature range of applicability is quite extensive, as shown in Table I, right up to the critical region apart from the Lu Chart method, but the pressure range over which these expressions have been tested is significantly smaller than that of industrial interest. Furthermore, the overall fit to experimental density data for *n*-alkanes is at best about 0.6% and the difference can exceed 10%. Moreover, the use of the critical compressibility factor, Z_c , when the specific constant, Z_{RA} , in the Rackett equation is not known, may lead to an error in density of the order of 30% [14].

Although these general methods would give better agreement by restricting attention to a given class of compounds such as, in this case, normal alkanes, it was decided to use a different approach, based on the widely used Tait equation [16], to develop an accurate prediction method

Table I. Comparison of Requirements and Range of Applicability of Methods Studied by Rea et al. [14]

	Lu Chart [14]	Yen and Woods [11]	Chueh and Prausnitz [13]
Input	T_c, P_c, ρ_{ref}	$T_c, P_c, Z_c, V_c, Z_{RA}$	T_c, V_c, ω, Z_{RA}
T range, T_R	0.50–0.76	0.3–1.0	0.4–0.98
P range, P_R	Sat.–3.0	0.2–60	Sat.–60

for liquid density at elevated pressures. This has the advantage that it is not dependent on input parameters such as the critical constants and Z_{RA} (Table I).

2.1. Tait Equation

The Tait equation is frequently expressed in the form:

$$(v_0 - v)/v_0 = C \log[(B + P)/(B + P_0)] \quad (1)$$

where v_0 and v are liquid volumes at the corresponding pressures, P_0 and P , and B and C are parameters.

In different applications, C has been taken either to be a constant for a given compound independent of temperature [17–21], to have the same value for a series of compounds [22–31], or to be a weak function of temperature [32–34]. Thomson et al. [35] generalized C for all fluids, by making it a linear function of the acentric factor.

Parameter B definitely varies with temperature, and this has been represented by a linear temperature dependence [20, 21] and by a quadratic equation [22–26]. Atanov [36] related B exponentially to the difference between the temperature and the critical temperature.

This same expression, used also by Golik et al. [37], leads to the result that at the critical temperature, B is equal to three times the critical pressure, P_c . However, Ginell [32] pointed out that B should equal $-P_c$ at this temperature and showed that this was supported by the results for helium. Liquid ammonia densities have been shown [34] to give a similar conclusion. The parameters in all these equations for B are specific for the compounds considered.

However, Thomson et al. [35] have given a general equation for B/P_c in terms of reduced temperature, T/T_c , and an acentric factor, which reduces to the value -1 at the critical temperature and which, with their generalized expression for C and the saturated liquid correlation of Hankinson and Thomson [38], allows a prediction of high-pressure densities to be made. Applications were generally limited to pressures up to 69 MPa. Even so, the discrepancies between the predicted densities and the observed values for hydrocarbons (Table II), even taking into consideration the errors in the estimation of the saturated-pressure density values, are so great that large errors will be introduced in using this method for the calculation of molar volumes for the prediction of high-pressure viscosities by the free-volume approach.

Table II. Percentage Error in Density Estimation Using the Thomson Form [35] of the Tait Equation

Compound	Average absolute percentage error	
	High pressure [35]	Saturated [38]
<i>n</i> -Hexane	1.45	0.32
2,2-Dimethylbutane	0.534	0.27
Benzene	0.902	0.06
Cyclohexane	0.444	0.07

3. THE CORRELATION

3.1. Experimental Data

Since the earlier correlation work for the densities of compressed liquids and liquid mixtures [14, 35], new measurements over a wider range of thermodynamic states have been reported. Dymond, Isdale, and their co-workers [39, 40] have reported data on *n*-hexane, *n*-octane, *n*-decane, *n*-dodecane, and *n*-hexadecane in the temperature range of 298.15 to 373.15 K at pressures up to 500 MPa. They claim the maximum uncertainty in all sets of measurements to be $\pm 0.2\%$. Kashiwagi and Makita [41] have also made compressed liquid density measurements recently for *n*-hexane, *n*-octane, *n*-decane, and *n*-dodecane in the same temperature range of 298.15 to 373.15 K but at lower pressures, up to 110 MPa. In all sets of measurements, the claimed uncertainty is expected to be better than $\pm 0.2\%$. The other reliable source of data for high-pressure liquid densities for *n*-heptane, *n*-nonane, *n*-undecane, *n*-tridecane, and *n*-heptadecane has been that of Doolittle [42], and the experimental uncertainty is about the same.

