

R = constant
 T = surface tension, dynes/cm.
 \hat{T} = separation of variables parameter, only a function of t
 t = time, sec.
 u_i = i component of velocity, cm./sec.
 u_j = j component of velocity, cm./sec.
 v = y component of velocity, cm./sec.
 x_i = i coordinate distance
 x_j = j coordinate distance
 x_2 = y coordinate distance
 X_i = i component of body force
 y = y coordinate distance
 Y = separation of variables parameter, only a function of y

Greek Letters

α = ultrasonic wave attenuation coefficient, neper/cm.
 γ = characteristic Mathieu function index
 γ_c = critical value of Mathieu function index
 ϵ = fluid particle displacement, cm.
 ϵ^* = maximal value of fluid particle displacement at the crystal surface, cm.
 ϵ_c = particle displacement caused by the crystal at the crystal surface, cm.
 ϵ_i = particle displacement at gas-liquid interface, cm.
 η = capillary wave amplitude
 η^* = time dependent part of capillary wave amplitude
 θ = dimensionless time
 λ = wavelength of surface wave cm.⁻¹
 λ_u = wavelength of ultrasonic wave, cm.⁻¹

μ = surface viscosity
 $\frac{\mu}{\rho}$ = constant defined by Equation (6)
 ρ = density of fluid, g./cc.
 ρ_0 = constant equilibrium density, g./cc.
 σ = phase angle in solution to Mathieu's equation = $\pi/4$
 ϕ = velocity potential
 ϕ^* = time dependent part of velocity potential

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A Generalized Equation for Computer Calculation of Liquid Densities

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A generalized equation explicitly relating reduced density to reduced temperature and reduced pressure has been developed for calculating liquid densities of pure compounds on a digital computer. The analytical formulation is based mainly on a modified corresponding states principle and the graphical correlation of Lydersen, Greenkorn, and Hougen. The calculated densities from the equation reproduce the literature data within 2% for sixty-two saturated liquids and nineteen compressed liquids.

With the aid of a pseudocritical method and a generalized equation of vapor pressure, the same equation is readily applied to the estimation of liquid mixture densities. The method of Prausnitz and Gunn is chosen for the evaluation of pseudocritical constants. For fifteen binary systems, one ternary system, and one quinary system, the one hundred fifty-nine calculated densities agree with the literature data to within 3%. This method is limited to pseudoreduced temperatures less than 1.0.

The principle of corresponding states has been used extensively in correlating and predicting volumetric properties and thermodynamic functions. Based on a modified form of the principle which contains a third parameter, z_c , Lydersen, Greenkorn, and Hougen (9) have developed a generalized correlation for liquid densities of pure compounds over a wide range of temperatures and pressures. This correlation, presented in both graphical and tabular form, has been thoroughly reviewed and found to

to be accurate to about 2 to 5% (13). When experimental data for liquid densities are not available, the generalized correlation has been widely used for the prediction of liquid densities in engineering work. However, this use has been limited to manual calculations. While digital computers are increasingly used in process calculations throughout the industry, the adoption of the Lydersen-Greenkorn-Hougen correlation for computer work would require a storage for a large number of points and exten-

sive interpolation methods, unless some form of analytical expressions is available. Hirschfelder, Buehler, McGee, and Sutton (5) have developed such analytical expressions, but the density is implicitly formulated. In order to calculate a liquid density at a given temperature and pressure, some kind of iterative method has to be developed. This iterative method so far has not been reported. Furthermore, in most engineering work it is the liquid mixture densities which are needed, but they have been studied only to a limited extent (8, 15). The purpose of this paper is to present an analytical formulation, explicit in density, based mainly on the correlation of Lydersen, Greenkorn, and Hougen so that liquid densities can be directly calculated on a digital computer, and to demonstrate its possible application to the estimation of liquid mixture densities.

