Reduced Density Correlation for Carbon Dioxide

Gaseous and Liquid States

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MODERN technological advances, particularly in the field of high pressures, require a more exacting knowledge of physical properties in the gaseous state, and in the liquid and dense phase regions of substances. Thermodynamic and transport properties, as well as both physical and chemical equilibria, are dependent on the density prevailing at the temperature and pressure conditions of interest.

Several equations of state have appeared in the literature (*4, 5, 7, 50, 60*) for the prediction of *P-V-T* behavior of gases at elevated pressures and conditions in the vicinity of the critical point. Of these equations, those capable of predicting the *P-V-T* behavior accurately require several constants and involve calculations that ordinarily become prohibitive without modern computer techniques. On the other hand, the equations of state that require fewer constants do not possess the exactness possible with the more comprehensive equations of state. A comprehensive comparison of the validity of various equations of state for carbon dioxide has been presented by Davy and Bell (*16*). Although these relationships are capable of producing acceptable values of density at elevated pressures, they become less accurate when applied to the critical region. These equations of state, although capable of producing reasonable density values for the gaseous state, are inadequate in the liquid region.

Compressibility factor correlations have been presented for the estimation of the *P-V-T* relations of gases. Hougen and Watson (*25*), Dodge (*19*), and, of late, Nelson and Obert (*43*) have prepared compressibility factor charts that enable the calculation of densities for the gaseous state. Pitzer and others (*47*) introduce the "acentric factor" as a third parameter in addition to reduced temperature and pressure in order to correlate more exactly the *P-V-T* behavior of many gases.

For the estimating of densities in the liquid region, Watson (*62*) has introduced the ω-factor. Because the compressibility factor, z, and ω-factors do not necessarily produce density values that are continuous, Lydersen, Greenkorn, and Hougen (*34*) have applied the theorem of corresponding states to both gases and liquids simultaneously. As a result of their development, the theorem of corresponding states becomes applicable to nonpolar substances having similar critical compressibility factors (*37*). Hamrin and Thodos (*23*) examined the *P-V-T* behavior of argon, krypton, and xenon which have critical compressibility factors, 0.291, 0.292, and 0.290, respectively. These inert gases produce a single correlation when the reduced density, ρ_R, is plotted as a function of reduced temperature, T_R, for parameters of reduced pressure, P_R.

Tabulated density data are presented for a number of substances by Hilsenrath and others (*24*) and Din (*18*). Kendall and Sage (*28*) and Price (*49*) present thermodynamic functions calculated from *P-V-T* data for carbon dioxide. The density data of Cook (*14*), McCormack and Schneider (*36*), and Schäfer (*54*) on carbon dioxide are expressed in terms of virial coefficients, and as such do not present direct experimental values. As a result, virial type data for carbon dioxide are not included here. *P-V-T* data for carbon dioxide are reported by Cooper and Maass (*15*) and Maass and Mennie (*35*) for pressures ranging up to 1 atm., while those of Cawood and Patterson (*13*) are at 0° C. in the limited pressure range of 0.9578 to 2.8717 mm. of mercury. Their low pressure data do not appear here because pressures below 2.0 atm. fall outside the range of this study.

An extension of this study is presented to include the density behavior of carbon dioxide for the liquid and gaseous states. In this investigation, the theorem of corresponding states has been re-examined for substances having critical compressibility factors similar to carbon dioxide. From these generalized considerations the reduced density correlation of carbon dioxide should prove useful for predicting thermodynamic (*20,48*) and transport properties (*9, 44*) at elevated temperatures and pressures.

DEVELOPMENT OF REDUCED DENSITY CORRELATION

For the construction of this correlation experimental density values reported in the literature have been considered beginning with the early work of Thilorier (*59*) in 1835 and D'Andreef (*17*) in 1859 and more recently the work of Kennedy (*30*) in 1954. Although, data can be accounted for as early as 1835, reliable density values did not become available until the work of Amagat (*1*) in 1891.

In this study, the critical values reported by Michels, Blaisse, and Michels (*40*) have been used; these are $T_c = 304.2°$ K, $P_c = 72.85$ atm., and $\rho_c = 0.467$ gram per cc. More recent work by Wentorf (*63*) and Palmer (*45*) corroborates these critical values.

