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Equation of State for Nonpolar Fluids: Prediction from Boiling Point Constants

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A new corresponding states correlation for the second virial coefficient of nonpolar fluids in terms of the boiling point constants is presented. The scaling constants are the normal boiling point temperature, T_{bp}, which is used to form a dimensionless temperature and the liquid density at the normal boiling point, ρ_{bp}, which is used to form a dimensionless second virial coefficient. The procedure has been examined for a large number of substances including noble gases, diatomic molecules, saturated hydrocarbons up to C_8, and a number of aliphatic, aromatic, and cyclic hydrocarbons. The resulting correlation has been applied to predict the equation of state of fluids over the range from the vapor-pressure curve to the freezing curve at various temperatures from the triple point up to the nonanalytical critical region. The equation of state has been applied to reproduce the liquid density of a great number of compounds both in the saturation and compressed states, at temperatures up to 2000 K and pressures up to 10000 bar, within an accuracy of a few percent. In particular we have shown that knowledge of two readily measurable constants is sufficient to determine the *pvT* surface of pure normal fluids having a variety of structural complexities.

KEY WORDS: compressed liquids; corresponding states; equation of state; gases; second virial coefficient.

1. INTRODUCTION

The prediction of bulk properties of matter from molecular level models appears to be a well-known science. Statistical-mechanical perturbation theories of fluids have been developed over the past years [1, 2] based on the recognition that the structure of a liquid is determined primarily by repulsive forces, so that fluids of hard bodies can serve as useful reference

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systems. An accurate analytical equation of state is presented by Ihm et al. [3], which is based on the statistical-mechanical perturbation theory of the liquid state in the perturbation scheme of Weeks–Chandler–Anderson [2]. In the final form, the equation of state is

$$\frac{p}{\rho kT} = 1 + \frac{(B_2 - \alpha)\,\rho}{1 + 0.22\lambda b\rho} + \alpha\rho G(b\rho) \tag{1}$$

where p is the pressure, ρ is the density, kT is the thermal energy per molecule, B_2 is the second virial coefficient, α is a parameter that accounts for repulsive forces, b is the van der Waals covolume, $G(b\rho)$ is the pair distribution function for hard-sphere bodies at contact [4], and λ is the slope of $[G(b\rho)]^{-1}$ as a function of $b\rho$. In Eq. (1), the characteristic parameters B_2, α, and b all are temperature-dependent. $\alpha(T)$ and $b(T)$ vary slightly with temperature and do not depend on the details of the potential function used for their calculation. $B_2(T)$ depends highly on temperature and serves as an interaction coefficient of the system in the equation of state.

The values of $B_2(T)$, $\alpha(T)$, and $b(T)$ can be calculated from the potential function by integration. If an accurate potential function is not known, α and b can be calculated from the experimental second virial coefficient by means of a two-constant scaling rule [5]. This can be done because α and b depend on the intermolecular repulsive forces and are relatively insensitive to the details of the intermolecular potential energy function. Thus the second virial coefficient serves to predict the entire equation of state for fluids in terms of two scaling constants, over the whole range of temperatures and pressures, excluding the nonanalytical critical point and the two-phase region.

A new strong principle of corresponding states is contained in Eq. (1), and this principle has led to an appreciable accuracy and simplicity [6]. The principle is that the plot of $[G(b\rho)]^{-1}$ vs. $b\rho$ is a linear function, with the slope of λ, a free parameter that is known to compensate for any uncertainties in the second virial coefficient, i.e.,

$$[G(b\rho)]^{-1} = \alpha\rho\left[Z - 1 + \frac{(\alpha - \beta_2)\,\rho}{1 + 0.22\lambda b\rho}\right]^{-1} = 1 - \lambda b\rho \tag{2}$$

where $Z \equiv p/\rho kT$ is the compressibility factor [3]. Substituting the above functional form for $G(b\rho)$ in Eq.(1), Ihm et al. [3] obtained

$$\frac{p}{\rho kT} = 1 - \frac{(\alpha - B_2)\,\rho}{1 + 0.22\lambda b\rho} + \frac{\alpha\rho}{1 - \lambda b\rho} \tag{3}$$

The purpose of this paper is to show how the equation of state can be used with even less input information. In particular, we have shown that knowledge of two constants is sufficient to determine the pvT surface of a pure normal fluid with an accuracy of a few percent. The two scaling constants are the normal boiling point temperature and liquid density.

2. CORRELATION

The minimum input information needed to use Eq. (3) consists of experimental values of the second virial coefficient plus some high density data to fix the value of an adjustable constant, λ, in $G(b\rho)$. The second virial coefficient plays a central role in Eq. (3) in that it is used both directly and indirectly, as a source of scaling constants for the calculation of $\alpha(T)$ and $b(T)$ [5]. Although accurate direct measurements of B_2 are best, in their absence there are several correlation schemes, usually based on the law of corresponding states, by which B_2 can be estimated with reasonable accuracy.

