The P.V.T.-Behavior of Diatomic Substances in Their Gaseous and Liquid States

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The critical compressibility factor has been used as a criterion for the correlation of the P.V.T.-behavior in the gaseous and liquid states for the diatomic gases, nitrogen, oxygen, carbon monoxide, nitric oxide, chlorine, and hydrogen chloride. Reduced densities for these substances have been calculated from available literature data. These are presented as functions of reduced temperature and reduced pressure to produce extensive density correlations for nitrogen, oxygen, and carbon monoxide. These correlations exhibit similar behavior as expected from their similar critical compressibility factors.

The validity of the critical compressibility factor as a correlating parameter has been verified by the favorable comparison of the P.V.T.-behavior of substances other than diatomic gases including argon, krypton, xenon, and methane. These results indicate that the P.V.T.-behavior of substances having critical compressibility factors of approximately 0.291 can be predicted from the reduced state correlation developed in this study as long as polarity effects are absent.

Considerable attention is presently directed to the study of the P.V.T.-behavior of substances. The object of these investigations is to obtain generalized means for predicting reliable values of the thermodynamic and transport properties of both gases and liquids. The introduction of the critical compressibility factor as a third correlating parameter (*41*, *47*) has produced results sufficient to warrant a further examination of this parameter when applied specifically to common diatomic substances. In this investigation emphasis has been placed on a correlation of the P.V.T.-behavior of nitrogen, oxygen, and carbon monoxide. Since the critical compressibility factors for these compounds are essentially identical, a unique density correlation should result which should be applicable to other substances having similar critical compressibility factors.

The dependence of reduced density on both reduced temperature and reduced pressure has been shown to apply for the inert gases (*21*). In those studies argon, krypton, and xenon were found to exhibit a similar P.V.T.-behavior in accordance with their respective z_c values of 0.291, 0.292, and 0.290. On the other hand helium, with a value of $z_c = 0.306$, was found to have a unique P.V.T.-behavior,

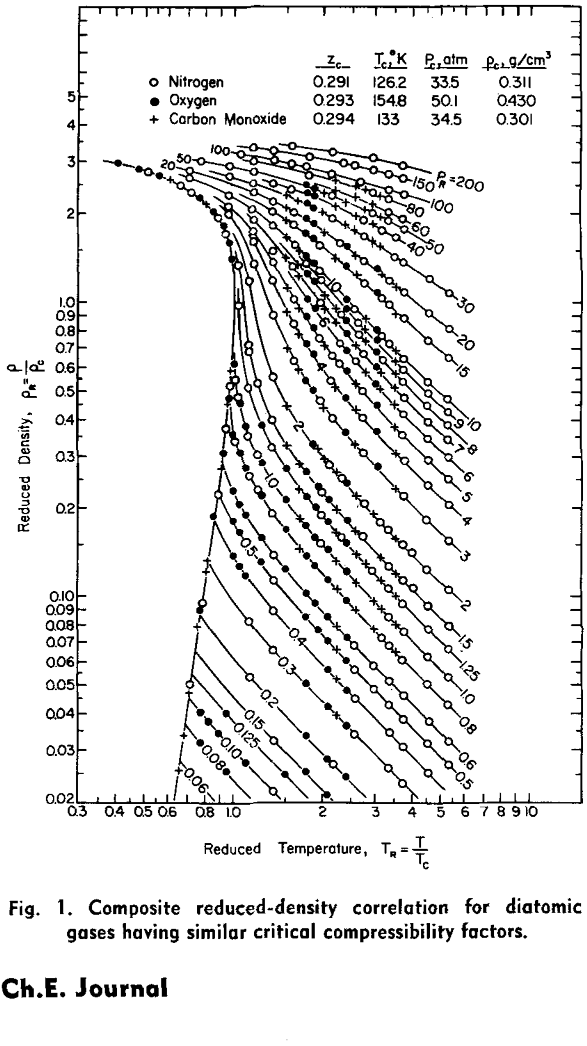

Fig. 1. Composite reduced-density correlation for diatomic gases having similar critical compressibility factors.

