$$\log_{10} P(\text{torr}) = 45.559 - 6770T^{-1} - 11.575 \log_{10} T \quad (\sigma = 0.0072)$$

which gives:

$$\Delta G^\circ = 30{,}980 + 23T \ln T - 195.3T \quad \text{kcal mol}^{-1}$$

$$\Delta H^\circ = 30{,}980 - 23T \quad \text{kcal mol}^{-1}$$

$$\Delta S^\circ = 172.3 - 23 \ln T \quad \text{cal deg}^{-1}\,\text{mol}^{-1}$$

Estimated uncertainties are ±0.3 kcal mol and 0.1 kcal in ΔH° and ΔG°, respectively, and ±1.0 eu in ΔS°. The equations lead to predicted values of $\Delta H^\circ = 15.1$ kcal mol^{-1} and $\Delta S^\circ = 21.9$ eu at the normal boiling point. At the melting point of 155°, the value, $\Delta H^\circ = 5.6 \pm 1$ kcal mol^{-1}, is derived.

Thermodynamic data found in the literature for vaporization of polycyclic aromatic hydrocarbons have generally been measured in different temperature ranges and are difficult to compare without knowledge of the heat capacities of the phases involved. The results of the present study seem reasonably in accord with expectation. A comparison with perylene ($C_{20}H_{12}$), a symmetrical, planar polycyclic aromatic hydrocarbon, is of interest; this substance may be supposed to be more closely packed in the crystal than the nonplanar 9-phenylanthracene ($C_{20}H_{14}$) (2); the former is reported to have an enthalpy of sublimation of 31 kcal and an entropy of sublimation of 52.5 cal deg^{-1} mol^{-1} (at a mean temperature of 140°) (*11*). Both these values are somewhat larger than those predicted by the equations derived for 9-phenylanthracene at 140° (27 and 46.7, respectively). The latter may be expected to have lower values if steric factors reduce the lattice energy and increase the entropy in the solid state.

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Prediction of Saturated Vapor Densities for Nonpolar Substances

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A relationship was developed capable of predicting saturated vapor densities for a variety of nonpolar substances. Data available in the literature for 25 substances were correlated to produce a relationship between the reduced saturated vapor density, $\rho_{R(v)}$, and the reduced temperature, T_R, reduced pressure, P_R, and z_c, the critical compressibility factor. For the 276 points considered, an average deviation of 1.9% resulted for T_R up to 0.985 and included helium (12 points, average deviation 4.3%) and hydrogen (12 points, average deviation 2.6%). An attempt to use this relationship for polar substances such as methyl alcohol, ethyl alcohol, ammonia, sulfur dioxide, and water produced an average deviation of 2.7% for 42 data points and therefore should be acceptable for most applications.

The saturated vapor density of a substance represents a point on the saturated vapor envelope of a PVT surface and therefore depicts the limiting value of a gaseous isotherm just before condensation is initiated. Thus, the saturated vapor state becomes the point of intersection between a super-heated gas isotherm and the corresponding vaporizing liquid isotherm.

Cailletet and Mathias (5) in their pioneering work in 1886 attempted to relate the saturated liquid and saturated vapor densities with temperature. Their efforts resulted in the formulation of the empirical "rectilinear diameter rule" which states that the mean saturated density is linearly dependent on temperature:

$$\tfrac{1}{2}(\rho_{\mathrm{l}} + \rho_{\mathrm{v}}) = mT + k \tag{1}$$

which in reduced form becomes

$$\tfrac{1}{2}(\rho_{R_\mathrm{l}} + \rho_{R_\mathrm{v}}) = sT_R + t \tag{2}$$

where $s = mT_c/\rho_c$ and $t = k/\rho_c$. At the critical point, $\rho_{R(l)} = 1$, $\rho_{R(v)} = 1$, at $T_R = 1$ produces the relation, $1 = s + t$. Substituting for t in Equation 2 produces

$$\tfrac{1}{2}(\rho_{R_\mathrm{l}} + \rho_{R_\mathrm{v}}) = 1 - s(1 - T_R) \tag{3}$$

In 1957 Guggenheim (*16*) related the difference between the reduced saturated liquid and vapor densities to the reduced temperature as

$$\tfrac{1}{2}(\rho_{R_\mathrm{l}} - \rho_{R_\mathrm{v}}) = \tfrac{7}{4}(1 - T_\mathrm{R})^{1/3} \tag{4}$$

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The sum of Equations 3 and 4 yields an expression that should be capable of producing reliable saturated liquid densities. However, the difference between Equations 3 and 4 cannot be used to obtain reliable saturated vapor values since the relative magnitudes of the liquid and vapor densities are very different, when removed from the critical point region.