Initially, the temperature and the pressure ranges of the selected data were restricted. The temperature range was restricted to either the boiling point or 0.66 times the critical temperature. The pressure was restricted to 150 MPa for two reasons, the range of industrial interest and the uniformity in the range of pressure for mainly all selected sets. The details of the data used for the correlation are given in Table III. The selected data sets covered a reduced temperature range from 0.42 to 0.66 and a reduced pressure range from 0.27 to 114.

Table III. Description of the Selected Data Sets

Compound	No. of points	T_R	Max. P (MPa)	P_R	Ref. No.
<i>n</i> -Hexane	8	0.59–0.64	142.5	47.9	39
	72	0.59–0.66	111.8	37.1	41
<i>n</i> -Heptane	14	0.56–0.60	150.0	54.8	42
<i>n</i> -Octane	14	0.52–0.61	202.5	81.4	40
	78	0.52–0.61	111.8	43.0	41
<i>n</i> -Nonane	21	0.51–0.63	150.0	65.6	42
<i>n</i> -Decane	8	0.48–0.61	150.5	71.5	40
	75	0.48–0.56	111.1	52.8	41
<i>n</i> -Undecane	21	0.48–0.58	150.0	76.3	42
<i>n</i> -Dodecane	67	0.45–0.53	111.2	61.0	41
<i>n</i> -Tridecane	28	0.45–0.63	150.0	87.2	42
<i>n</i> -Hexadecane	17	0.42–0.52	154.2	108.7	39
<i>n</i> -Heptadecane	28	0.44–0.65	150.0	113.9	42

3.2. Correlation Development

In order to develop a correlation of the Tait form, each isotherm for normal alkanes varying from *n*-hexane to *n*-heptadecane was individually fitted by Eq. (1), rearranged in the form

$$(\rho - \rho_0)/\rho = C \log[(B + P)/(B + P_0)] \quad (2)$$

and the adjustable parameters C and B were optimized. It was observed that C can be definitely considered constant. The extensive, accurate data gave the result that C is equal to 0.2000 instead of the slightly higher values suggested by Eduljee et al. [27] and Cutler and co-workers [28], as shown in Table IV.

It was observed that the corresponding optimized B values had a reasonable flexibility limit of 3 to 4% on either side, which will still correlate the observed densities within experimental uncertainties. However, the flexibility decreases as the temperature increases toward $0.7T_c$. It was found that the B values for P - V isotherms for any normal alkane, when adjusted for the number of carbon atoms in the chain length, have a smooth variation with reduced temperature which can be given by

$$B_c = 341.539 - 734.292T_R + 411.189T_R^2 \quad (3)$$

where B_c is the B value adjusted for the length of the carbon chain using the simple expression

$$B_c = B + (C_n - 6) \quad (4)$$

Table IV. Comparison of the Tait Equation Representation Using $C = 0.2000$, $C = 0.2058$ [28], and $C = 0.2172$ [27]^a

Liquid	Temperature (K)	$C = 0.2000$		$C = 0.2058$		$C = 0.2172$	
		AAD (%)	Max. dev. (%)	AAD (%)	Max. dev. (%)	AAD (%)	Max. dev. (%)
<i>n</i> -Heptane [42]	303.15	0.03	0.06	0.05	0.10	0.09	0.25
	323.15	0.03	0.07	0.06	0.15	0.10	0.27
<i>n</i> -Undecane [42]	303.15	0.04	0.10	0.05	0.16	0.08	0.27
	323.15	0.03	0.09	0.05	0.16	0.09	0.29
	373.15	0.01	0.04	0.04	0.11	0.08	0.29
	423.15	0.06	0.10	0.09	0.15	0.15	0.41
<i>n</i> -Heptadecane [42]	323.15	0.06	0.19	0.07	0.24	0.10	0.34
	373.15	0.07	0.19	0.09	0.26	0.12	0.40
	423.15	0.05	0.12	0.08	0.22	0.13	0.42
	473.15	0.03	0.09	0.02	0.06	0.10	0.30

