Greek Letters

φ = transition matrix of dimension $n \times n$
Δ = forcing function weighting matrix of dimension $n \times r$
λ = discretized vector function

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A Corresponding States Correlation of Saturated Liquid Volumes

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A corresponding states correlation is presented for the prediction of saturated liquid volumes. Parameters required are the critical temperature, the acentric factor, and a scaling volume. The correlation is valid over the entire useful range of reduced temperatures from 0.2 to 1.0. The full temperature range has not been covered by previous corresponding states correlations. Average absolute deviations in predicted liquid volumes is one-quarter of 1% for 26 compounds. The correlation is also useful for calculating critical temperatures, pressures, and volumes when experimental critical data are lacking. The proposed method also provides a convenient means for calculating rapidly and accurately the statistical mechanical parameters used in the cell model correlation developed by Renon, Eckert, and Prausnitz.

Liquid densities or liquid molar volumes are often needed for a wide variety of engineering calculations, such as the design of storage facilities, flow metering, high pressure vapor-liquid equilibrium studies, and fluid flow analyses. Not infrequently densities and other properties are required at temperatures for which no experimental data exist. In such cases the three-parameter corresponding states formulation of Pitzer et al. (*4*, *15*) is widely used for predicting the thermodynamic properties of dense gases and liquids. However, Pitzer's correlation for saturated liquid compressibility factors, and similar correlations by Lyckman, Eckert, and Prausnitz (*12*) and by Halm and Stiel (*6*), are valid only over a range of reduced temperatures of 0.56 to 1.00.

THE CORRELATION

A new correlation is presented here which is valid over essentially the entire useful range of 0.20 to 1.00 in the reduced temperature. This correlation has the following form linear in the acentric factor

$$V/V_{SC} = V_R^{(0)}(1.0 - \omega\ \delta) \qquad (1)$$

The generalized functions $V_R^{(0)}$ and δ, which are dependent only upon the reduced temperature, are tabulated in Table 1. These values were calculated from density data for the following 10 substances: argon, methane, nitrogen, propane, *n*-pentane, *n*-heptane, *n*-octane, benzene, ethyl ether and ethylbenzene.

To establish the correlation at the lowest temperatures, the functions $V_R^{(0)}$ and δ are extrapolated from a T_R of 0.225 to a T_R of 0.20. Density data for slightly subcooled propane liquid determine $V_R^{(0)}$ at a reduced temperature of 0.225, and the product $\delta\ V_R^{(0)}$ is virtually constant below a reduced temperature of 0.5.

The scaling volume V_{SC} is used in Equation (1) rather than the critical volume. It is widely recognized that the experimental determination of critical volumes is very difficult and subject to rather large errors. For this reason

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TABLE 1. REDUCED VOLUME FUNCTIONS FOR NONPOLAR FLUIDS

T_R	$V_R^{(0)}$	δ	T_R	$V_R^{(0)}$	δ
0.20	0.3113	0.2760	0.90	0.5289	0.1754
0.30	0.3252	0.2646	0.92	0.5501	0.1719
0.40	0.3421	0.2521	0.94	0.5771	0.1683
0.50	0.3625	0.2387	0.95	0.5941	0.1664
0.60	0.3862	0.2244	0.96	0.6147	0.1646
0.70	0.4157	0.2090	0.97	0.6410	0.1628
0.75	0.4341	0.2010	0.98	0.6771	0.1609
0.80	0.4562	0.1927	0.99	0.7348	0.1591
0.85	0.4883	0.1842	1.00	1.000	0

* Functions defined by Equation (1).

the parameter V_{SC} is based on liquid densities which can generally be measured with high accuracy far from the critical point. Table 2 lists scaling volumes for a number of compounds. This table also shows that for many compounds the scaling volume and the critical volume differ very little.

The scaling volume is defined by the following relationship:

$$V_{SC} = \frac{V_{0.6}}{0.3862 - 0.0866\omega} \tag{2}$$

The quantity $V_{0.6}$ is the molar volume of the liquid evaluated at a reduced temperature of 0.6. For compounds not listed in Table 2 the definition may be used to evaluate V_{SC}, provided that the critical temperature, the acentric factor, and a liquid density are known. If liquid densities are known only at temperatures far from 0.6, alternative calculations are possible with the aid of Equation (1) and Table 1. Density measurements made at the normal boiling point or at 20° to 25°C. are frequently the most accurate.