Barile and Thodos (*1*) have attempted to overcome this limitation by treating the saturated liquid and vapor states separately. In this connection, they used relationships involving reduced vapor pressures, P_R, to establish the corresponding reduced compressibility factors for the saturated state, $z_{R(v)}$ and $z_{R(l)}$. For the saturated vapor state, Barile and Thodos proposed the following empirical relationship:

$$z_{R_v} = A + BP_R + \frac{CD}{D + P_R} + EP_R{}^n \qquad (5)$$

where A, B, and n depend on z_c, and C and E are fixed by the boundary conditions. The value of D was a constant for all substances. Hobson and Weber (*18*) also justify the use of separate treatments for the saturated vapor and liquid states and present graphically relationships between the compressibility factor, z, and reduced pressure, P_R, for parameters of critical compressibility factors ranging from $z_c = 0.22$ to $z_c = 0.29$.

Lydersen et al. (*31*) have shown graphically that the saturated densities of pure substances are functions of either reduced temperature, T_R, or reduced pressure, P_R, and utilize the critical compressibility factor, z_c, as the third correlating parameter but do not present analytical relationships between these variables.

Equation 5 has been shown (*1*) to reproduce accurately the saturated vapor densities of 23 substances, including those of the polar substances, sulfur dioxide, ammonia, and water with an overall average deviation of 1.5% for nearly 500 points. This equation reproduces accurate saturated vapor densities, but for its application it requires values of A, B, C, E, and n for each substance. The present investigation was undertaken to attempt a more fundamental approach for the development of a relationship capable of predicting saturated vapor densities of pure substances. Of necessity, such a relationship must be simpler in form and more general in application than Equation 5.

Treatment of experimental saturated vapor densities. Experimental density measurements for the saturated vapor state for a total of 32 different substances were obtained from the available literature. Of these, 27 were nonpolar substances, including helium and hydrogen. The five polar substances were methyl alcohol, ethyl alcohol, ammonia, sulfur dioxide, and water.

Since it is expected that a more general expression will result from a study dealing with nonpolar substances, the main emphasis of this study deals with these substances. However, to extend its application, the method developed for these nonpolar substances has been applied to the five polar compounds.

To confirm the conclusion proposed by Meissner and Seferian (*40*) that besides reduced temperature, T_R, and reduced pressure, P_R, the compressibility factor at the critical point, z_c, represents a third correlating parameter, reduced saturated vapor densities, $\rho_{R(v)}$, were plotted against T_R for the nonpolar substances krypton (z_c = 0.289), carbon dioxide (z_c = 0.275), and n-octane (z_c = 0.255) as shown in Figure 1. The resulting $\rho_{R(v)}$ vs. T_R relationships, which converge at the critical point, exhibit increasing divergence with the approach of ideal state conditions realized at low reduced densities. This behavior may be rationalized by considering the reference state of vapors to be ideal. With increasing vapor pressure, this ideality does not continue to exist, and the resulting deviations must be considered until the critical point is reached. These extremes establish the two boundary conditions:

$$\rho_{R_v} = 0, \text{ for } T_R = 0 \text{ and } P_R = 0 \qquad (a)$$

$$\rho_{R_v} = 1.00, \text{ for } T_R = 1 \text{ and } P_R = 1 \qquad (b)$$

The general gaseous state behavior, which may be expressed as

$$P_V = zRT \qquad (6)$$

becomes in the reduced state

$$P_R v_R = z_R T_R \qquad (7)$$

or

$$P_R = z_R \rho_R T_R \qquad (8)$$

Boundary condition (a) suggests that for the ideal gaseous state, $z_R = 1/z_c$; therefore,

$$\rho_R T_R = z_c P_R \qquad (9)$$

To investigate deviations from Equation 9, $\rho_{R(v)} T_R$ was plotted against P_R for substances with similar z_c values. The data for argon, krypton, nitrogen, oxygen, carbon monoxide, and methane are presented on log–log coordinates in Figure 2. As expected, the slope of the resulting relationship at low P_R values is unity, and its intercept at P_R = 1 is z_c = 0.291. This procedure was applied to all the substances included in this study.