For the development of the saturated vapor line, the available data of Jenkin and Pye (*27*), Plank and Kuprianoff (*48*), and Michels, Blaisse, and Michels (*40*) have been used to establish the saturated vapor density from the critical point to a reduced density of $\rho_R = 0.01$. To complete the saturation envelope, the liquid densities reported by Behn (*6*) have been included in addition to those of Plank and Kuprianoff (*48*) and Michels, Blaisse, and Michels (*40*). The data of these investigators enable the establishment of the saturated liquid line from the critical point to the triple point of carbon dioxide. Since the density at the triple point is not reported in the literature, this value has been estimated from the saturated liquid line and the vapor pressure data of Meyers and Van Dusen (*38*) at the triple point, $-56.602°$ C. and 3885.2 mm. of mercury. Following this procedure, the reduced density at the triple point has been determined as $\rho_R = 2.52$.

Density data are reported either in absolute units or relative units. The absolute units can be converted directly to reduced densities, whereas the relative units, expressed in either Amagat densities or Amagat units, depend upon the density at a selected reference state. To prepare the final reduced density correlations with convenient parameters of reduced pressure, it became necessary to cross plot all density values, except those of the saturated envelope. Thus, it became possible to translate the experimental density data and present them at suitable reduced pressures from $P_R = 0.03$ to $P_R = 50$ and for a reduced temperature as high as $T_R = 4.18$. The density data of Michels, Michels, and Wouters (*42*) extend this correlation over a limited temperature range up to a pressure of $P_R = 50$, whereas the data of Kennedy (*30*) account for pressures of $P_R \leq 20$ and for temperatures ranging from the saturated envelope to $T_R = 4.18$. In addition to the data of these investigators, Amagat (*1*), Keesom (*29*), and Michels and Michels (*41*) contribute experimental values that establish the *P-V-T* behavior of carbon dioxide in the critical and dense phase regions. The areas covered by the different investigators are presented in Figure 1 on log-log coordinates to emphasize the less dense region, and in Figure 2 on rectilinear coordinates to accentuate the vicinity around the critical point. For the completeness of these correlations, considerable credit should be given to Kennedy (*30*), whose experimental data have enabled the extension of the temperature and pressure ranges where no other data exist.

Densities have been extrapolated into regions where no data exist by using the experimental density value corresponding to the highest temperature as a reference point and the compressibility factor chart of Nelson and Obert (*43*). As a result, each isobar was extended with the relationship,

$$\frac{\rho_R}{\rho_{R_1}} = \frac{z_1}{z}\,\frac{T_{R_1}}{T_R}\,\frac{P_R}{P_{R_1}} \qquad (1)$$