SATURATED LIQUID DENSITIES

Experimental density data for pure liquids at saturation states are abundant and have been subjected to various treatments. Francis (2, 3) has fitted the following equation to the experimental liquid density data with good accuracy.

$$\rho = A - Bt - \frac{C}{E - t} \quad (1)$$

Although Equation (1) has been used extensively in previous works, it is not applicable at temperatures near the critical point. Martin (10) has suggested the following equation:

$$\rho_{r,s} = \frac{\rho_s}{\rho_c} = 1 + A(1 - T_r)^{1/3} + B(1 - T_r)^{2/3} + C(1 - T_r) + D(1 - T_r)^{4/3} \quad (2)$$

which holds up to the critical temperature. It has been found in this work that almost an equal accuracy can be obtained without the use of the fourth term on the right-hand side of Equation (2). Literature data for sixty-two pure compounds have been fitted with the following equation:

$$\rho_{r,s} = 1 + A(1 - T_r)^{1/3} + B(1 - T_r)^{2/3} + D(1 - T_r)^{4/3} \quad (3)$$

with an average deviation of less than 0.3%. The specific coefficients A, B, and D in Equation (3) and the critical constants for these pure compounds are given in Table 1.*

Based on the literature density data mentioned above, a generalized graphical correlation relating $\rho_{r,s}$ to T_r and z_c has been developed. This graphical correlation agrees well with those developed earlier by Lydersen et al. (9) and Hobson and Weber (6), and can be analytically represented by Equation (3) with the following generalized coefficients A, B, and D.

$$A = 17.4425 - 214.578 z_c + 989.625 z_c^2 - 1522.06 z_c^3 \quad (4)$$

$$B = -3.28257 + 13.6377 z_c + 107.4844 z_c^2 - 384.211 z_c^3 \quad (5A)$$

if $z_c \leq 0.26$

$$B = 60.2091 - 402.063 z_c + 501.0 z_c^2 + 641.0 z_c^3 \quad (5B)$$

if $z_c > 0.26$

$$D = 0.93 - B \quad (6)$$

Equations (3) through (6) have been used to calculate saturated liquid densities of sixty-two pure compounds

whose z_c values range from 0.21 to 0.29. For a total of six hundred and ninety-three points, the calculated values agree with the literature data within 2.1%. While for most of the pure liquids the generalized coefficients reproduce the experimental data to within 2%, a maximum deviation of 10% has been observed for methylcyclohexane. This is probably due to the uncertainty of the critical density which generally is not so reliable as the critical temperature.

COMPRESSED LIQUID DENSITIES

At pressures higher than the vapor pressure, the liquid densities increase with increasing pressures. The density of a compressed liquid is defined in reduced quantities as

$$\rho_r = \frac{\rho}{\rho_c} = \rho_{r,s} + (\Delta\rho_r)_{27} + \delta z_c \quad (7)$$

In Equation (7) the isothermal pressure effect on density for a pure liquid with reference to the saturated liquid density is the sum of $(\Delta\rho_r)_{27}$ and δz_c . The former term $(\Delta\rho_r)_{27}$ represents the reduced density increase caused by ΔP_r , or $(P_r - P_{r,s})$, the reduced pressure increase from the vapor pressure to the given pressure, for compounds with a z_c value of 0.27. The latter term δz_c , which equals zero for $z_c = 0.27$, further corrects the isothermal pressure effect on density for compounds with other z_c values. In this work, three other z_c values, 0.29, 0.25, and 0.23, have been selected and are designated by subscripts 29, 25, and 23, respectively. For a pure compound, $\rho_{r,s}$ can be calculated by using the equations given in the previous section. The problem of calculating compressed liquid densities then becomes one of calculating $(\Delta\rho_r)_{27}$ and δz_c .

For compounds with z_c values of 0.27, Lydersen et al. (9) have correlated the reduced liquid densities, including those on the saturated liquid line as a function of reduced temperatures and reduced pressures. Based on this correlation, values of $(\Delta\rho_r)_{27}$ have been calculated in this work as a function of ΔP_r and T_r , and then fitted to the following equation:

$$(\Delta\rho_r)_{27} = E_{27} + F_{27} \ln \Delta P_r + G_{27} e^{H_{27} \Delta P_r} \quad (8)$$

where

$$E_{27} = 0.714 - 1.626 (1 - T_r)^{1/3} - 0.646 (1 - T_r)^{2/3} + 3.699 (1 - T_r) - 2.198 (1 - T_r)^{4/3} \quad (9)$$

$$F_{27} = \frac{0.268 T_r^{2.0097}}{1.0 + 0.8 (-\ln T_r)^{0.441}} \quad (10)$$

$$G_{27} = 0.05 + 4.221 (1.01 - T_r)^{0.75} e^{-7.848(1.01 - T_r)} \quad (11)$$

$$H_{27} = -10.6 + 45.22 (1 - T_r)^{1/3} - 103.79 (1 - T_r)^{2/3} + 114.44 (1 - T_r) - 47.38 (1 - T_r)^{4/3} \quad (12)$$