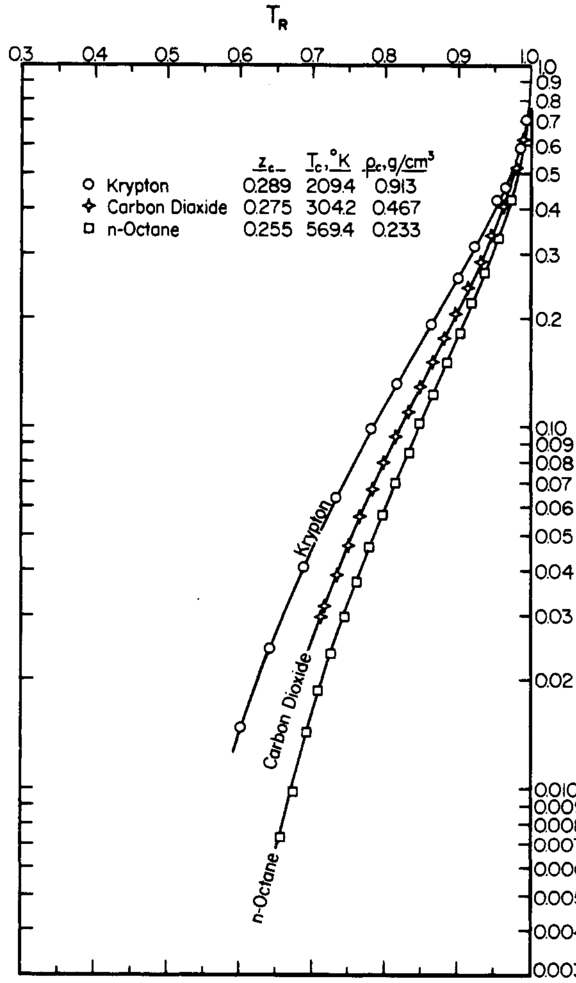

Figure 1. Relationships between $\rho_{R(v)}$ and T_R for nonpolar substances krypton, carbon dioxide, and n-octane in their saturated states

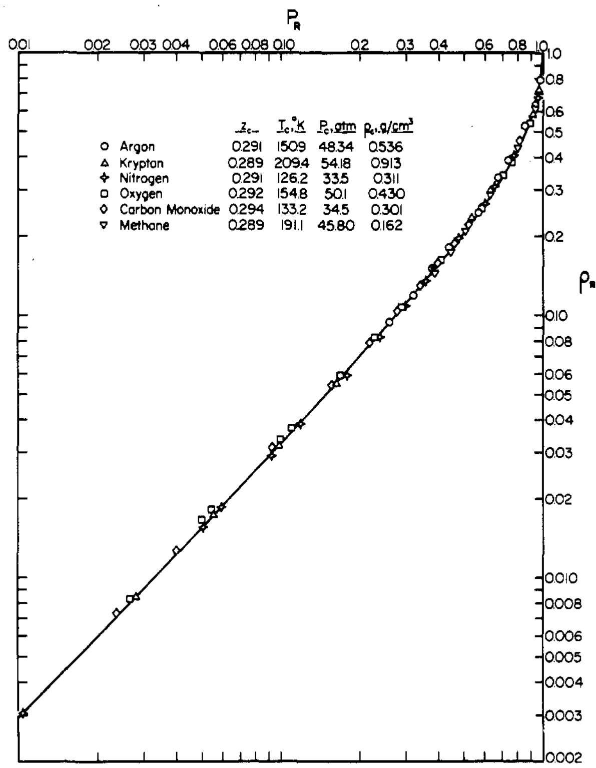

Figure 2. Relationship between $\rho_{R(v)}T_R$ and P_R for six substances having similar z_c values ($z_c \approx 0.291$)