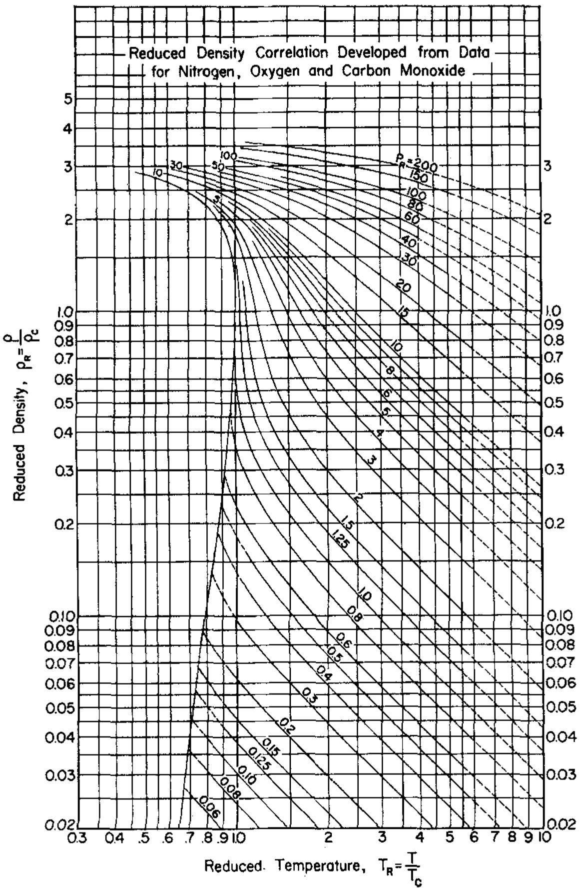

Fig. 2. Combined reduced-density correlation for nitrogen, oxygen, and carbon monoxide (log-log coordinates.)

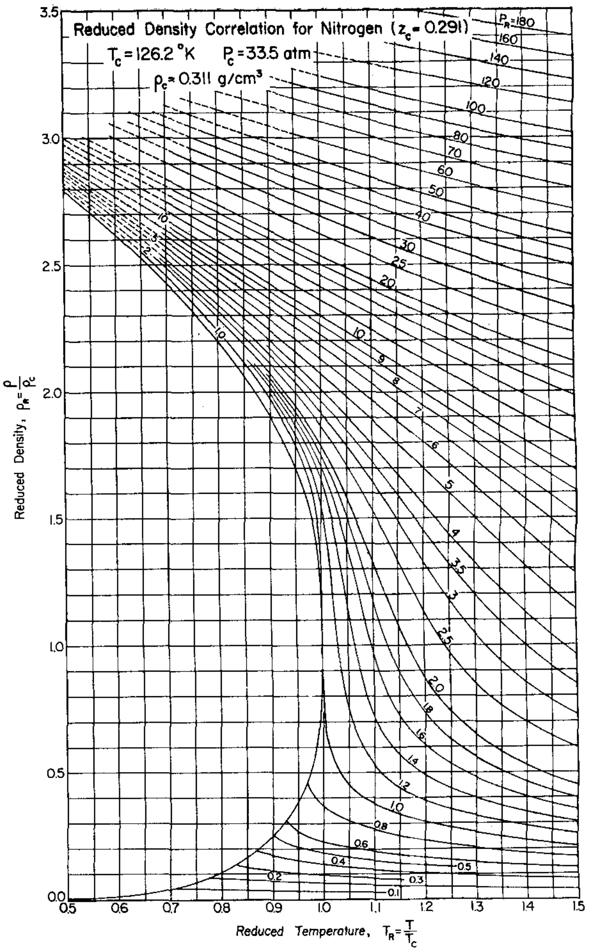

Fig. 3. Reduced-density correlation developed from nitrogen data.

while neon with a value of $z_c = 0.307$ exhibited a P.V.T.-behavior which approximated that for argon, krypton, and xenon. The extremely irregular behavior of helium is attributed to its extensive quantum deviation as accounted for by its quantum mechanical parameter (*13*). Similarly hydrogen having a value of $z_c = 0.305$ was found to behave irregularly (*65*). This behavior is also attributed to excessive quantum deviations associated with hydrogen.

The P.V.T.-behavior for eighty-two compounds having z_c values ranging from 0.23 to 0.29 are presented by Lydersen, Greenkorn, and Hougen (*41*). The present investigation has been limited to a study of the P.V.T.-behavior of diatomic gases with particular emphasis on the precise establishment of a reduced density correlation for nitrogen, oxygen, and carbon monoxide for which considerable density data are available. For other diatomic gases, such as chlorine, nitric oxide, and hydrogen chloride, a limited amount of density data are reported, and consequently a comprehensive study of their behavior cannot be presented.