where the subscript refers to the corresponding conditions

Experimental density data for carbon dioxide expressed in reduced states

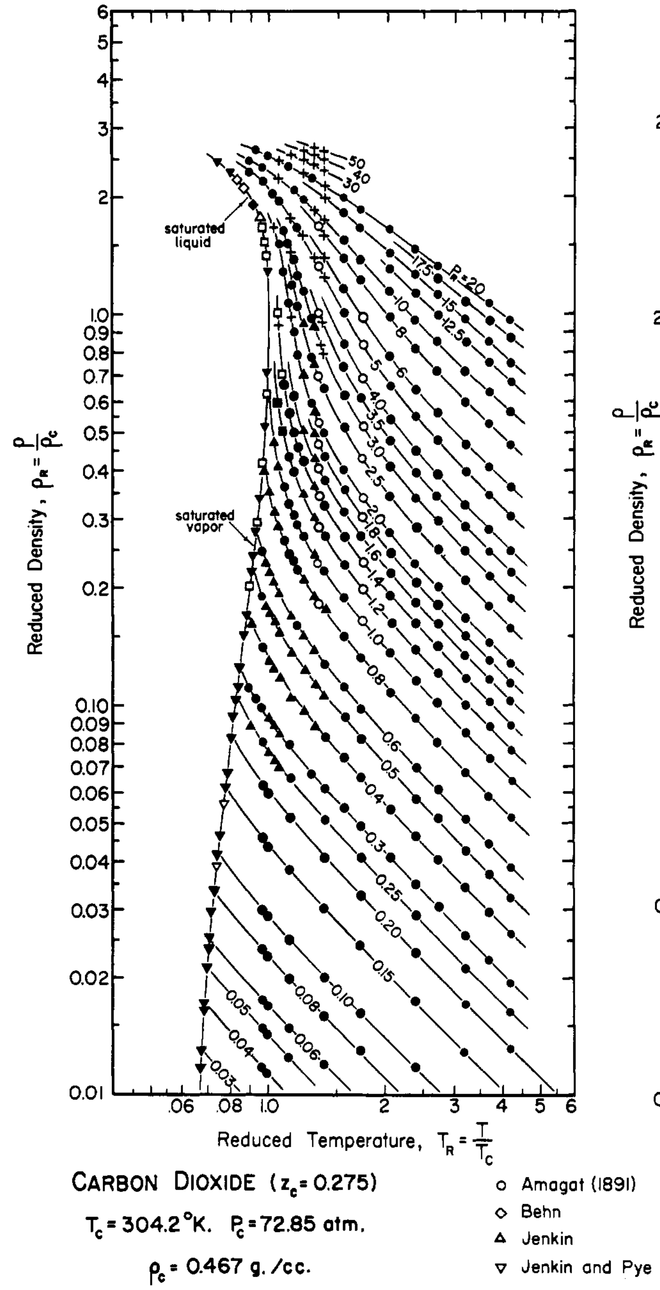

Figure 1. Log-log coordinates

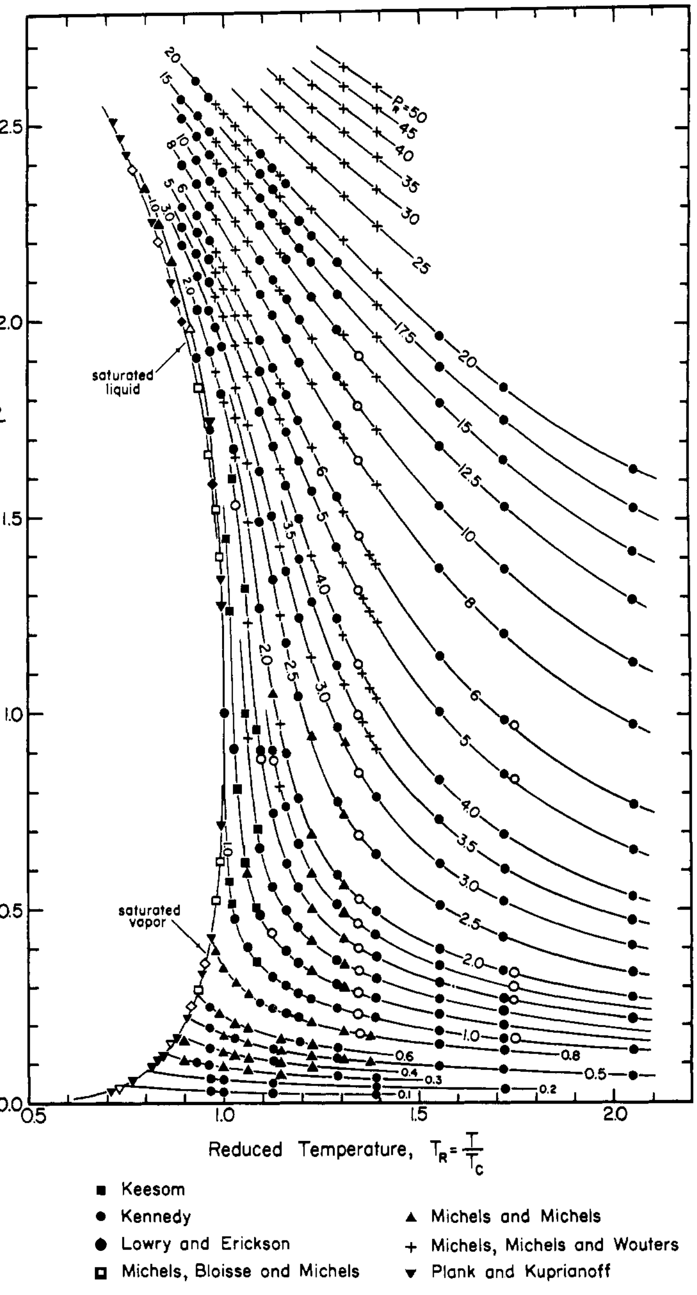

Figure 2. Rectilinear coordinates

Reduced density correlation for carbon dioxide

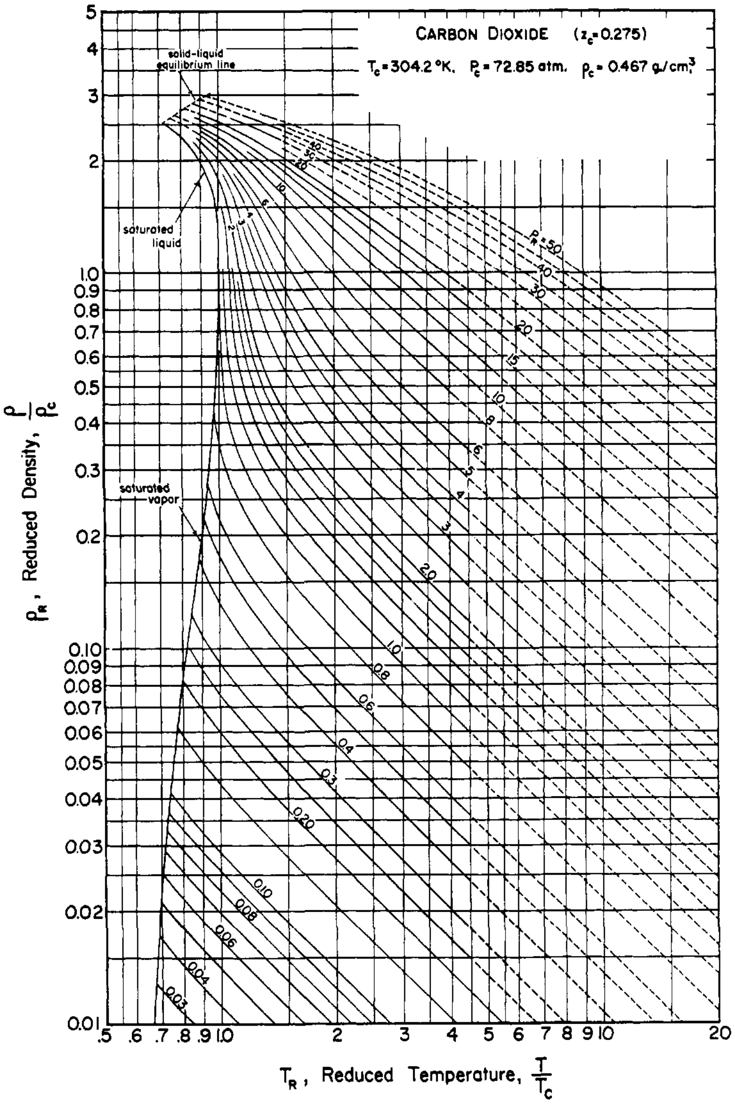

Figure 3. Log-log coordinates

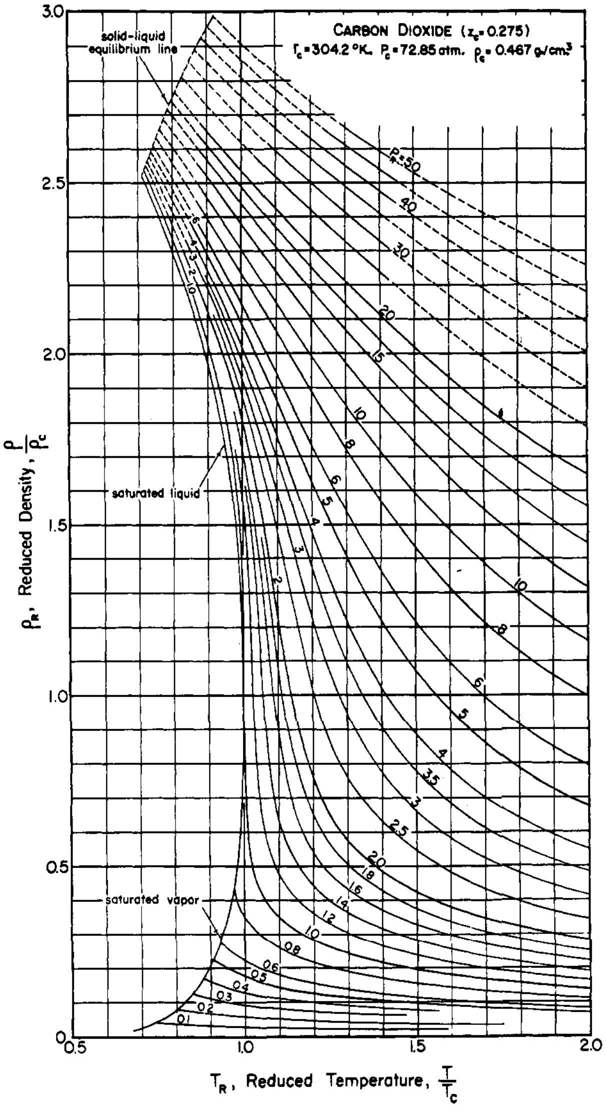

Figure 4. Rectilinear coordinates

of the experimental value. Since these calculations involve extensions of constant pressure lines, Equation 1 reduced to

$$\frac{\rho_R}{\rho_{R_1}} = \frac{z_1}{z}\frac{T_{R_1}}{T_R} \tag{2}$$

Equation 2 has been used to extend the densities to temperatures of T_R = 20 for constant pressure lines of $P_R \leqq 20$. These calculated regions appear in Figures 3 and 4 as dashed lines. Densities in the high pressure region above P_R = 20 have been calculated with Equation 1 for the isothermal conditions when $T_R = T_{R_1}$. With this restriction, Equation 1 becomes

$$\frac{\rho_R}{\rho_{R_1}} = \frac{z_1}{z}\frac{P_R}{P_{R_1}} \tag{3}$$