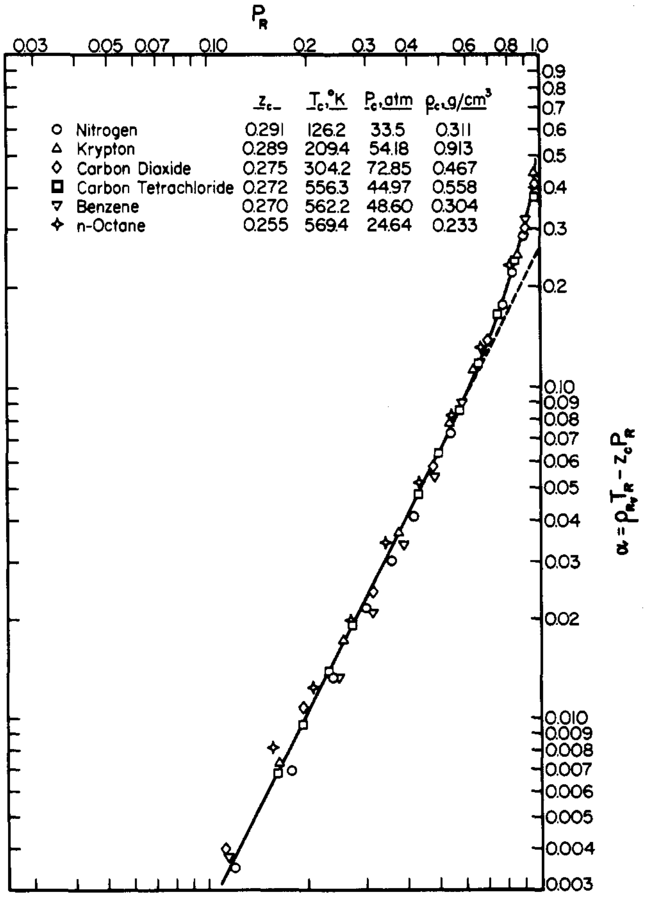

Figure 3. Relationship between α and P_R for six nonpolar substances having different z_c values

The single relationship of Figure 2 suggests that the nonideal behavior of a substance may be expressed in terms of Equation 9 as the residual quantity,

$$\alpha = \rho_{R_v}T_R - z_cP_R \qquad (10)$$

Residual quantities α were calculated for all nonpolar substances included in this study, and these values were plotted against P_R. Figure 3 presents the single relationship resulting from a plot of α vs. P_R on log-log coordinates for six different substances having different z_c values. For values below $P_R \approx 0.6$, this relationship is linear and can be expressed in equation form as follows:

$$\alpha = 0.26\,P_R^2 \qquad (11)$$

Combining Equations 10 and 11, the saturated vapor density becomes for $P_R \leq 0.60$

$$\rho_{R_v}T_R = z_cP_R + 0.26\,P_R^2 \qquad (12)$$

For reduced pressures $P_R > 0.60$, the residual quantity β follows directly from Equation 12 to be

$$\beta = \rho_{R_v}T_R - z_cP_R - 0.26\,P_R^2 \qquad (13)$$

Boundary condition (b) requires that $\beta = 0.74 - z_c$ when $T_R = 1.00$, $P_R = 1.00$, and $\rho_{R(v)} = 1.00$. To satisfy both boundary conditions, the analytical form of this second residual was taken to be of the form:

$$\beta = (0.74 - z_c)\mathrm{P_R}^w(2 - P_R^x)^y \qquad (14)$$

where w, x, and y are constants. Exponent w was taken to be unity, and x was varied until a straight line resulted when $\beta/(0.74 - z_c)P_R$ was plotted against $2 - P_R^x$ on log-log coordinates. When $x = 3$, the quantity $\beta/(0.74 - z_c)P_R$ gave a linear relationship having a slope of $y = -6.33$. Thus, Equation 14 becomes

$$\beta = \frac{(0.74 - z_c)P_R}{(2 - P_R^3)^{6.33}} \qquad (15)$$

Combining Equations 13 and 15, the final expression for the reduced saturated vapor density becomes

$$\rho_{R_v} = \frac{P_R}{T_R}\left[z_c + 0.26\,P_R + \frac{0.74 - z_c}{(2 - P_R^3)^{6.33}}\right] \qquad (16)$$

Equation 16 relates T_R and the corresponding reduced vapor pressure, P_R, and includes z_c as a basic parameter to account for the limiting conditions of the ideal gaseous state. In this final form, Equation 16 satisfies both boundary conditions.

Comparison of Calculated and Experimental Saturated Vapor Densities

Equation 16 has been applied for the calculation of saturated vapor densities for all 32 substances for which experimental measurements are available. These substances with their critical constants are listed in Table I. The sources of experimental saturated vapor densities are also listed.