Statistical theory [7] shows that a group of substances will conform to the principle of corresponding states only if the intermolecular potentials are identical except for distances and energy-scale parameters characteristic of each substance. Also quantum effects and multibody effects must be negligible. Various types of molecular shapes and molecular dipole moments might be expected to cause different deviations from macroscopic properties of simple fluids. It is found, however, that the reduced second virial coefficients for a wide variety of molecular types are nearly identical functions of reduced parameters. Generally two scaling constants, a temperature constant and a volume constant, are needed to estimate the second virial coefficient from the corresponding states correlation. The temperature constant, which is a measure of the strength of intermolecular forces, is used to form a dimensionless temperature, and the volume constant is used to form a dimensionless second virial coefficient.

A simple reduced equation for the second virial coefficient is proposed by Berthelot [8], which uses the critical temperature, T_C, as the temperature reducing constant and the pseudocritical molar volume, RT_C/p_C, as the volume reducing constant. The correlation reproduces the properties of real gases at low pressures to the ideal gas state. Pitzer and Curl [9] showed that the introduction of the acentric factor, ω, as an additional parameter increases the accuracy of the correlation, so that plots of $B_2 p_C/RT_C$ versus T_C/T form a universal family of curves indexed by the third parameter, ω. It should be noted that the critical volume, v_C, is subject to higher experimental inaccuracy than that of T_C and p_C. Therefore, it is recommended to use the pseudocritical volume, $B_2 p_C/RT_C$, instead

[10]. The values of these constants are known for many substances [11], but for some liquids T_C and p_C are not known, either because they have not been measured or because the liquid decomposes at high temperatures. Boushehri and Mason [12] introduced the latent heat of vaporization and the liquid density at the triple point or freezing point as two scaling constants for correlating second virial coefficient data. A new correlation is also proposed by Boushehri and Ghatee [13] based on the surface tension and the liquid density at the freezing point.

In this work, we show that two easily measurable scaling constants, namely the liquid density at the normal boiling point as the volume constant and the normal boiling point temperature as the temperature constant, are sufficient for correlating second virial coefficients of normal fluids, ranging in complexity from noble gases, $\omega = 0$, to hydrocarbons up to C_8, $\omega = 0.398$.

We have checked experimental data to see whether a dimensionless plot of $B_2 \rho_{bp}$ as a function of T_{bp}/T results in a universal curve or, at least, a close family of curves that can be indexed by a third constant. We have chosen 25 normal fluids having a variety of structural types for testing the present prediction scheme. The fluids examined can be classified into the following seven groups for convenience:

Noble gases: Ne, Ar, Kr, Xe

Diatomic molecules: N_2, O_2, CO

C_1–C_4 alkanes: CH_4, C_2H_6, C_3H_8, n-C_4H_{10}, i-C_4H_{10}

C_5–C_8 alkanes: n-C_5H_{12}, i-C_5H_{12}, n-C_6H_{14}, n-C_7H_{16}, n-C_8H_{18}

Aliphatic hydrocarbons: propene, 1-butene, cis-2-butene, trans-2-butene

Aromatic hydrocarbons: C_6H_6, $C_6H_5CH_3$

Cyclic hydrocarbons: cyclo-C_5H_{10}, cyclo-C_6H_{12}.

Figure 1 shows that a single curve serves to correlate a wide variety of nonpolar fluids with sufficient accuracy (see Section 3). The smoothed values of the second virial coefficient recommended by Dymond and Smith [14] have been used to construct Fig. 1. In the cases where smoothed values are not cited, the other sources compiled by Dymond and Smith [14] are used. Therefore, we have used the second virial coefficients by Michels et al. [15] for CO, by Strein et al. [16] for i-C_4H_{10}, by Al-Bizreh et al. [17] for n-C_7H_{16} and n-C_8H_{18}, by Roper [18] for cis- and trans-C_4H_{10}, by Kolysko et al. [19] for toluene, and by McGlashan et al. [20] for 1-butene and cyclo-C_5H_{10}. Examination of the results plotted in Fig. 1 shows that it is not necessary to index the curves by the third parameter ω for our purpose. Apparently the shape effects described by the acentric factor affect the values of T_{bp} and ρ_{bp} to compensate for their influence on the second