The critical constants for the diatomic gases considered in this study have been obtained from Kobe and Lynn (*38*), and along with other pertinent physical constants are presented in Table 1.

Nitrogen ($z_c = 0.291$)

A comprehensive literature search for P.V.T.-data has been carried out to obtain information for the gaseous and liquid states of nitrogen. Twenty-three sources of data (*1, 2, 4, 5, 6, 7, 11, 12, 15, 17, 24, 25, 26, 29, 36, 45, 46, 54, 55, 59, 62, 67, 70*) have been consulted and utilized to produce the nitrogen values presented in Figure 1. This figure includes density values obtained as early as 1880 by Amagat (*1*) and as recent as the work of van Itterbeek, de Bock, and Verhaegen (*29*) reported in 1949.

Consistent density values were found for the saturated envelope by Mathias, Kamerlingh Onnes, and Crommelin (*45, 46*); Baly and Donnan (*5*); and van Itterbeek de Bock and Verhaegen (*29*). The data of Bartlett, Cupples, and Tremearne (*6*) and those of Holborn and Otto (*24, 25, 26*) have enabled the calculation of densities for temperatures up to $T_R = 5.33$. The high-pressure region is well established from the extensive work of Manson Benedict (*11, 12*) reported in 1937. This excellent contribution permitted the construction of isobars in Figure 1 to reduced pressures as high as $P_R = 200$. Good agreement in this high-pressure region was found to exist between the data of Benedict and those of Amagat (*4*) obtained in 1893, while those of Bridgman (*15*) were somewhat inconsistent and did not check those of Benedict except in limited regions.

Oxygen ($z_c = 0.293$)

The density data for oxygen were not as plentiful as those found for nitrogen. The data of fourteen references (*1, 3, 4, 5, 24, 27, 30, 34, 35, 39, 44, 51, 57, 69*) have been compiled to produce the oxygen density values shown in Figure 1. The data of Mathias and Kamerlingh Onnes (*44*) have been used to produce the saturated density values. The additional data of van Itterbeek and van Paemel (*30*) and those of Baly and Donnan (*5*) for the saturated liquid region were found to be consistent with those of Mathias and Kamerlingh Onnes (*44*) and therefore confirm the saturated liquid-density behavior for oxygen.

High-pressure data below a reduced temperature of $T_R = 1.77$ are unavailable. However in the temperature interval, $1.77 < T_R < 3.05$, Amagat (*1, 3, 4*) presents information up to pressures of $P_R = 20$ and up to $P_R = 60$ over a more restricted temperature interval. The contributions of Nijhoff and Keesom (*57*) and Kamerlingh Onnes and Kuypers (*35*) have produced

TABLE 1. CRITICAL CONSTANTS OF DIATOMIC GASES

	M	T_c, °K.	P_c, atm.	ρ_c, g./cc.	z_c
Bromine, Br_2	159.83	584	102	1.18	0.307
Carbon monoxide, CO	28.01	133	34.5	0.301	0.294
Chlorine, Cl_2	70.91	417.2	76.1	0.573	0.275
Hydrogen chloride, HCl	36.46	324.6	81.5	0.42	0.266
Nitric oxide, NO	30.01	180	64	0.517	0.251
Nitrogen, N_2	28.02	126.4	33.5	0.311	0.291
Oxygen, O_2	32.00	154.8	50.1	0.430	0.293

data for oxygen in the low-pressure interval, $0.08 < P_R < 2.0$

The results of this study on oxygen emphasize the lack of data in the compressed liquid region and the region in the vicinity of the critical point.

Carbon Monoxide ($z_c = 0.294$)

Only eight sources of P.V.T.-data (*5, 7, 14, 19, 42, 49, 66, 68*) were available for carbon monoxide. As was found to be the case for nitrogen and oxygen, sufficient saturated density data were also found for carbon monoxide. In this connection the saturated data of Mathias, Crommelin, Bijleveld, and Grigg (*42*) were found to be consistent with the limited data of Baly and Donnan (*5*).

The extensive data of Michels, Lupton, Wassenaar, and de Graaff (*49*) have contributed to the establishment of carbon monoxide values from $P_R = 0.6$ to $P_R = 80$. The work of Bartlett, Hetherington, Kvalnes, and Tremearne (*8*) was found to be in agreement with that of Michels et al. and permitted the establishment of density values at lower temperatures from $P_R = 0.8$ to $P_R = 30$. Density data for carbon monoxide are unavailable between the saturated values and $T_R = 1.53$.