Table 3 records deviations between calculated and experimental saturated liquid volumes for 32 different substances. Average deviations are 0.5% or less and maximum deviations are less than 2.2% for all compounds except the highly polar ones. Below a reduced temperature of 0.8, maximum deviations are almost always less than 0.5%. The correlation is not intended to give satisfactory results for polar substances without the introduction of an additional correlating parameter (*6*). Nevertheless, moderately good results are obtained even for acetic acid, acetonitrile, and the alcohols. Similar findings have also been recorded by other investigators (*6*, *12*).

An electronic computer was used for calculating the data recorded in Table 3. For computerized computations the following equations may be used:

for $0.20 < T_R < 0.80$

$$V_R^{(0)} = 0.33593 - 0.33953\,T_R + 1.51941\,T_R^2 - 2.02512\,T_R^3 + 1.11422\,T_R^4 \tag{3}$$

for $0.80 < T_R < 1.0$

$$V_R^{(0)} = 1.0 + 1.3\,(1 - T_R)^{1/2}\log_{10}(1 - T_R) - 0.50879\,(1 - T_R) - 0.91534\,(1 - T_R)^2 \tag{4}$$

for $0.20 < T_R < 1$

$$\delta = 0.29607 - 0.09045\,T_R - 0.04842\,T_R^2 \tag{5}$$

Equation (4) gives the correct limiting conditions at the critical point. For T_R equal to 1

$$V_R = 1.0$$

$$\frac{dV_R}{dT_R} = \infty$$

The function δ, which is correlated from experimental data, has an apparent discontinuity at the critical point, that is, the function does not approach 0 as T_R approaches 1. Similar findings are recorded by other investigators (*12*). Neither the correlation nor most experimental data are very reliable in the reduced temperature range of 0.99 to 1.00, and the discontinuity in δ has no apparent adverse effect upon the predictive accuracy of the proposed method.

TABLE 2. SCALING VOLUMES AND CRITICAL VOLUMES FOR A NUMBER OF COMPOUNDS

Compound	V_{SC}, cu. cm./g.-mole	V_C, cu.cm./g.-mole (Ref. 9)	Compound	V_{SC}, cu.cm./g.-mole	V_C, cu. cm./g.-mole (Ref. 9)
Argon	75.25	75.20(7)	Acetylene	116.38	113.0
Nitrogen	89.64	90.1(7)	Etylene	130.41	129.0
Methane	99.10	99.0	Propylene	183.02	181.0
Ethane	145.42	148.0	Butylene	235.95	240.0
Propane	199.79	203.0	Benzene	255.53	259.0
n-Butane	254.07	255.0	Toluene	313.67	316.0
iso-Butane	256.72	263.0	Ethylbenzene	373.55	374.0
n-Pentane	310.97	304.0	*o*-Xylene	365.67	369.0
iso-Pentane	308.17	306.0	Bromobenzene	320.09	324.0
neo-Pentane	311.09	303.0	Chlorobenzene	305.18	308.0
n-Hexane	368.48	370.0	Carbon tetrachloride	274.84	276.0
2,3-Dimethylbutane	360.35	358.0	Methyl formate	168.92	172.0
n-Heptane	429.28	432.0	Ethyl acetate	284.61	286.0
2,2,3-Trimethylbutane	411.87	398.0	Ethyl ether	281.15	280.0
n-Octane	490.30	492.0	Carbon monoxide	91.79	93.1
Cyclohexane	307.98	308.0	Carbon dioxide	93.55	94.0
Ave. % Dev.* = 1.10%					

(7) Indicates critical volume data taken from reference 7.

* Ave. % Dev. $= \dfrac{100}{N}\displaystyle\sum_{1}^{N}\left|\frac{V_{SC} - V_C}{V_c}\right|$