^a Maximum deviation % = $|\rho_{\text{expt}} - \rho_{\text{calc}}| 100 / \rho_{\text{expt}}$. AAD% = $\sum_i |\rho_{\text{expt}} - \rho_{\text{calc}}| 100 / \rho_{\text{expt}} / n$, where n is the total number of data points.

C_n is the characteristic carbon number, which equals the actual number of carbon atoms in the case of normal alkanes. The values of B_c are plotted against reduced temperature in Fig. 1.

3.3. Mixtures

The Tait equation has been extended to include the high-pressure density data of binary and ternary mixtures of *n*-alkanes. It has been observed that C can still be considered constant at 0.2000. B can be determined from the one-fluid approximation with $C_{n,\text{mix}}$, the carbon number for the equivalent *n*-alkane:

$$C_{n,\text{mix}} = \sum_i x_i C_{ni} \quad (5)$$

The critical temperature, T_c , required in Eq. (3) is considered the same as that for an *n*-alkane having a carbon number equivalent to $C_{n,\text{mix}}$. This will of course involve interpolation where $C_{n,\text{mix}}$ is noninteger. Systems with the same carbon number are thus expected to exhibit identical pressure dependences of density.

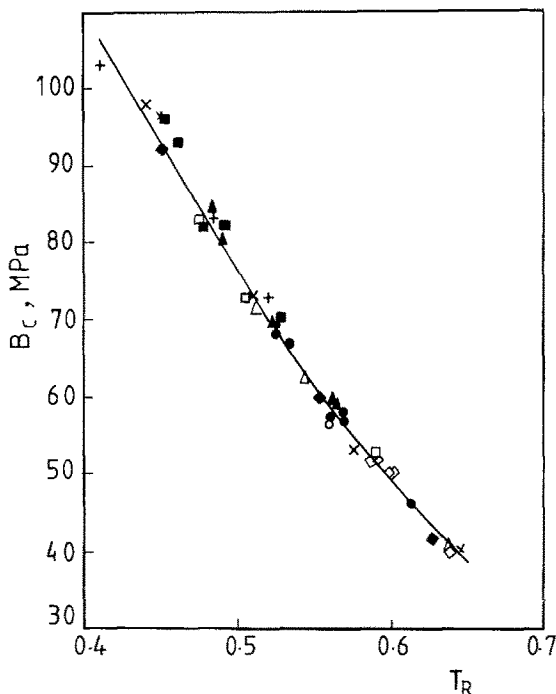


Fig. 1. Plot of B_c against T_R . Solid line, Eq. (3); \diamond , *n*-hexane; \circ , *n*-heptane; \bullet , *n*-octane; \triangle , *n*-nonane; \blacktriangle , *n*-decane; \square , *n*-undecane; \blacksquare , *n*-dodecane; \blacklozenge , *n*-tridecane; $+$, *n*-hexadecane; \times , *n*-heptadecane.

4. RESULTS

4.1. Pure Liquids

A comparison between densities predicted on the basis of the Tait equation, with C equal to 0.2000 and B given by Eqs. (3) and (4), and experimental values is given in Table V, which also lists the number of data points in each pure-compound data set and the temperature and pressure range covered. The analysis shows that the present correlation can fit all the *n*-alkane data, comprising 1394 data points, with an average absolute percentage deviation of 0.09%.

The maximum percentage deviations in density for individual isotherms for the hydrocarbons from *n*-hexane to *n*-heptadecane are plotted in Fig. 2. In only a few cases does this maximum percentage deviation exceed 0.2%, and this nearly always occurs at the highest pressures. Such data comprise only 3% of the total data considered in the study.