GENERALIZED BEHAVIOR OF NITROGEN, OXYGEN, AND CARBON MONOXIDE

Since the critical compressibility factors for nitrogen, oxygen, and carbon monoxide are nearly alike, it is expected that the P.V.T.-characteristics of these substances follow a corresponding state behavior. The data of these three substances were found to be superimposable in both the gaseous and liquid regions and thus permitted the construction of a single correlation applicable to these three substances. The composite data presented in Figure 1 extend from the saturated envelope to temperatures up to $T_R = 5.33$ and pressures as high as $P_R = 200$. In order to extend the temperature of the gaseous state above $T_R = 5.33$, the Nelson-Obert compressibility factor charts (*56*) have been utilized in conjunction with the relationship

$$\frac{\rho_R}{\rho'_R} = \frac{z'}{z} \frac{P_R}{P'_R} \frac{T'_R}{T_R} \qquad (1)$$

For the case when $P_R = P'_R$, this equation enables the extension of an isobar from a single experimental density value. The correlation obtained in Figure 1 has been used to produce Figure 2. The extensions of the isobars up to $T_R = 10$, by means of Equation (1) and the Nelson-Obert compressibility factor charts, are shown as dashed lines in Figure 2. Since the upper pressure limit of the Nelson-Obert charts is restricted to $P_R = 40$, it was necessary to use information from an available reduced-density correlation for helium (*21*) in order to extend the pressure range in Figure 2 from $P_R = 40$ to $P_R = 200$. The quantum deviations present in helium are minimized at these elevated temperatures where these extrapolations were effected. These extensions in the elevated-pressure region show regularity with the general shape of the isobars produced from the experimental data of nitrogen, oxygen, and carbon monoxide. The vapor-pressure data of nitrogen have been utilized to extend the incomplete isobars to the saturated envelope. These extensions also appear as dashed lines in Figure 2. In order to present the dense-phase region for these substances in a more exacting manner the density data of nitrogen were used to construct Figure 3, where reduced density is plotted against reduced temperature on rectilinear coordinates for different reduced-pressure parameters. This rectilinear correlation allows the expansion of the saturated-liquid and dense-

TABLE 2. REDUCED DENSITY VALUES FOR NITROGEN

T_R	Saturated vapor	Saturated liquid	$P_R=0.1$	$P_R=0.2$	$P_R=0.4$	$P_R=0.6$	$P_R=0.8$	$P_R=1.0$	$P_R=1.2$	$P_R=1.4$	$P_R=1.6$	$P_R=1.8$	$P_R=2.0$	$P_R=3.0$
0.50	0.002	2.791						2.808					2.822	2.839
0.60	0.0130	2.619						2.638					2.662	2.683
0.70	0.0431	2.426						2.455					2.486	2.520
0.80	0.103	2.199	0.0393	0.0835				2.239					2.293	2.342
0.90	0.246	1.902	0.0340	0.0710	0.164			1.972	1.995	2.014	2.035	2.053	2.071	2.144
0.95	0.382	1.700	0.0321	0.0665	0.148	0.255		1.788	1.832	1.868	1.895	1.921	1.944	2.035
1.00	1.000	1.000	0.0302	0.0622	0.137	0.227	0.368	1.000	1.545	1.652	1.706	1.750	1.788	1.918
1.05			0.0287	0.0588	0.127	0.206	0.310	0.450	0.747	1.148	1.384	1.501	1.575	1.784
1.10			0.0273	0.0557	0.118	0.191	0.277	0.377	0.529	0.718	0.965	1.173	1.325	1.644
1.20			0.0248	0.0505	0.105	0.166	0.233	0.307	0.389	0.490	0.612	0.735	0.872	1.326
1.30			0.0229	0.0462	0.0955	0.148	0.204	0.264	0.330	0.399	0.479	0.562	0.652	1.044
1.40			0.0213	0.0428	0.0870	0.134	0.182	0.233	0.289	0.343	0.403	0.462	0.528	0.855
1.50			0.0198	0.0398	0.0799	0.123	0.168	0.209	0.258	0.303	0.353	0.398	0.447	0.728