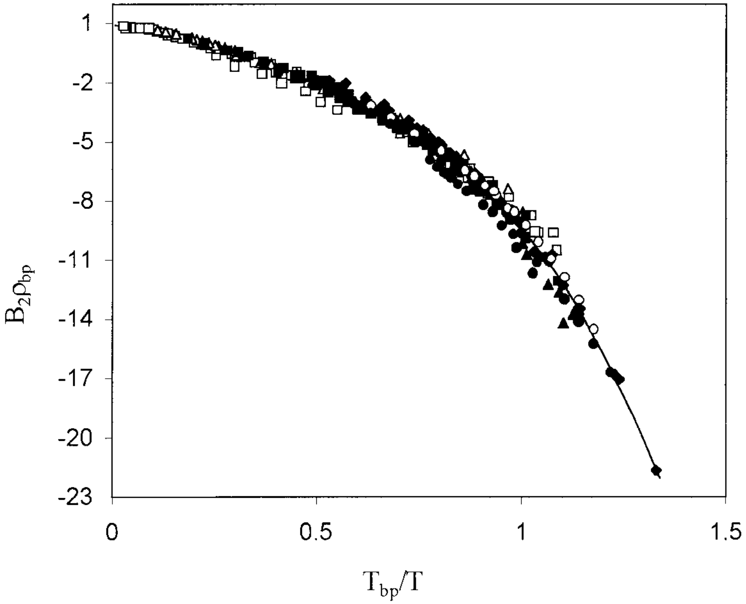

Fig 1. Correlation plot for the reduced second virial coefficient of (□) Ne, Ar, Kr, Xe; (△) O_2, N_2, CO; (■) CH_4, C_2H_6, C_3H_8, n-C_4H_{10}, i-C_4H_{10}; (◆) n-C_5H_{12}, i-C_5H_{12}, n-C_6H_{14}, n-C_7H_{16}, n-C_8H_{18}; (▲) propene, 1-butene, cis-2-butene, trans-2-butene; (●) C_6H_6, $C_6H_5CH_3$; (○) cyclo-C_5H_{10}, cyclo-C_6H_{12}. The markers are experimental data [14–20] and the solid line represents the best polynomial fit, Eq. (4). The same marker is used for each group listed in Table II.

virial coefficient. Our resulting correlation for B_2 can be represented by the empirical expression

$$B_2\rho_{\mathrm{bp}} = 1.033 - 3.0069\left(\frac{T_{\mathrm{bp}}}{T}\right) - 10.588\left(\frac{T_{\mathrm{bp}}}{T}\right)^2 + 13.096\left(\frac{T_{\mathrm{bp}}}{T}\right)^3 - 9.8968\left(\frac{T_{\mathrm{bp}}}{T}\right)^4 \tag{4}$$

which is shown as the solid curve in Fig. 1. A total of 296 points are included in Fig. 1, and the R-squared value for the fit is 0.9891. Equation (4) can be used to predict B_2 over the entire temperature range cited in Ref. 14. Also shown in Fig. 2 is the standard two-parameter correlation, $B_2 p_C/RT_C$ as a function of T/T_C, for the sake of comparison. It is clear that the present correlation is more accurate than the correlation based on the critical constants.

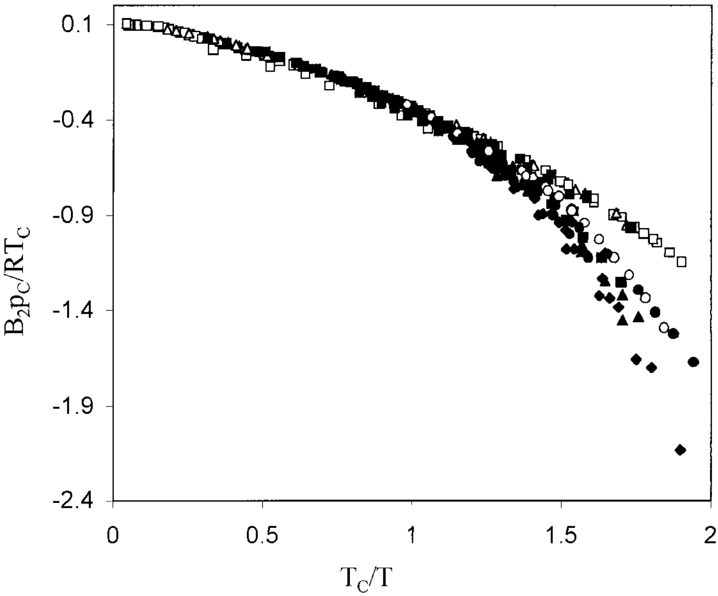

Fig. 2. Same as Fig. 1 for B_2p_C/RT_C versus T/T_C.