TABLE 3. DEVIATIONS BETWEEN CALCULATED AND EXPERIMENTAL SATURATED LIQUID VOLUMES

Compound	Temperature range, °C.	Reduced temperature range	No. of data points	Volume % deviation		Data source
				Ave.	Max.	
Argon	−199 to −119	0.56 to 1.0	27	0.39	1.58	*5, 14*
Nitrogen	−210 to −152	0.50 to 0.95	14	0.36	0.92	*5*
Carbon monoxide	−205 to −140	0.51 to 1.0	26	0.48	2.16	*5*
Carbon dioxide	−10 to 40	0.86 to 1.0	9	0.54	1.41	*5*
Methane	−182 to −88	0.47 to 0.97	20	0.24	0.85	*5*
Propane	−190 to 96	0.22 to 0.99	40	0.19	0.53	*5, 19*
iso-Butane	30 to 125	0.74 to 0.98	5	0.43	0.59	*23*
n-Pentane	−136 to 195	0.291 to 1.0	55	0.16	0.55	*23, 28, 19*
n-Hexane	10 to 210	0.56 to 0.95	21	0.11	0.40	*28*
2,3-Dimethylbutane	0 to 210	0.55 to 0.97	22	0.27	0.82	*28*
n-Heptane	−90 to 264	0.34 to 0.99	65	0.17	0.88	*19, 28*
2,2,3-Trimethylbutane	10 to 30	0.53 to 0.57	3	0.14	0.17	*19*
n-Octane	0 to 260	0.48 to 0.94	27	0.15	0.63	*28*
Cyclohexane	15 to 104	0.52 to 0.68	8	0.25	0.64	*15, 23*
Ethylene	−145 to 6	0.45 to 0.99	14	0.40	0.81	*23*
Benzene	0 to 284	0.49 to 0.99	30	0.26	0.83	*28*
Toluene	57 to 127	0.56 to 0.67	4	0.07	0.16	*23*
o-Xylene	70 to 110	0.54 to 0.61	5	0.05	0.10	*19*
Ethylbenzene	−96 to 126	0.29 to 0.65	26	0.11	0.19	*19, 23*
Chlorobenzene	−45 to 329	0.36 to 0.86	34	0.12	0.50	*23, 28*
Bromobenzene	100 to 270	0.56 to 0.81	18	0.12	0.48	*48*
Methanol*	0 to 230	0.53 to 0.98	24	2.84	5.38	*28*
Ethanol*	0 to 240	0.53 to 0.99	25	3.03	5.63	*28*
n-Propanol*	80 to 260	0.66 to 0.99	19	3.20	4.23	*28*
Ethyl ether	−123 to 193	0.32 to 0.99	37	0.11	0.52	*23, 28*
Acetic acid*	0 to 310	0.46 to 0.98	34	0.70	1.72	*23, 28*
Methyl formate*	0 to 210	0.56 to 0.99	22	0.54	1.76	*28*
Ethyl acetate	−83 to 240	0.36 to 0.98	36	0.19	1.45	*23, 28*
Methyl butyrate	40 to 280	0.56 to 0.99	25	0.27	1.33	*28*
Acetone	15 to 50	0.56 to 0.64	5	0.06	0.12	*23*
Acetonitrile*	212 to 264	0.89 to 0.98	6	4.90	7.8	*23*
Ethyl mercaptan	0 to 218	0.55 to 0.99	16	0.42	0.81	*23*
Grand ave. dev.			592	0.22		

*** Polar components not included in overall average deviation.**

ESTIMATION OF THE CRITICAL CONSTANTS

Liquid densities are available for a much wider selection of compounds than are critical volumes, especially for the more complex molecules for which thermal decomposition often occurs before the critical point is reached. Table 2 shows that V_{SC}, which is readily calculated from liquid densities at low reduced temperatures, is a good approximation for the critical volume. A comparison between V_{SC} and V_C for nonpolar compounds shows an average deviation of 1.1% and a maximum deviation of 3.5% in the case of 2,2,3-trimethyl butane. This is well within the range of experimental error for the critical volume.

The small differences between the two volumes, V_{SC} and V_C, may be ascribed to two causes. First, it is well known that experimental difficulties in determining V_C are very great and that the measurements are subject to rather large errors. Second, the critical region is unstable and consequently very sensitive to minor intermolecular or structural effects. For this reason, V_C may not be a good scaling factor for predicting thermodynamic properties by means of the correspondence principle.

The scaling volume may also be used as the basis for a critical compressibility factor correlation. For this purpose a compressibility factor is defined as

$$Z_{SC} = \frac{P_C V_{SC}}{R T_C} \qquad (6)$$

Next Z_{SC} is correlated as a function of the acentric factor. A linear regression analysis of Z_{SC} for 26 nonpolar substances produces the following result:

$$Z_{SC} = 0.2920 - 0.0967\ \omega \qquad (7)$$

For the same 26 compounds the following correlation is obtained for the experimental critical compressibility factor:

$$Z_C = 0.2918 - 0.0928\ \omega \qquad (8)$$

The standard deviation for Equation (7) is 0.87% for the 26 fluids. This is about half of the standard deviation of 1.57% for Equation (8).