For these calculations, the reference densities were obtained from the isobar P_R = 20, and the compressibility factors corresponding to this isobar from the Nelson-Obert charts. As a result, the high pressure region has been extended to include pressures up to P_R = 50. The calculated portions of these curves are also shown as dashed lines in Figures 3 and 4. To complete the dense phase region properly, the isobars were extended to the solid-liquid equilibrium line established from the data of Bridgman (*11*), Michels, Blaisse, and Hoogschagen (*39*), and Tammann (*57*).

The vapor pressure data of Meyers and Van Dusen (*38*) have been used to locate the limiting values of the isobars terminating on the saturated vapor line. Figures 3 and 4, presented for completeness, cover the regions represented by experimental data and those obtained through the computational procedures. Reduced density values for temperatures up to T_R = 20 and pressures up to P_R = 50 are presented in convenient intervals in Table I.

REDUCED VAPOR PRESSURE EQUATION

Vapor pressure data available in the literature for carbon dioxide have been comprehensively examined in order to obtain reliable values, starting with the early work of Faraday in 1845, Regnault in 1862, and Andrews in 1876. Of the 14 references examined (*2, 3, 10, 21, 22, 31, 32, 38, 40, 46, 48, 51, 61, 65*), the data obtained by Meyers and Van Dusen at the National Bureau