Because α and b are insensitive to the detailed shape of the intermolecular potential, they can be calculated if B_2 is known, by means of some effective mean-spherical potential [5, 6]. The results of such calculations are conveniently recorded [3] as the dimensionless quantities α/V_B and b/V_B, which are almost universal functions of the dimensionless temperature T/T_B. Here the scale factors T_B and V_B are the Boyle temperature at which $B_2 = 0$, and the Boyle volume, $V_B = T_B(dB_2/dT)_{TB}$, which can be expressed in terms of the boiling point parameters as $T_B = 4.629T_{bp}$ and $V_B = 1.3277/\rho_{bp}$. The empirical equations given in Ref. 6 for α/V_B and b/V_B as a function of T/T_B, using an LJ (12-6) potential, can then be rescaled by T_{bp} and ρ_{bp}, instead of T_B and V_B as:

$$\alpha\rho_{bp} = a_1\left\{\exp\left[-c_1\left(\frac{T}{T_{bp}}\right)\right]\right\} + a_2\left\{1-\exp\left[-c_2\left(\frac{T_{bp}}{T}\right)^{1/4}\right]\right\} \tag{5}$$

and

$$b\rho_{bp} = a_1\left[1-c_1\left(\frac{T}{T_{bp}}\right)\right]\exp\left[-c_1\left(\frac{T}{T_{bp}}\right)\right] + a_2\left\{1-\left[1+0.25c_2\left(\frac{T_{bp}}{T}\right)^{1/4}\right]\exp\left[-c_2\left(\frac{T_{bp}}{T}\right)^{1/4}\right]\right\} \tag{6}$$

Table I. Comparison of the Calculated Boyle Parameters with the Values Determined from Tabulated Data in Ref. 14

	T_B (K)			V_B ($cm^3 \cdot mol^{-1}$)		
Substance	Cal.	Expt.	D (%)	Cal.	Expt.	D (%)
Ne	125.4	127.1	−1.3	22.24	22.44	−0.80
Ar	404.1	409.1	−0.2	37.98	43.60	−12.88
Kr	554.5	579.8	−4.3	45.98	51.10	−10.02
Xe	764.5	774.5	−1.3	58.99	72.30	−18.41
N_2	358.1	321.0	11.5	46.11	55.69	−17.2
O_2	417.5	409.4	2.0	37.23	41.87	−11.08
CO	378.0	342.0	10.5	47.14	56.21	−16.13
CH_4	515.7	508.7	1.4	50.19	54.55	−8.00

where

$$a_1 = -0.0860, \qquad c_1 = 0.5624$$

$$a_2 = 2.3988, \qquad c_2 = 1.4267$$

For the sake of comparison, we have calculated the Boyle parameters for several compounds and compared the results with values determined from tabulated data in Ref. 14. The results are shown in Table I. It is worth mentioning that the Boyle volume involves the first derivative of the second virial coefficient with respect to temperature, and, hence, the errors become exaggerated. For the other fluids, listed in Table II, the experimental second virial coefficient data do not extend to high enough temperatures to allow the calculation of Boyle parameters.

To determine the value of the constant λ, at least one experimental high-density pvT point is needed. For consistency, to find λ we have used normal boiling point pvT data. Once this is done, the parameter λ can be calculated using Eqs. (3)–(6). The right hand side of Eq. (3) contains parameters $B_2\rho$, $\alpha\rho$, $b\rho$, and λ. According to Eqs. (4)–(6), the former three parameters have the same value at the normal boiling point for different fluids. The left hand side, Z, is very small compared with unity for the liquid phase at the normal boiling point, but varies very slightly for different fluids. Therefore, the parameter λ must have nearly the same value for all fluids. Calculation of λ at the normal boiling point leads to the value of 0.495, close to that obtained for noble gases, 0.454, by Ihm et al. [3]. Equation (3) therefore simplifies to the more convenient form

$$\frac{p}{\rho kT} = 1 - \frac{(\alpha - B_2)\,\rho}{1 + 0.11b\rho} + \frac{\alpha\rho}{1 - 0.495b\rho} \tag{7}$$

Table II. Calculated Results for the Density of Nonpolar Fluids Compared with Experiment as Average Absolute Deviation (AAD) and Maximum Deviation (MD) Percentage