Although this correlation has been developed with the high-pressure density data on *n*-alkanes from *n*-hexane to *n*-heptadecane (Table III), it will correlate very satisfactorily the high-pressure densities of lower alkanes also, as shown in Table V.

An evaluation of the present Tait correlation [Eqs. (2)–(4)] together with other recommended correlations [14] is presented in Table VI, based

Table V. Tait Correlation for the *n*-Alkanes^a

Compound	No. of points	Temp. range, T_R	Max. press (MPa)	Max. % dev.	Av. abs. % dev.	Ref. No.
Ethane	50	0.33–0.49	32.1	0.18	0.08	44
Propane	50	0.24–0.38	31.5	0.11	0.05	45
<i>n</i> -Butane	62	0.33–0.59	32.2	0.23	0.14	43
<i>n</i> -Hexane	8	0.59–0.64	144.5	0.21	0.11	39
	28	0.44–0.59	126.8	0.21	0.09	46
	72	0.59–0.66	111.8	0.22	0.09	41
<i>n</i> -Heptane	14	0.56–0.59	150.0	0.24	0.10	42
	71	0.55–0.65	111.6	0.24	0.11	41
	47	0.51–0.68	117.7	0.09	0.04	48
<i>n</i> -Octane	13	0.52–0.61	152.2	0.15	0.06	40
	78	0.52–0.61	111.4	0.12	0.05	41
	47	0.53–0.69	117.7	0.14	0.07	48
<i>n</i> -Nonane	21	0.51–0.63	150.0	0.19	0.08	42
	91	0.51–0.71	140.0	0.19	0.08	49
	48	0.51–0.66	117.7	0.27	0.12	48
<i>n</i> -Decane	75	0.48–0.56	111.1	0.09	0.04	41
	8	0.48–0.60	150.5	0.33	0.16	40
	109	0.48–0.58	490.1	0.27	0.10	47
<i>n</i> -Undecane	21	0.48–0.58	150.0	0.15	0.07	42
<i>n</i> -Dodecane	67	0.45–0.53	111.2	0.22	0.13	41
	31	0.45–0.57	441.9	0.24	0.10	40
	91	0.45–0.54	416.9	0.18	0.09	47
	24	0.46–0.60	117.7	0.31	0.14	48
<i>n</i> -Tridecane	28	0.45–0.63	150.0	0.19	0.06	42
<i>n</i> -Tetradecane	113	0.43–0.52	366.8	0.12	0.05	47
<i>n</i> -Hexadecane	17	0.42–0.52	154.2	0.18	0.08	39
	89	0.42–0.50	290.2	0.29	0.13	47
<i>n</i> -Heptadecane	21	0.44–0.65	150.0	0.34	0.11	42

^a Total data points, $n = 1394$. Av. abs. % error = 0.09 %.

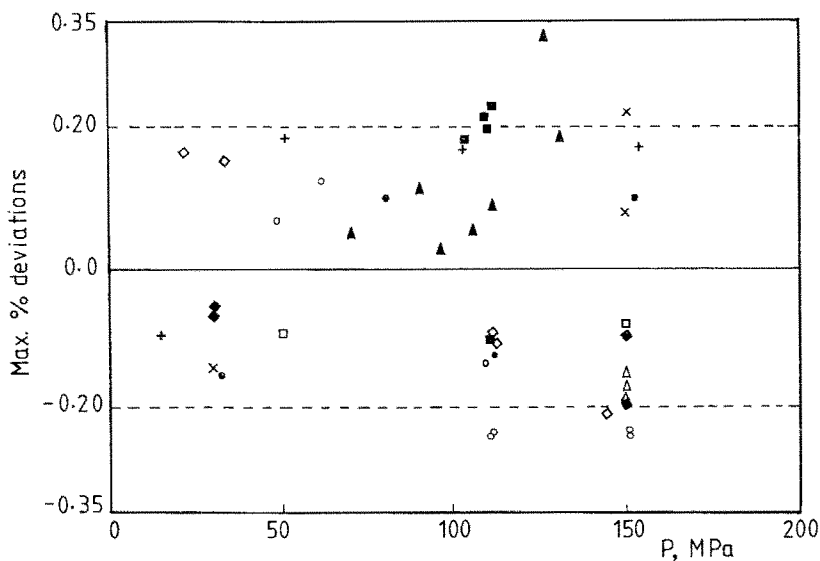


Fig. 2. Plot of the maximum percentage deviations in ρ for individual PV isotherms using Eqs. (2)–(4). Key as in the legend to Fig. 1.