and the subscript 27 designates z_c of 0.27. When used to calculate the isothermal pressure effect on density for pure compounds and to compare calculated values with experimental data, Equations (8) through (12) have been found to give satisfactory results for reduced temperatures from 0.3 to 1.0 and ΔP_r from 0.2 to 60, although the original correlation of Lydersen et al. was only good to a reduced pressure of 30. For ΔP_r values less than 0.2, the following equation is recommended in addition to Equations (8) through (12):

$$(\Delta\rho_r)_{27} = [(\Delta\rho_r)_{27} \text{ at } \Delta P_r = 0.2] \left(\frac{\Delta P_r}{0.2} \right) \quad (13)$$

For compounds of the other selected z_c values, 0.29, 0.25, and 0.23, it is necessary to calculate δz_c as mentioned previously. The subscript z_c is 29, 25, or 23 designating z_c values of 0.29, 0.25, or 0.23, respectively. Based on the correlation of Lydersen et al., the δz_c values have

* Tabular material has been deposited as document 8638 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for \$1.25 for photoprints or 35-mm. microfilm.

been calculated in this work as a function of ΔP_r and T_r , and then fitted to the following equation,

$$\delta z_c = I + J \ln \Delta P_r + K e^{L \Delta P_r} \quad (14)$$

where

$$I = a_1 + a_2 (1 - T_r)^{1/3} + a_3 (1 - T_r)^{2/3} + a_4 (1 - T_r) + a_5 (1 - T_r)^{4/3} \quad (15)$$

$$J = b_1 + b_2 (1 - T_r)^{1/3} + b_3 (1 - T_r)^{2/3} + b_4 (1 - T_r) + b_5 (1 - T_r)^{4/3} \quad (16)$$

$$K = c_1 + c_2 T_r + c_3 T_r^2 + c_4 T_r^3 \quad (17)$$

$$L = d_1 + d_2 (1 - T_r)^{1/3} + d_3 (1 - T_r)^{2/3} + d_4 (1 - T_r) + d_5 (1 - T_r)^{4/3} \quad (18)$$

and a_i through a_5 , b_i through b_5 , c_i through c_4 , and d_i through d_5 are given in Table 2. Equation (14) is valid for reduced temperatures from 0.3 to 1.0 and ΔP_r from 0.2 to 60. For ΔP_r less than 0.2, the following equation is recommended in addition to Equations (14) through (18).

$$\delta z_c = [\delta z_c \text{ at } \Delta P_r = 0.2] \left(\frac{\Delta P_r}{0.2} \right) \quad (19)$$

To use the above equations in calculating the compressed liquid density of a pure compound at a given temperature and pressure on a digital computer, the following computation procedure is followed.

1. If $z_c = 0.27$, set δz_c in Equation (7) equal to zero. For other z_c values, select one of the three sets of constants in Table 2 according to the z_c value of the given compound, and calculate δz_c from Equations (14) through (19). If the z_c value is not exactly 0.29, 0.25, or 0.23, use the closest one. The vapor pressure data needed in calculating ΔP_r are generally available in the literature or can be estimated by using Equation (24), to be given later.

2. Calculate $(\Delta \rho_r)_{\infty}$ from Equations (8) through (13).

3. Calculate ρ_{rs} from Equation (3). Use the specific coefficients A , B , and D if available. Otherwise, use the generalized coefficients obtained from Equations (4) through (6).

4. Calculate ρ_r and ρ from Equation (7).

In order to check the accuracy of Equations (7) through (19), compressed liquid densities for nineteen pure compounds over a wide temperature and pressure range have been calculated on an IBM 7094 computer. The nineteen compounds are: *n*-butane, *n*-octane, *n*-nonane, *n*-decane, ethylene, propylene, 1-butene, benzene, nitrogen, carbon dioxide, ammonia, water, nitrous oxide,

ethanol, sulfur dioxide, hydrogen sulfide, methyl chloride, and ethyl chloride. For a total of six hundred and eighty-seven points, the calculated densities give an average deviation of 1.9% when compared with experimental data. The maximum deviation, 5.8%, has been found for ethyl chloride. This result is considered to be satisfactory, since this is about the same accuracy as one can expect from the original graphical correlation of Lydersen, Greenkorn, and Hougen.