Substance	T_{bp} (K)	ρ_{bp} ($\mathrm{mol \cdot L^{-1}}$)	ΔT (K)	Δp^a (bar)	AAD (%)	MD (%)	NP^b	Ref.
Ne	27.09	59.690	25–300	0–200	2.6	4.2	135	22
Ar	87.29	34.953	83.8–1300	0–5000	1.5	5.5	105	23
Kr	119.78	28.875	116–1300	0–1000	2.3	5.8	98	22
Xe	165.15	22.504	161.5–1300	0–1000	2.8	5.9	102	22
N_2	77.35	28.794	63–2000	0–10000	2.8	9.3	104	24
O_2	90.19	35.663	54.5–300	0–800	2.4	7.9	121	25
CO	81.65	28.167	71–1800	0–800	2.83	6.3	114	22
CH_4	111.42	26.456	91–800	0–1000	1.9	7.9	84	22
C_2H_6	184.52	18.174	90–400	0–500	0.7	5.6	60	22
C_3H_8	231.10	13.199	190–500	0–600	0.8	5.7	68	22
n-C_4H_{10}	272.65	10.341	213–510	0–600	1.3	2.7	78	22
i-C_4H_{10}	261.42	10.224	203–393	0–200	1.2	2.5	67	22
n-C_5H_{12}	309.22	8.456	183–423	0–200	1.0	6.2	59	22
i-C_5H_{12}	301.00	8.477	173–413	0–15.5	1.2	4.6	25	22
n-C_6H_{14}	341.89	7.118	193–483	0–300	2.3	9.0	56	22
n-C_7H_{16}	371.78	6.127	183–573	0–1500	3.6	9.2	42	22
n-C_8H_{18}	398.81	5.353	273–570	0–500	3.8	5.6	44	22
Propene	225.45	14.464	123–250	0–215	0.9	2.4	41	22
1-Butene	266.89	10.764	173–444	0–680	1.7	3.8	54	22
cis–2-Butene	276.87	11.422	223–283	0–1.3	0.1	0.5	7	22
trans–2-Butene	274.03	11.157	223–283	0–1.4	0.1	0.4	7	22
C_6H_6	353.25	10.529	278.5–600	0–500	1.7	4.7	69	22
$C_6H_5CH_3$	383.77	8.457	173–343	0–0.3	0.4	2.4	18	22
Cyclo-C_5H_{10}	322.41	10.219	223–373	0–4.1	0.2	0.5	16	22
Cyclo-C_6H_{12}	353.89	8.544	283–353	0–1	0.2	0.6	8	22

[a] The lower pressure limit is the vapor pressure at the lower temperature reported.
[b] The number of points considered.

which is the major result of this work. Since there are not parameters such as the acentric factor [9], the latent heat of vaporization [12], or the surface tension [13] in the correlation of Eqs. (4)–(6), the reduced quantities $B_2\rho_{\mathrm{bp}}$, $\alpha\rho_{\mathrm{bp}}$, and $b\rho_{\mathrm{bp}}$ are all independent of substance. This is the reason that the parameter λ has the same value for all substances. Theoretically, λ is a fluid-dependent property [21], but in the current empirical treatment, it is sufficient to regard it as a constant. The scaling constants, T_{bp} and ρ_{bp}, may change in such a way to compensate for the shape effects in λ. The present method for determining λ, 0.495, reproduces pvT results which are almost within $\pm 5\%$ of experimental values [22–25].

3. RESULTS

We can now check for the strong principle of corresponding states, Eq. (2), obtained by Ihm et al. [3]. According to this principle, for all the pvT data of fluids, except the nonanalytical critical region, the graph of $[G(b\rho)]^{-1}$ versus $b\rho$ should cover a straight line given by $1-\lambda b\rho$, or, in our method, $1-0.495b\rho$. The parameter $b\rho$ in Eq. (2) can be written as a product of dimensionless quantities, $b\rho_{\mathrm{bp}}$ and ρ/ρ_{bp}, and similarly for $B_2\rho$ and $\alpha\rho$. Such a graph is shown in Fig. 3 for a number of substances listed in Table II. The points in Fig. 3 fall along a single line of slope -1 shown by the solid line. The small scatter is not surprising, since the underlying theory is based on the perturbation treatment of the statistical mechanics for fluids of hard convex bodies [3]. The experimental density region that is covered in Fig. 3 ranges along the vapor-pressure curve at various temperatures from the triple point up to the critical region.

Equation (7) has been used to calculate the density of substances listed in Table II both in the saturation and compressed states. The results are