on the same high-pressure density data of n-alkanes measured by Doolittle [42]. Modification of the Yen–Woods and Chueh and Prausnitz correlations using the Rackitt equation with Z_c gives a substantially poorer fit for these compounds. It is observed that the correlation developed here is applicable beyond its recommended temperature range and at pressures up to 150 MPa with relatively little loss in accuracy compared with the discrepancies found by Rea et al. [14] with the other methods at pressures limited to 50 or 100 MPa. These discrepancies may be reduced in the case of the Yen–Woods and Chueh–Prausnitz methods on using experimental densities at saturation rather than predicted values, but only by about 1%.

It has been found that the general correlation suggested by Thomson et al. [35] (referred to by the name COSTALD) is more reliable than the previously recommended correlations [8]. This is confirmed in Table VII for n-alkane densities, where a comparison is made among the present correlation, the COSTALD method [35], and that of Yen and Woods [14], using the same data sets. Only a slight improvement in the fit ($<0.3\%$) is obtained by the other methods with experimental densities at 0.101 MPa.

The modified corresponding-states procedure TRAPP of Ely and Hanley [50] for the prediction of densities (and viscosities) based on methane as reference fluid is of limited applicability. Higher n-alkanes exist

Table VI. Summary of Evaluation of Correlations

Compound	No. of data points	Temperature range of data (K)	Pressure range of data (MPa)	Average absolute percentage deviation				
				This work, Tait equation	Regression of Lu Chart [14]	Yen and Woods [14]	Chueh and Prausnitz [14]	
<i>n</i> -Undecane [42]	21 ^a	303-373	5-150	0.07				
	42	303-523	5-150	0.37				
	42	303-573	5-100		2.09	2.47	5.26	
<i>n</i> -Tridecane [42]	28 ^a	303-423	5-150	0.06				
	42	303-523	5-150	0.15				
	42	303-573	5-100		1.91	4.22	8.91	
<i>n</i> -Heptadecane [42]	21 ^a	323-423	5-150	0.07				
	42	323-573	5-150	0.43				
	30	323-573	5-50		2.85	4.64	14.05	

^a This work: data analyzed in the recommended temperature range, i.e., at temperature $T < T_B$ or $T < 0.66 T_C$, whichever is smaller.

Table VII. Comparison of Test Results with Those of Thomson et al. [35]

n-Alkanes	No. of points	Temp. (K)	Max. press (MPa)	Average absolute error (%)			Ref. No.
				Yen Woods	COSTALD	This work	
<i>n</i> -Hexane	28	223–298	150			0.09	46
	20		69	1.71	1.45		
<i>n</i> -Decane	109	298–358	490			0.10	47
	51		94	1.20	0.81		
<i>n</i> -Dodecane	91	298–358	417			0.09	47
	48		94	1.09	0.49		
<i>n</i> -Tetradecane	113	298–358	113			0.05	47
	66		94	1.18	0.88		
<i>n</i> -Hexadecane	89	298–358	290			0.13	47
	19		69	1.82	0.15		

in the liquid state at reduced temperatures well below that at which methane freezes, but density calculations by this method are not possible at these temperatures. Where a comparison with experimental data is possible, significant differences are found, for example, the average absolute percentage deviation of predicted *n*-hexane densities from measured values [39] is greater than 0.4% at pressures up to 150 MPa. These differences exceed 1% at higher pressures.

The method described in this paper fits the density data up to high pressures with a much higher accuracy. Although the pressure range considered was restricted to 150 MPa, it should be noted, as shown in Tables V and VII, that the data for certain hydrocarbons can be fitted to well within an average absolute percentage deviation of 0.2% up to much higher pressures, even close to 500 MPa in some instances.