To apply Equation 16, the vapor pressures corresponding to each temperature were obtained for monatomic gases (*17*), diatomic gases (*49*), hydrocarbons (*4, 44, 53, 54*), and the miscellaneous substances (*49*). For these calculations a CDC 3400 digital computer was used. The results and comparisons with experimental values for the 32 substances are given elsewhere (*15*), and four of the nonpolar substances, krypton, carbon monoxide, *n*-pentane, and carbon dioxide are listed in Table II. The average deviations of each of the 32 substances are listed in Table I. The average deviation of

Table I. Critical Constants, Sources of Experimental Data, and Average Deviations for Substances Included in This Study

						Av dev, %	
	z_c	T_c, K	P_c, atm	ρ_c, g/cm^3	Sources of saturated vapor densities	Barile and Thodos (*1*)	This investigation
Helium	0.314	5.20	2.26	0.0695	*2, 14, 27, 35*	3.2	4.3
Neon	0.307	44.5	26.86	0.484	*34*	1.8	2.1
Argon	0.291	150.9	48.34	0.536	*10, 42*	1.3	1.9
Krypton	0.289	209.4	54.18	0.913	*36*	0.5	0.9
Xenon	0.290	289.7	57.64	1.099	*45*	2.6	2.5
Hydrogen	0.305	33.2	12.81	0.0310	*22, 33*	2.1	2.6
Nitrogen	0.291	126.2	33.5	0.311	*39*	2.5	2.4
Oxygen	0.292	154.8	50.1	0.430	*23, 38*	3.4	3.1
Carbon monoxide	0.294	133.2	34.5	0.301	*32*	1.7	1.8
Chlorine	0.275	417.2	76.1	0.573	*60*	2.2	2.8
Methane	0.289	191.1	45.80	0.162	*8*	3.2	3.1
Ethylene	0.284	283.1	50.50	0.215	*37*	2.7	3.5
Ethane	0.284	305.4	48.20	0.205	*29, 47, 51*	1.1	1.1
Propane	0.279	370.0	42.00	0.220	*12, 48*	1.7	1.7
1,3-Butadiene	0.270	425.2	42.70	0.245	*52*	0.8	1.2
n-Butane	0.274	425.4	37.43	0.228	*11, 26, 51*	0.9	1.7
i-Pentane	0.267	491.0	32.92	0.234	*59*	1.1	0.8
n-Pentane	0.269	469.8	33.31	0.232	*30, 50, 56*	1.6	0.9
Benzene	0.270	562.2	48.60	0.304	*59*	2.5	3.2
Cyclohexane	0.272	553.0	40.00	0.273	*59*	0.8	1.8
2,3-Dimethylbutane	0.267	500.5	23.34	0.241	*59*	0.6	1.3
n-Hexane	0.264	507.9	29.94	0.233	*55*	1.0	1.2
n-Heptane	0.260	540.6	26.95	0.235	*25, 57*	0.5	1.9
2,5-Dimethylhexane	0.262	550.0	24.55	0.237	*59*	2.1	1.6
n-Octane	0.255	569.4	24.64	0.233	*58*	3.4	1.9
Carbon dioxide	0.275	304.2	72.85	0.467	*21, 41, 46*	1.1	1.6
Sulfur dioxide	0.269	430.7	77.81	0.525	*6, 9, 24*	4.5	3.6
Ammonia	0.242	405.5	111.3	0.235	*3, 13*	3.4	3.5
Carbon tetrachloride	0.272	556.3	44.97	0.558	*59*	0.7	1.4
Methyl alcohol	0.219	513.2	78.50	0.272	*59*	2.1	2.0
Ethyl alcohol	0.248	516.3	62.96	0.275	*59*	0.7	2.1
Water	0.231	647.4	218.3	0.32	*7, 19, 20, 28, 43*	1.7	2.4

Table II. Comparison Between Calculated Saturated Vapor Densities and Values Obtained from Experimental Data