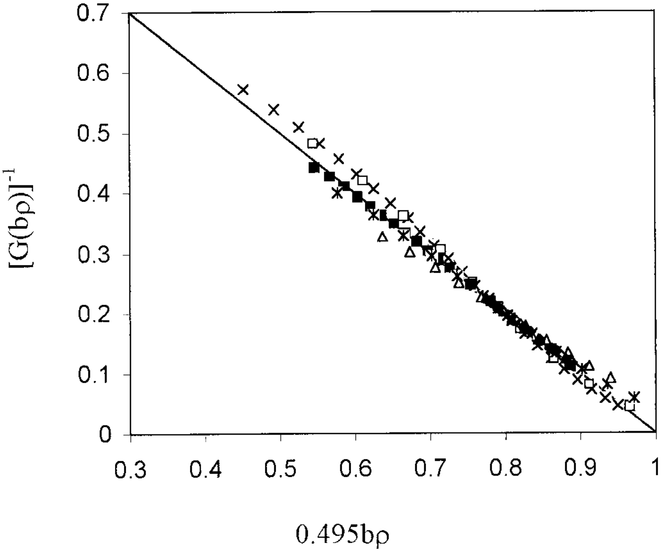

Fig. 3. Correlation plot of $[G(b\rho)]^{-1}$ vs. $0.495b\rho$ for (◆) Xe, (×) O_2, (□) C_2H_6, (●) n-C_4H_{10}, (∗), i-C_5H_{12}, (△) n-C_8H_{18}, (■) C_6H_6, (◇) cyclo-C_6H_{12}, (○) cis-2-butene, (▲) trans-2-butene. The markers are from tabulated pvT values [22–25] and the solid line is $1-0.495b\rho$.

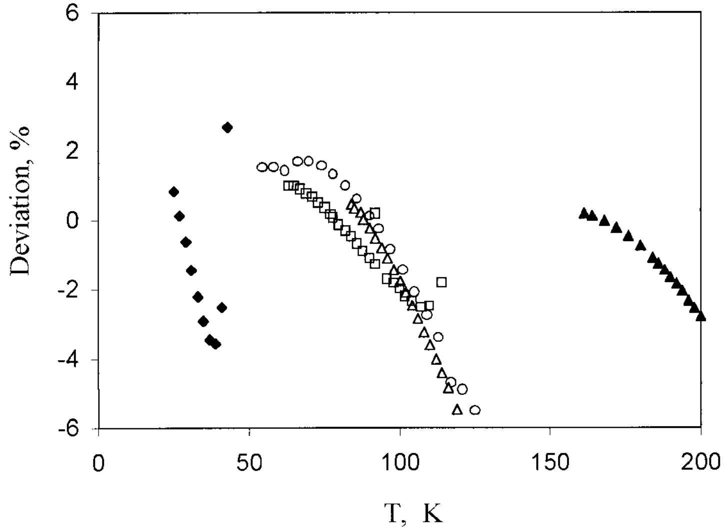

Fig. 4. Deviation plot for the density of (◆) Ne, (△) Ar, (▲) Xe, (○) O_2, and (□) N_2.

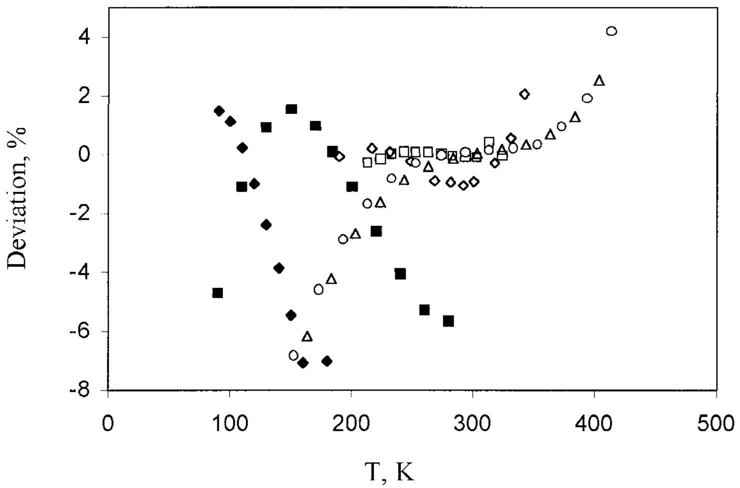

Fig. 5. Same as Fig. 4 for (◆) CH_4, (■) C_2H_6, (◇) C_3H_8, (□) n-C_4H_{10}, (△) n-C_5H_{12}, (○) i-C_5H_{12}.

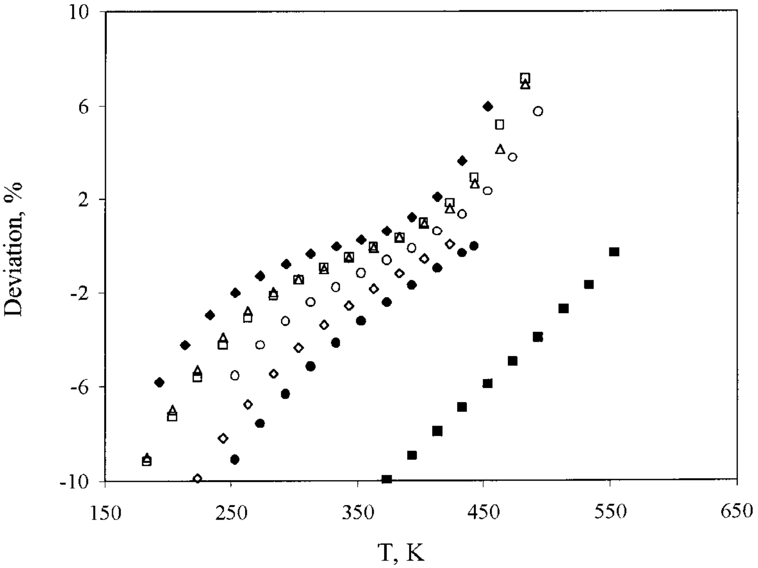

Fig. 6. Same as Fig. 4 for (◆) *n*-C_6H_{14}, (□) *n*-C_7H_{16}, (○) *n*-C_8H_{18}, (△) *i*-C_8H_{18}, (◇) *n*-C_9H_{20}, (●) *n*-$C_{10}H_{22}$, and (■) *n*-$C_{16}H_{34}$.