of Standards and those reported by Amagat (*2*), Michels, Blaisse, and Michels (*40*), Plank and Kuprianoff (*48*), and Zelany and Smith (*65*) represent data for carbon dioxide that are internally consistent. Following an approach similar to that proposed by Sondak and Thodos (*55*), these data were used to develop the vapor pressure function of carbon dioxide in terms of reduced temperature and reduced pressure as follows:

$$\log P_R = 4.2397 - \frac{4.4229}{T_R} - 5.3795 \log T_R + 0.1832 \frac{P_R}{T_R^2} \tag{4}$$

Equation 4 is capable of evaluating accurate vapor pressures that range from the triple point to the critical point and accounts for the reversal of curvature of the vapor pressure function in the vicinity of the critical point. The validity of Equation 4 has been checked with six vapor pressure measurements reported by Myers and Van Dusen (*38*) that cover the entire vapor pressure curve. The reported and calculated vapor pressure values in millimeters of mercury are:

Temp., ° K.	Meyers and Van Dusen (*38*)	Equation 4
217.94	4,122.1	4,122.0
228.15	6,246.8	6,229.8
243.16	10,717	10,675
283.16	33,759	33,682
288.16	38,149	38,075
298.16	48,247	48,246

The deviation between the actual and the calculated values is 0.18% for these six vapor pressures.