From the preceding information it is clear that the saturated liquid volume correlation, Equation (1), may be used to predict critical temperatures and pressures as well as critical volumes. The only information required is a vapor pressure or a normal boiling point measurement and two liquid density measurements preferably at widely separated temperatures. The vapor pressure is used to calculate the acentric factor. From the density data two simultaneous equations equivalent to Equation (1) are solved to obtain V_{SC} and T_C. The critical pressure is calculated next with the aid of Equation (7) and the following relationship:

$$P_C = \frac{Z_{SC} R T_C}{V_{SC}} \qquad (9)$$

TABLE 4. COMPARISON BETWEEN CALCULATED AND EXPERIMENTAL CRITICAL CONSTANTS

Property	Type of data	Toluene	Ethane	Ethylene	Ethyl acetate
Critical temperature, °K.	Calc.	592.41	304.30	282.27	521.52
	Exp.	591.7 to 593.7	305.2 to 305.7	282.3 to 283.0	523.25
Critical pressure, atm.	Calc.	41.44	48.45	49.94	38.55
	Exp.	40.5 to 41.6	48.2 to 49.8	49.90	37.8 to 38.0
Critical volume, cu.cm./g.-mole	Calc.	313.29	145.25	131.44	284.09
	Exp.	316.3	141.8 to 148.1	123.6 to 133.7	285.9 to 286.3
Temp. range for liquid densities, °K.		174 to 319	93 to 183	128 to 225	190 to 413
Data source		*1, 2, 8, 22*	*3, 11, 20, 21*	*13, 24, 25*	*2, 26*

Table 4 compares the experimental critical constants for four compounds with those predicted by the method just described. The agreement is very good and in most cases the predicted critical constants fall within the range of experimental values obtained in recent investigations. The method, however, should not be used to predict critical parameters of polar materials.

COMPARISON WITH OTHER METHODS

The Lydersen method (*10*) is recommended by Reid and Sherwood (*17*) as one of the best for estimating the critical constants of compounds. This method shows an average deviation of 1.2% between experimental and calculated critical volumes for the compounds listed in Table 2 of this work. The scaling volume shows a slightly lower average deviation of 1.1%. The real advantage of the proposed method results because the scaling volume is derived from the principle of corresponding states. For corresponding states correlations, therefore, at least in the liquid state, V_{SC} is expected to be a more accurate correlating parameter than the critical volume predicted by the Lydersen method.

The accuracy of critical temperature and pressure predictions by the two methods is reported below:

	Average % deviation	
	T_C	P_C
Lydersen method (*10*)	0.4	3.0
Proposed method	0.2	1.0

Although both methods are quite satisfactory, the corresponding states technique is more accurate. However, for predicting critical temperatures and pressures in general, the authors recommend the use of Lydersen method because of its greater simplicity. The proposed method for calculating critical volumes is quite simple, but the simultaneous solution for both T_C and V_{SC} is tedious without the aid of a computer. This method, therefore, is justified primarily if a high degree of accuracy is required or if the critical properties are desired for an unusual compound for which the Lydersen group contributions are poorly established.

Two of the most accurate generalized methods for predicting liquid volumes are those of Lyckman et al. (*12*) and of Yen and Wood (*26*). The predictive accuracy of these two methods and of the one proposed are tested with experimental liquid density data for 20 nonpolar fluids selected from those listed in Table 3. The results of this test are summarized next:

Method	Ave. deviation %	Ave. of maximum deviations %
Yen and Woods (*26*)	1.6	2.2
Lyckman et al. (*12*)	1.4	2.1
Proposed (20 comp.)	0.22	0.66
Proposed (16 comp.)	0.27	0.75

In the final row of the preceding table average deviations are also reported for a list of 16 compounds, none of which is used to establish the proposed correlation.

RELATIONSHIP BETWEEN MACROSCOPIC AND STATISTICAL MECHANICAL THEORIES

The correlation proposed in this work utilizes a macroscopic corresponding states approach to predicting thermodynamic properties with the aid of critical constants. Equations (1) through (5), however, establish a link between this approach and the statistical mechanical method developed by Renon, Eckert, and Prausnitz (*18*).