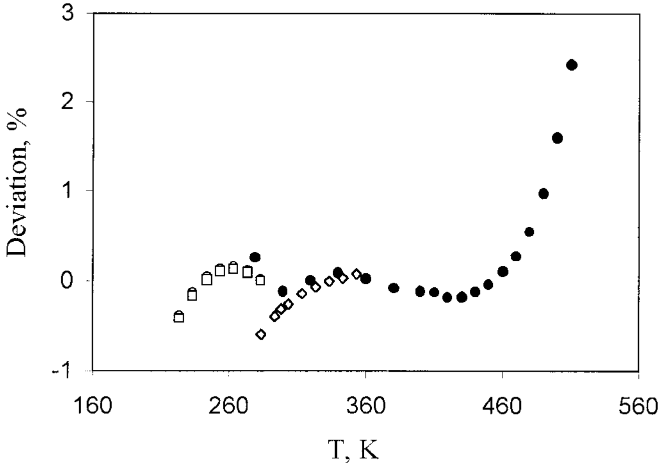

Fig. 7. Same as Fig. 4 for (●) C_6H_6, (◇) cyclo-C_6H_{12}, (○) cis-C_4H_8, (□) trans-C_4H_8.

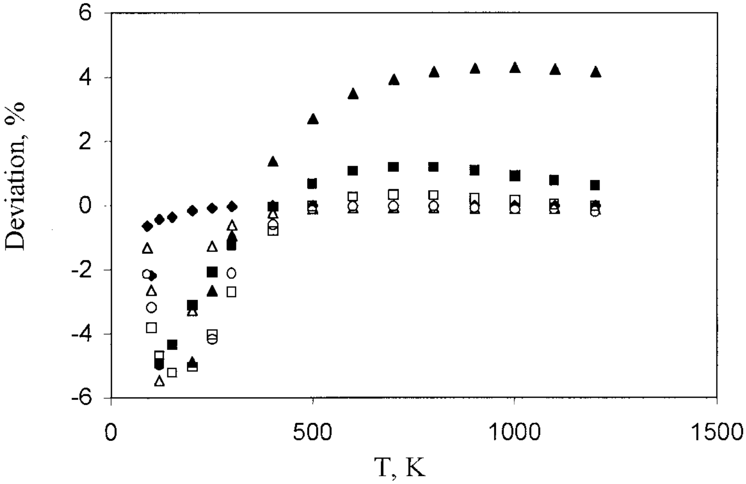

Fig. 8. Deviation plot for the density of compressed Ar as a function of temperature at (◆) 10 bar, (△) 100 bar, (○) 250 bar, (■) 500 bar, (□) 1000 bar, and (▲) 5000 bar.

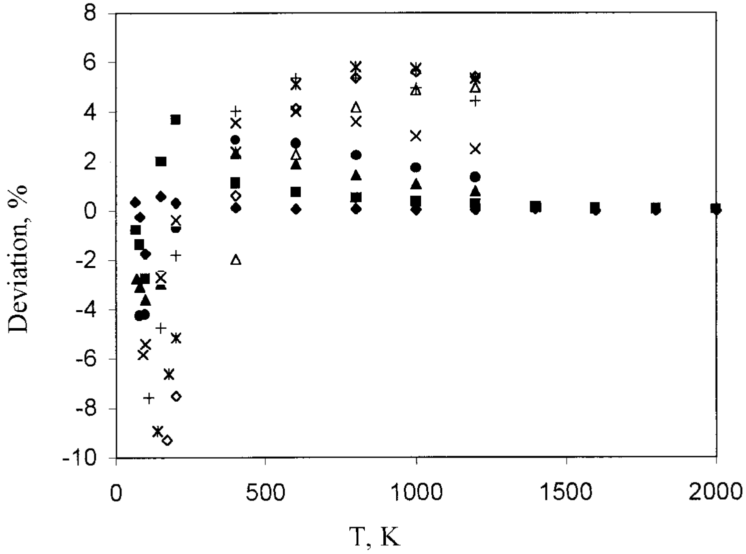

Fig. 9. Same as Fig. 8 for N_2 at (◆) 10 bar, (■) 100 bar, (▲) 300 bar, (●) 500 bar, (×) 1000 bar, (+) 2500 bar, (⁎) 5000 bar, (◇) 7000 bar, and (△) 10000 bar.