		ρ_{R_v}				ρ_{R_v}		
T_R	P_R	Exptl	Equation 16	Dev, %	P_R	Exptl	Equation 16	Dev, %
		Krypton, $z_c = 0.289$				Carbon monoxide, $z_c = 0.294$		
1.000	1.000	1.000	1.000	0.0	1.000	1.000	1.000	0.0
0.985	0.917	0.590	0.605	2.5	0.919	0.610	0.613	0.5
0.975	0.865	0.517	0.515	−0.4	0.862	0.532	0.514	−3.3
0.950	0.744	0.400	0.397	−0.8	0.740	0.400	0.397	−0.8
0.925	0.636	0.323	0.322	−0.4	0.630	0.315	0.321	1.8
0.900	0.540	0.261	0.263	0.9	0.527	0.252	0.258	2.2
0.850	0.379	0.173	0.176	1.6	0.368	0.168	0.172	2.1
0.800	0.256	0.114	0.116	1.5	0.245	0.1125	0.1113	−1.0
0.750	0.165	0.0735	0.0742	0.9	0.157	0.0726	0.0713	−1.8
0.700	0.0997	0.0455	0.0456	0.3	0.0932	0.0448	0.0431	−3.8
0.650	0.0557	0.0267	0.0265	−0.8	0.0500	0.0246	0.0240	−2.3
0.600	0.0281	0.0141	0.0141	0.0	0.0236	0.01225	0.0120	−1.9
0.550					0.0099	0.0054	0.0054	0.0
		n-Pentane, $z_c = 0.269$				Carbon dioxide, $z_c = 0.275$		
1.000	1.000	1.000	1.000	0.0	1.000	1.000	1.000	0.0
0.985	0.898	0.550	0.550	0.0	0.905	0.560	0.568	1.5
0.975	0.835	0.462	0.461	−0.3	0.840	0.480	0.471	−1.9
0.950	0.694	0.342	0.342	0.0	0.700	0.350	0.351	0.2
0.925	0.572	0.262	0.265	1.2	0.584	0.268	0.277	3.2
0.900	0.468	0.207	0.207	0.0	0.480	0.211	0.218	3.1
0.850	0.303	0.125	0.126	0.9	0.315	0.131	0.135	2.8
0.800	0.186	0.0745	0.0752	0.9	0.195	0.0805	0.0808	0.4
0.750	0.107	0.0428	0.0432	0.9	0.113	0.0468	0.0467	−0.1
0.700	0.0561	0.0233	0.0232	−0.4	0.0592	0.0254	0.0251	−1.4
0.650	0.0263	0.0120	0.0114	−5.0				

the nonpolar substances, excluding helium and hydrogen, is 1.9%. For helium and hydrogen the deviations are 4.3 and 2.6%, respectively. For the five polar substances, the average deviation is 2.7%.

This initial test for this limited number of polar substances suggests that Equation 16 produces values of a good degree of accuracy. However, a general conclusion concerning its application for polar substances must be reserved until this work is extended to include a broader range of this class of compounds.

The equation suggested by Barile and Thodos (*1*) was applied to the same data used in this study. Equation 5 produced saturated vapor densities which for the same points give the average deviations listed in Table I. The average deviation for the nonpolar substances, excluding helium and hydrogen, is 1.7%, and the average deviation for the five polar substances is 2.5%. These comparisons show that these two methods produce comparable results. However, the method of Barile and Thodos (*1*) possesses inherently a purely empirical development and does not approach the ideal gaseous state, $\rho_R = z_c P_R / T_R$. Equation 16 possesses the simplicity of having constant coefficients and exponents which account for deviations from ideal behavior.

Nomenclature

A, B, C, D, E = constants, Equation 5
k = constant, Equation 1
m = constant, Equation 1
n = exponent, Equation 5
P = pressure
P_c = critical pressure, atm
P_R = reduced pressure, P/P_c
R = gas constant
s = constant, Equation 2
t = constant, Equation 2
T = temperature, K
T_c = critical temperature, K
T_R = reduced temperature, T/T_c
v = molar volume, cm^3/g-mol
v_c = critical volume, cm^3/g-mol
v_R = reduced volume, v/v_c
w, x, y = exponents, Equation 14
z = compressibility factor
z_c = critical compressibility factor, $P_c v_c / RT_c$
z_R = reduced compressibility factor, z/z_c

Greek Letters

α = residual quantity, Equation 10
β = residual quantity, Equation 13
ρ = density, g/cm^3
ρ_c = critical density, g/cm^3
ρ_R = reduced density, ρ/ρ_c

Subscripts

v = saturated vapor
l = saturated liquid

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