Using a cell model, these investigators derived a liquid equation of state requiring only three parameters V^*, T^*, and U^*, which are, respectively, the characteristic volume, temperature, and internal energy. Renon et al. derived the following equations for predicting the three characteristic parameters:

$$V^* = \frac{V'}{1.3138} \tag{10}$$

$$T^* = \frac{T'}{0.66188} \tag{11}$$

$$U^* = \frac{U'}{-0.88203} \tag{12}$$

The primed quantities V', T', and U' are all evaluated at the condition $(\partial \ln V_R/\partial \ln T_R) = 0.4$. This same condition applied to Equation (1) in this work leads to the following relationship:

$$\frac{\partial \ln V_R}{\partial \ln T_R} = \frac{T_R}{V_R}\,\frac{\partial V_R^{(0)}}{\partial T_R} + \frac{\omega \partial (V_R^{(0)}\,\delta)}{\partial T_R} = 0.4 \tag{13}$$

The simultaneous solution of Equations (1) and (13) yields the following results for T' and V':

$$T' = T_C(0.60 - 0.072\omega) \tag{14}$$

$$V' = V_{SC}(0.386 - 0.106\omega) \tag{15}$$

By combining Equations (14) and (15) with (10) and (11), one obtains relationships which relate the characteristic parameters T^* and V^* to the critical constants and the acentric factor:

$$T^* = T_C\,(0.906 - 0.109\omega) \tag{16}$$

$$V^* = V_{SC}\,(0.293 - 0.018\omega) \tag{17}$$

Values of T^* calculated for 11 liquids from Equation (*16*) averaged 2% greater than those reported by Renon et al. for these fluids. Values of V^* calculated from Equation (17) averaged less than 1% higher than those originally reported. However, there appears to be little loss in predictive accuracy for liquid volumes when T^* and V^* are evaluated from Equations (16) and (17). These equations, therefore, provide a simple, rapid method for evaluating the characteristic parameters. In addition these relationships demonstrate the considerable degree of equivalence between the macroscopic and the statistical mechanical methods.

There is no direct relationship between the proposed method for predicting liquid volumes and the configurational internal energy function; and therefore we cannot derive an equation for predicting U^* comparable to Equations (16) and (17). These latter relationships, however, suggest that a similarly simple form should exist for the characteristic energy; and a direct curve fit of the parameters reported by Renon et al. (*18*) yields the following result:

$$U^* = RT_C\,(5.243 + 8.692\omega) \tag{18}$$

Equation (18) deviates by an average of 1.1% from the reported values for U^*.

SUMMARY

The correlation, described by Table 1 and Equations (1) through (5), is shown to predict saturated liquid volumes accurately for a wide variety of nonpolar compounds. Three parameters are required: the critical temperature, the acentric factor, and the scaling volume. The method preserves the simple linear form with respect to the acentric factor as first proposed by Pitzer et al. (*4, 15*). The correlation is extended down to a reduced temperature of 0.20, a greater range than that covered by previous corresponding states methods. In addition, the accuracy appears to be better than that of other methods of comparable simplicity and generality. The correlation may be used for predicting critical temperatures, volumes, and pressures when experimental ones are not available. Finally, Equations (16) and (17), which are derived in this paper, are shown to provide a simple method for evaluating the characteristic parameters used by Renon et al. (*18*) for their correlation of liquid properties based on a cell model partition function.

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NOTATION

P_C, T_C, V_C, Z_C = critical constants (pressure, temperature, volume, and compressibility factor, respectively)
T = temperature
T_R = T/T_C, reduced temperature
U = configurational internal energy
V = saturated liquid volume
V_{SC} = scaling volume as defined by Equation (2)
V_R = V/V_{SC}, a reduced volume
$V_R^{(0)}$ = generalized reduced volume for a simple fluid, for example, argon as defined by Equation (1)
Z = compressibility factor
Z_{SC} = characteristic compressibility factor as defined by Equation (7)
δ = generalized deviation function as defined by Equation (1)
ω = acentric factor

Superscripts

$*$ = characteristic parameter for the statistical mechanical correlation of Renon et al. (*18*)
$'$ = property of liquid evaluated at the condition $(\partial \ln V/\partial \ln T_R) = 0.4$

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