T_R	$P_R=4$	$P_R=6$	$P_R=8$	$P_R=10$	$P_R=20$	$P_R=30$	$P_R=40$	$P_R=60$	$P_R=80$	$P_R=100$	$P_R=120$	$P_R=140$	$P_R=160$	$P_R=180$
0.50	2.852	2.882	2.899	2.928										
0.60	2.704	2.740	2.770	2.800	2.943	3.051								
0.70	2.548	2.595	2.636	2.679	2.840	2.958	3.046							
0.80	2.378	2.440	2.502	2.553	2.738	2.867	2.964	3.112						
0.90	2.201	2.284	2.363	2.424	2.634	2.777	2.882	3.048	3.185					
1.00	2.011	2.128	2.226	2.300	2.537	2.693	2.806	2.985	3.127	3.247	3.335			
1.10	1.792	1.967	2.087	2.175	2.441	2.608	2.734	2.926	3.069	3.192	3.293	3.380	3.468	
1.20	1.552	1.803	1.948	2.052	2.352	2.532	2.667	2.869	3.017	3.142	3.251	3.345	3.432	3.495
1.30	1.326	1.627	1.805	1.927	2.267	2.460	2.603	2.813	2.968	3.095	3.210	3.304	3.390	3.461
1.40	1.129	1.454	1.662	1.806	2.188	2.390	2.545	2.762	2.924	3.054	3.171	3.264	3.350	3.428
1.50	0.963	1.282	1.514	1.688	2.117	2.326	2.494	2.717	2.886	3.016	3.132	3.226	3.314	3.395

phase regions for the procurement of more precise values of reduced density. Accurate reduced-density values for nitrogen are presented in Table 2. These values were obtained from the original enlarged plot of Figure 3. A limited extrapolation has been indicated in the highly compressed regions that appear in Figure 3 as dashed lines. The recent density data for argon reported by Michels, Levelt, and de Graaff (*48*) have been utilized to confirm the region in the vicinity of the critical point. This approach was necessary since no data for nitrogen exist in this region, preventing the continuation of isobars in the vicinity where their slopes become excessive.

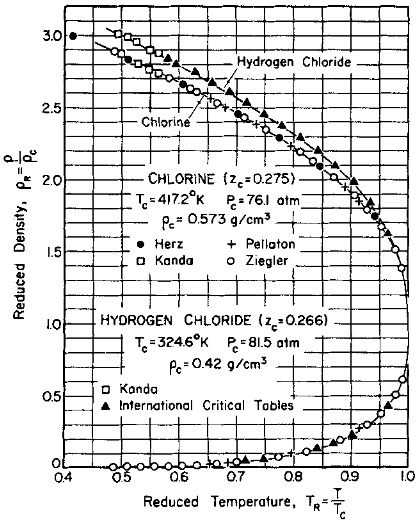

Fig. 4. Saturated vapor and liquid states for chlorine and hydrogen chloride.

OTHER DIATOMIC GASES

A limited amount of density data for diatomic gases other than nitrogen, oxygen, and carbon monoxide are presented in the literature. In general such information is confined to the saturated vapor and liquid regions. For fluorine (*18, 32, 37, 72*), hydrogen fluoride (*31*), hydrogen bromide (*28*), and hydrogen iodide (*28*) for which saturated data exist this information could not be translated to reduced values because of the lack of reliable critical constants. Critical constants are available for chlorine and hydrogen chloride. The saturated density data of Herz (*22*), Pellaton (*60*), Ziegler (*73*), and Kanda (*37*) for chlorine and Kanda (*37*) for hydrogen chloride have been reduced to produce the saturated envelopes presented in Figure 4. The reduced densities of the saturated vapor for chlorine and hydrogen chloride were found to be essentially alike, whereas those of the saturated liquid were found to differ. This difference at $T_R = 0.50$ amounts to 4.5% deviation.

Comparisons of densities for the saturated liquid state for chlorine and hydrogen chloride with nitrogen values taken from Figure 3 show that the reduced densities for nitrogen are always lower than those for chlorine and hydrogen chloride at corresponding reduced temperatures. With decreasing z_c values, reduced-density values increase. This trend is evident for nitrogen and chlorine since no polarity effects exist. For hydrogen chloride the magnitude of increase in density is the result of not only a lower z_c value but also of dipole moment effects. Wesson (*71*) reports for hydrogen chloride a dipole moment of $\mu = 2.15$ debye units. A limited amount of data for chlorine are reported (*61, 63*) for the gaseous state at low pressures in the vicinity of $P_R = 0.01$. These data have not been included in this study because the order of magnitude of these pressures falls below the range of interest.