LIQUID MIXTURE DENSITIES

The correlations presented above can be applied to the estimation of liquid mixture densities which are often more important than pure compound densities in engineering calculations. Such applications have been made by Ritter et al. (15) in calculating the liquid density of a multicomponent hydrocarbon mixture and by Leland and Mueller (8) who calculated saturated liquid densities of various binary systems. In order to calculate liquid mixture densities with the equations developed in this work for pure compounds, it is only necessary to evaluate the pseudo values for the following properties of the given mixture: critical constants, saturated liquid density, and vapor pressure. Once these quantities are evaluated, the calculation of liquid mixture densities is the same as that of pure liquid densities.

Several methods for the evaluation of pseudocritical constants have been proposed. A summary of these methods is available elsewhere (7, 14, 18). For simplicity, two methods, one by Kay and another by Prausnitz and Gunn (12), have been tested. It has been found in this work that better results can be obtained by using the method of Prausnitz and Gunn, which states

$$T_c' = \sum_{i=1}^N x_i T_{c,i} \quad (20)$$

$$V_c' = \sum_{i=1}^N x_i V_{c,i} \quad (21)$$

$$z_c' = \sum_{i=1}^N x_i z_{c,i} \quad (22)$$

$$P_c' = z_c' RT_c' / V_c' \quad (23)$$

These pseudocritical constants of mixtures are used in place of the critical constants of pure compounds.

The pseudosaturated liquid density of a mixture is calculated in this work by using Equation (3) with generalized coefficients and pseudocritical constants.

The pseudovapor pressure of a mixture is calculated in this work based on a generalized correlation of vapor pressure by Lydersen, Greenkorn, and Hougen.

$$\log_{10} P_{rs}' = A \left(1 - \frac{1}{T_r'} \right) \quad (24)$$

where A is a function of z_c given in reference 9.

It is apparent that the above method of estimating liquid mixture densities should be limited to pseudoreduced temperature less than 1, since all the equations developed are for pure compounds in the liquid state which can only exist at reduced temperatures less than 1. Therefore, the calculation of liquid mixture densities in this work is limited to pseudoreduced temperatures less than 1, although liquid mixtures can exist at pseudoreduced temperatures higher than 1.0.

To test the method as outlined above, liquid densities for fifteen binary mixtures, one ternary mixture, and one quinary mixture at saturation states, as well as in the compressed liquid region, have been calculated and compared with experimental data. The result of this compari-

TABLE 2. CONSTANTS IN EQUATIONS (15) THROUGH (18)

	$z_c = 0.29$	$z_c = 0.25$	$z_c = 0.23$
a_1	-0.0817	0.0933	0.0890
a_2	0.3274	-0.3445	-0.4344
a_3	-0.5014	0.4042	0.7915
a_4	0.3870	-0.2083	-0.7654
a_5	-0.1342	0.05473	0.3367
b_1	-0.0230	0.0220	0.0674
b_2	-0.0124	-0.003363	-0.06109
b_3	0.1625	-0.07960	0.06261
b_4	-0.2135	0.08546	-0.2378
b_5	0.08643	-0.02170	0.1665
c_1	0.05626	0.01937	-0.01393
c_2	-0.3518	-0.03055	-0.003459
c_3	0.6194	0.06310	-0.1611
c_4	-0.3809	0	0
d_1	-21.0	-16.0	-6.550
d_2	55.174	30.699	7.8027
d_3	-33.637	19.645	15.344
d_4	-28.109	-81.305	-37.04
d_5	26.277	47.031	20.169