4.2. Mixtures

A summary of the results for correlation of the density data of binary and ternary *n*-alkane mixtures is given in Table VIII. The agreement with the experimental data is generally within 0.2%, although slightly larger discrepancies are found at temperatures above 0.6 times the critical temperature. However, this agreement is very encouraging, considering that there are no adjustable parameters when the correlation is applied to mixtures.

Table VIII. Tait Correlation for the Mixtures^a

Mixtures	No. of points	Temp. range, T_R	Max. press (MPa)	Max. % dev.	Av. abs. % dev.	Ref. No.
0.2 C6 + 0.8 C16	15	0.43–0.54	150	0.20	0.10	51
0.4 C6 + 0.6 C16	14	0.45–0.57	145	0.20	0.09	51
0.6 C6 + 0.4 C16	8	0.48–0.60	100	0.17	0.10	51
0.8 C6 + 0.2 C16	14	0.52–0.66	143	0.49	0.15	51
0.5 C6 + 0.5 C12	7	0.50–0.59	150	0.22	0.14	40
0.2 C6 + 0.8 C10	2	0.41	100	0.42	0.41	52
0.4 C6 + 0.6 C10	2	0.51	100	0.20	0.13	52
0.5 C6 + 0.5 C10	2	0.52	100	0.10	0.07	52
0.6 C6 + 0.4 C10	2	0.54	100	0.07	0.04	52
0.8 C6 + 0.2 C10	2	0.56	100	0.10	0.05	52
0.5 C8 + 0.5 C12	12	0.48–0.60	150	0.44	0.15	40
0.2 C8 + 0.8 C12	2	0.46	100	0.05	0.04	52
0.4 C8 + 0.6 C12	2	0.48	100	0.05	0.03	52
0.5 C8 + 0.5 C12	2	0.48	100	0.08	0.05	52
0.6 C8 + 0.4 C12	2	0.49	100	0.12	0.08	52
0.8 C8 + 0.2 C12	2	0.51	100	0.02	0.01	52

^a Binary mixtures: total data points = 287; ADD = 0.09%. Ternary mixture: total data points = 87; AAD = 0.04%.

Table VIII. (Continued)

Mixtures	No. of points	Temp. range, T_R	Max. press (MPa)	Max. % dev.	Av. abs. % dev.	Ref. No.
0.5 C10+ 0.5 C16	7	0.52-0.55	130	0.29	0.15	40
0.5 C10+ 0.5 C14	100	0.45-0.54	430	0.31	0.11	53
0.2 C10+ 0.8 C14	2	0.44	100	0.04	0.04	52
0.4 C10+ 0.6 C14	2	0.45	100	0.009	0.008	52
0.5 C10+ 0.5 C14	2	0.45	100	0.06	0.05	52
0.6 C10+ 0.4 C14	2	0.46	100	0.09	0.08	52
0.8 C10+ 0.2 C14	2	0.47	100	0.28	0.28	52
0.5 C12+ 0.5 C16	80	0.43-0.52	331	0.12	0.05	53
0.6 C10+ 0.2 C14+ 0.2 C16	87	0.45-0.54	359	0.10	0.04	53

5. CONCLUSIONS

It is shown that the Tait equation in the form $(\rho - \rho_0)/\rho = C \log[(B + P)/(B + P_0)]$, where subscript 0 refers to 0.101 MPa, C is equal to 0.2000, and B , adjusted for the number of carbon atoms in the alkane chain, depends only on reduced temperature, gives a very satisfactory correlation of n-alkane densities at pressures up to 150 MPa and at temperatures up to 0.66 times the critical temperature or the boiling point, whichever is lower. By a simple determination of the equivalent carbon number, this equation can be successfully applied to n-alkane mixtures as well. In the absence of experimental measurements, this method is recommended for use in the accurate prediction of densities for liquid n-alkanes and their multicomponent mixtures at elevated pressures with experimental values for the density at 0.101 MPa.

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