Although no saturated data are reported for nitric oxide, Golding and Sage (*20*) and Briner, Biedermann, and Rother (*16*) present data for the superheated region as shown in Figure 5. The data of these investigators exhibit a lack of continuity. From their general trend the behavior of the data of Golding and Sage appear to be consistent with the data of nitrogen, oxygen, and carbon monoxide. The lack of a direct correspondence of the data for nitric oxide with those of Figure 2 can be accounted for by its critical compressibility factor, $z_c = 0.251$.

CONCLUSIONS

Figures 2 and 3 represent the results of this investigation. In order to generalize the application of these correlations and to establish a magnitude of accuracy experimental density values for nitrogen, oxygen, and carbon monoxide were compared with those resulting from Figures 2 and 3. For values over the entire range of both plots the following average deviations result:

	Figure 2			Figure 3		
		% deviation			% deviation	
	Points	Avg.	Max.	Points	Avg.	Max.
Nitrogen	47	0.52	2.41	—	—	—
Oxygen	43	0.88	4.91	12	1.27	2.76
Carbon monoxide	50	0.95	7.88	7	2.32	5.29

From these comparisons it will be noted that the experimental values for nitrogen are in agreement with the values resulting from Figure 2. Similar comparisons for oxygen values with Figures 2 and 3 show average deviations of 0.88 and 1.27%, respectively, with the maximum deviations found to exist in the saturated vapor region. Similarly values from Figures 2 and 3 produced average deviations of 0.95 and 2.32%, respectively, for carbon monoxide. The deviation of 2.32% is the result of comparison of values existing on the saturated envelope.

In addition experimental density values for argon (*15, 23, 33, 53*), krypton (*10, 43*), xenon (9, *52*), and methane (*40, 50, 58*) have been compared with the corresponding values resulting from Figures 2 and 3 to produce the deviations reported below:

		Figure 2		Figure 3	
	z_c	Points	% deviation	Points	% deviation
Argon	0.291	14	2.10	11	1.82
Krypton	0.292	12	1.26	7	0.68
Xenon	0.290	11	1.13	9	2.06
Methane	0.289	16	0.68	14	0.47

These comparisons show that the density for methane is in excellent agreement with those resulting from Figures 2 and 3, while those for krypton and xenon can be considered to be generally good. The significant deviation of 2.06% for xenon is the result of comparisons of high-pressure values above $P_R = 5$. For argon the data of Bridgman (*15*) have been included in these comparisons and are restricted to the high-pressure dense-phase region. The resulting deviations for argon reflect that the data of Bridgman (*15*) are significantly different from the values resulting from the correlations developed in these studies.

Application of the critical compressibility factor for density studies of nonpolar substances indicates that good agreement can be expected. This

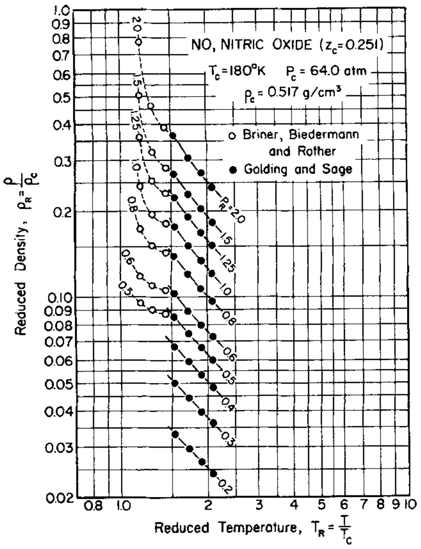

Fig. 5. Reduced-density behavior of nitric oxide.

study advances the utility of the critical compressibility factor as a valid correlating parameter when quantum deviations and dipole moment effects are negligible.

NOTATION

M = molecular weight, g./g.-mole

P = pressure, atm.

P_c = critical pressure, atm.

P_R = reduced pressure, P/P_c

R = gas constant, 82.055 (cc.) (atm.)/(g.-mole) (°K.)

T = absolute temperature, °K.

T_c = critical temperature, °K.

T_R = reduced temperature, T/T_c

V = molar volume, cc./g.-mole

V_c = critical molar volume, cc./g.-mole

z = compressibility factor, PV/RT

z_c = critical compressibility factor, P_cV_c/RT_c

μ = dipole moment, debye units

ρ = density, g./cc.

ρ_c = critical density, g./cc.

ρ_R = reduced density, ρ/ρ_c

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