TABLE 3. DEVIATIONS OF CALCULATED LIQUID MIXTURE DENSITIES FROM LITERATURE DATA

Mixture	No. of compositions	Temperature range, °F.	Pressure range, lb./sq. in. abs.	No. of points	% Deviation		Reference
					Average	Maximum	
Methane-propane	4*	100 to 160	500 to 1,000	4	5.77	12.79	16
Methane- <i>n</i> -butane	1	220	600 to 10,000	11	2.50	4.33	
	9*	100 to 250	100 to 1,000	9	4.74	10.23	
Methane- <i>n</i> -pentane	3*	100 to 160	200 to 800	3	1.81	4.14	
Methane- <i>n</i> -decane	1	100	1,750 to 10,000	8	6.65	7.75	
	4*	100 to 460	1,000 to 2,000	4	7.10	7.62	
Methane-hydrogen sulfide	2	40 to 160	1,000 to 10,000	20	2.21	9.37	17
	2*	100 to 160	1,000 to 1,500	2	3.64	5.04	
Ethane-propylene	1	160	800 to 10,000	10	1.81	7.41	
Propane-benzene	1	100	200 to 10,000	12	1.89	3.40	
	2*	220 to 340	200 to 500	2	2.46	3.12	
<i>n</i> -Butane-carbon dioxide	1	100	800 to 10,000	10	2.00	2.87	
<i>n</i> -Pentane-hydrogen sulfide	1	100	400 to 10,000	11	1.02	3.12	
	3*	40 to 220	150 to 1,000	3	0.95	1.07	
<i>n</i> -Decane-hydrogen sulfide	1	220	600 to 10,000	11	4.21	5.69	
	3*	160 to 220	600 to 1,000	3	7.79	8.43	
Propylene-1-butene	1*	100	200	1	0.03	0.03	
Propane-carbon dioxide	1*	40	200	1	1.31	1.31	
Dichlorodifluoromethane (Freon 12)-Monochloro- difluoromethane (Freon 22)	11	39 to 218	157 to 1,257	11	1.17	3.46	19
Propanol-water	10	73 to 122	14.7	10	2.37	3.46	11
Nitrogen- <i>n</i> -heptane	2*	175 to 260	2,111 to 3,022	2	3.33	4.02	1
Methane- <i>n</i> -butane- <i>n</i> -decane	2	460	2,000 to 10,000	10	1.10	2.47	16
Methane-ethane-propane - <i>n</i> -pentane- <i>n</i> -hexane	1	73	1,640	1	1.71	1.71	4

Total number of points: 159

Overall average deviation: 2.8%

* Liquid mixtures are at saturation states.

son is given in Table 3. For a total of one hundred fifty-nine points, the average deviation is 2.8%, which is of the same order of magnitude as that found for pure compounds. Mixtures consisting of polar components are also included in this test. In view of the approximate nature of the corresponding states principle and the method of evaluating pseudo properties for mixtures, this result is considered to be rather satisfactory.

DISCUSSION

The correlation developed in this work is explicit in density, a feature which enables direct calculation of liquid densities from temperatures and pressures on a digital computer. The accuracy in using this correlation to calculate liquid densities of pure compounds and mixtures has been presented in the previous section. It should be interesting to compare further the calculated results with those by other correlations developed prior to this work. The one correlation to be compared below is the method developed by Hirschfelder, Buehler, McGee, and Sutton (5).

The equation of state developed by Hirschfelder et al. is applicable to both gaseous and liquid states. The comparison to be made between Equation (7) developed in this work and the equation of Hirschfelder et al. is for the liquid state. In order to make a direct comparison, it is necessary to be able to calculate pressure from a given temperature and density by using Equation (7), since the equation of Hirschfelder et al. is implicit in density. Equation (7), when applied to a given temperature and density, gives density as a function of pressure. The Newton-Raphson iterative method is readily applicable to its solution. The pressures thus calculated are illustrated for *n*-butane and *n*-decane in Figures 1 and 2, and are favor-

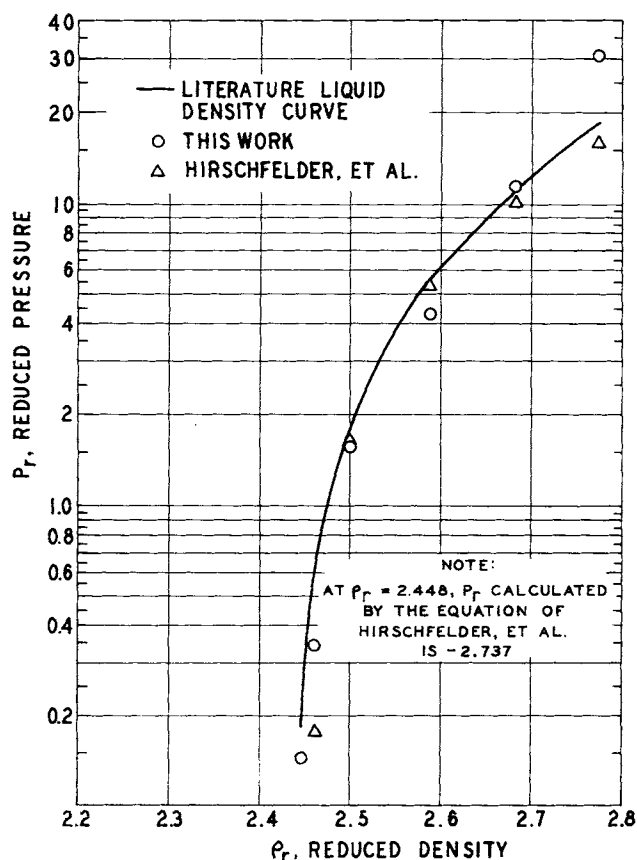


Fig. 1. Comparison of accuracy with the equation of Hirschfelder et al. (5) for *n*-butane at $T_r = 0.7313$.