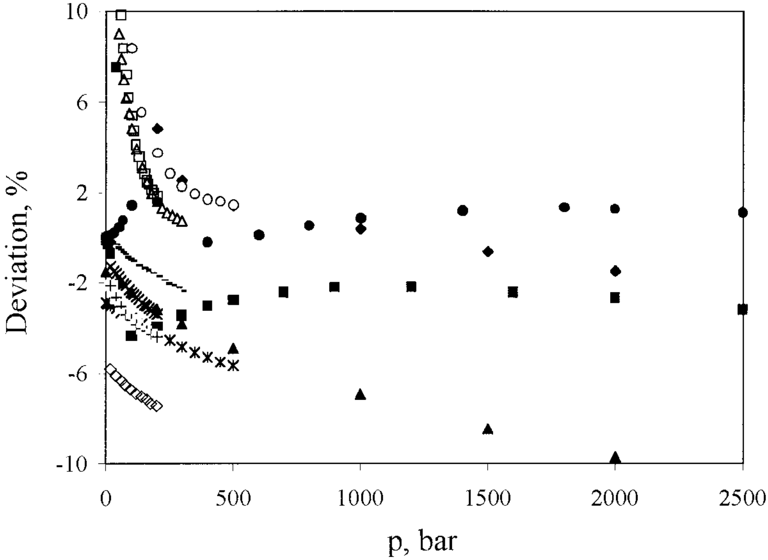

Fig. 10. Deviation plot for the density of n-C_6H_{14} at (—) 343.15 K and (△) 483.15 K, n-C_7H_{16} at (▲) 303.15 K and (◆) 573.15 K, n-C_8H_{18} at (⁎) 300 K and (○) 570 K, i-C_8H_{18} at (×) 373.15 K and (□) 553.15 K, n-$C_{10}H_{22}$ at (◇) 313.15 K and (+) 393.15 K, and C_2H_4 at (■) 273.15 K and (●) 423.15 K.

compared with the tabulated values [22–25], as the average absolute deviation and the maximum deviation, in a wide range of temperatures and pressures in Table II. The tabulated vapor-pressure data [22–25] have been used to calculate the liquid densities in the saturation state, and the Maxwell equal-area construction has not been applied. Although Table II is itself a measure of the accuracy of the present method, we have shown deviation plots for the density of compounds listed in Table II and also for C_2H_4 and a number of long-chain paraffins including n-C_9H_{20}, n-$C_{10}H_{22}$, and n-$C_{16}H_{34}$, which are more complex, in Figs. 4–10. It is clear that for almost all cases the deviations are not higher than $\pm 5\%$. The predicted densities at low temperatures get worse as the chain length gets longer.

4. CONCLUSIONS

Statistical-mechanical perturbation theory can be applied to determine the equation of state of real fluids if the intermolecular forces are known. For real fluids, however, the equation of state can be used with much less input information than the full potential, because the temperature-dependent parameters of the equation of state that depend only on the repulsion,

α and b, are insensitive to the detailed shape of the potential and can be scaled with two fixed constants, T_B and V_B, or in our procedure by T_{bp} and ρ_{bp}.

This work suggests that the *pvT* properties of a nonpolar fluid can be determined with reasonable accuracy from just the two scaling constants, T_{bp} and ρ_{bp}, which are readily available. The present work also shows that the volumetric behavior of molecular fluids can be obtained from relatively simple models, provided that suitable scaling factors are used. Without knowing any details of the intermolecular forces, we can predict the temperature-dependent parameters of the equation of state. Thus simple models suffice to determine thermodynamic properties.

Finally, our correlation scheme does not need the critical constants [8, 9], which are not available for many substances, or other parameters or properties such as acentric factor [9], the heat of vaporization [12], and the surface tension [13]. The previous correlations based on the critical constants, Fig. 2, and the heat of vaporization [12] both show higher scatter than the present one. The present correlation is less accurate than the correlation by Pitzer and Curl [9], but it does not require the critical parameters. Comparison of our predicted results for the density of substances examined with results using the previous correlations [12, 13, 9], shows that the accuracy of the present equation of state is nearly the same as the previous ones [12, 13, 21]. Meanwhile, the present equation of state is simpler than the previous ones [12, 13, 21] in that it requires only the boiling point parameters and the parameter λ is self-adjusted. Presumably, the scaling constants, T_{bp} and ρ_{bp}, vary in such a manner to compensate for the shape effects hidden in λ.

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