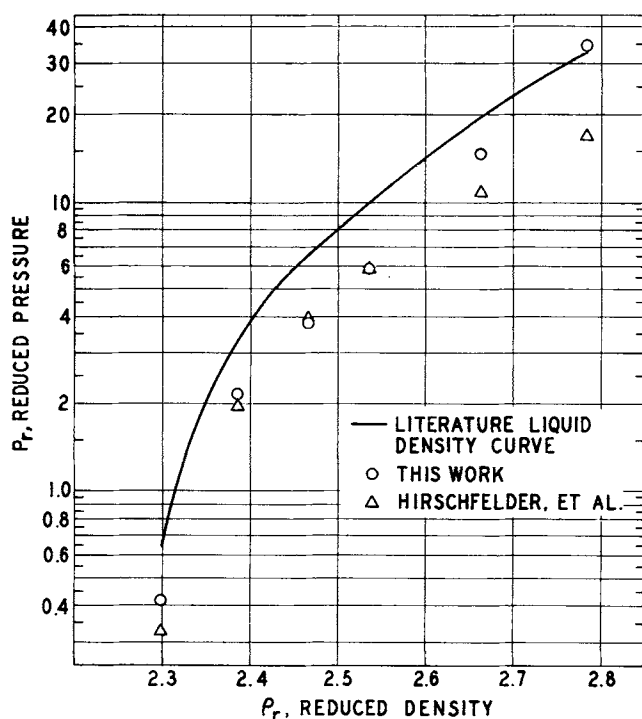


Fig. 2. Comparison of accuracy with the equation of Hirschfelder et al. (5) for *n*-decane at $T_r = 0.8254$.

ably compared with the corresponding pressures calculated by Hirschfelder et al. (5). It is noted that in the case of *n*-butane, a negative reduced pressure of -2.737 is calculated from the equation of Hirschfelder et al. (5). This point cannot be plotted in Figure 1.

When the equation developed in this work is applied to the calculation of liquid mixture densities, it has been pointed out previously that the pseudoreduced temperature should be between 0.3 and 1.0. Although limited, this represents a rather wide temperature range and is sufficient for most engineering work. It may be possible, however, in future studies to extend further the correlation into pseudoreduced temperatures higher than 1.0 by use of experimental density data for liquid mixtures.

CONCLUSION

A generalized equation of state for pure liquids, explicit in density, has been developed for direct calculations of liquid densities on digital computers, while the generalized equations previously available require iterative techniques for their solution. This generalized equation can also be used to calculate liquid densities of mixtures based on a pseudocritical method between pseudoreduced temperatures of 0.3 and 1.0. The calculated densities reproduce experimental data within 2% for pure liquids and 3% for liquid mixtures. The accuracy of the generalized equation has been found to compare favorably with the equation of Hirschfelder et al.

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NOTATION

$A, B, C \dots K, L$ = coefficients
 a_1, a_2, a_3, a_4, a_5 = coefficients in Equation (15)

b_1, b_2, b_3, b_4, b_5 = coefficients in Equation (16)
 c_1, c_2, c_3, c_4 = coefficients in Equation (17)
 d_1, d_2, d_3, d_4, d_5 = coefficients in Equation (18)
 N = number of components in a mixture
 P = pressure
 R = gas constant
 t = temperature
 T = absolute temperature
 V = molal volume
 x = mole fraction
 z = compressibility factor

Greek Letters

δ = liquid density change for compounds with z_c values other than 0.27, defined in Equation (7)
 ρ = liquid density
 $\Delta\rho$ = liquid density change due to pressure, defined in Equation (7)
 $\Delta P_r = P_r - P_{r,c}$

Superscripts

' = pseudo
 c = critical
 i = i^{th} component
 r = reduced
 s = saturated
 z_c = critical compressibility factors, 29, 25, or 23, denoting 0.29, 0.25, or 0.23, respectively
27 = critical compressibility of 0.27

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