Table I. Reduced Density Values for Carbon Dioxide

	Saturated									
T_R	Vapor	Liquid	Solid	$P_R = 0.03$	$P_R = 0.04$	$P_R = 0.06$	$P_R = 0.08$	$P_R = 0.10$	$P_R = 0.20$	$P_R = 0.30$
0.712	0.0268	2.520	2.520	0.0120	0.0164	0.0248				
0.750	0.0477	2.425	2.631	0.0113	0.0153	0.0232	0.0313	0.0407		
0.800	0.0822	2.296	2.752	0.0106	0.0142	0.0216	0.0291	0.0376	0.0822	
0.850	0.129	2.146	2.861	0.0100	0.0132	0.0200	0.0272	0.0346	0.0750	0.121
0.900	0.211	1.967	2.953		0.0124	0.0188	0.0254	0.0324	0.0692	0.108
0.950	0.346	1.733			0.0117	0.0177	0.0239	0.0303	0.0640	0.0988
0.975	0.470	1.550			0.0114	0.0171	0.0232	0.0295	0.0618	0.0952
1.000	1.000	1.000			0.0111	0.0166	0.0226	0.0288	0.0598	0.0918
1.05					0.0106	0.0158	0.0214	0.0272	0.0562	0.0855
1.10					0.0101	0.0152	0.0203	0.0259	0.0531	0.0803
1.20						0.0138	0.0185	0.0236	0.0478	0.0723
1.40						0.0118	0.0158	0.0200	0.0401	0.0605
1.60						0.0103	0.0137	0.0174	0.0348	0.0525
1.80							0.0122	0.0154	0.0307	0.0463
2.00							0.0109	0.0138	0.0276	0.0415
2.50								0.0111	0.0219	0.0329
3.00									0.0182	0.0272
4.00									0.0136	0.0202
5.00									0.0109	0.0160
6.00										0.0132

T_R	$P_R = 0.40$	$P_R = 0.60$	$P_R = 0.80$	$P_R = 1.00$	$P_R = 1.2$	$P_R = 1.4$	$P_R = 1.6$	$P_R = 1.8$	$P_R = 2.0$	$P_R = 3.0$
0.750				2.460					2.492	2.518
0.800				2.332					2.375	2.413
0.850				2.191					2.253	2.304
0.900	0.165			2.024					2.121	2.189
0.950	0.146	0.256		1.828					1.980	2.067
0.975	0.138	0.235	0.411	1.672			1.833		1.900	1.998
1.000	0.132	0.221	0.351	1.000	1.564		1.718		1.808	1.928
1.05	0.122	0.197	0.292	0.421	0.659	1.112	1.375		1.556	1.770
1.10	0.113	0.180	0.260	0.351	0.476	0.631	0.893	1.072	1.220	1.597
1.20	0.0998	0.156	0.218	0.284	0.372	0.442	0.540	0.650	0.758	1.221
1.40	0.0819	0.126	0.171	0.218	0.267	0.318	0.373	0.427	0.486	0.773
1.60	0.0700	0.107	0.143	0.182	0.217	0.257	0.298	0.337	0.382	0.583
1.80	0.0618	0.0936	0.125	0.158	0.186	0.219	0.252	0.284	0.320	0.480
2.00	0.0553	0.0835	0.111	0.141	0.165	0.192	0.222	0.250	0.280	0.415
2.50	0.0438	0.0657	0.0872	0.109	0.131	0.152	0.173	0.195	0.217	0.319
3.00	0.0363	0.0546	0.0722	0.0904	0.108	0.126	0.142	0.159	0.178	0.262
4.00	0.0272	0.0402	0.0537	0.0671	0.0806	0.0933	0.106	0.119	0.132	0.195
5.00	0.0216	0.0323	0.0432	0.0537	0.0644	0.0748	0.0853	0.0954	0.106	0.157
6.00	0.0178	0.0270	0.0360	0.0448	0.0538	0.0625	0.0710	0.0795	0.0884	0.131
8.00	0.0134	0.0202	0.0271	0.0338	0.0402	0.0468	0.0534	0.0600	0.0667	0.0990
10.0	0.0106	0.0161	0.0216	0.0269	0.0321	0.0374	0.0428	0.0481	0.0538	0.0793
15.0		0.0107	0.0143	0.0179	0.0215	0.0252	0.0287	0.0324	0.0363	0.0536
20.0			0.0107	0.0134	0.0161	0.0188	0.0216	0.0244	0.0274	0.0402

T_R	$P_R = 4.0$	$P_R = 5.0$	$P_R = 6.0$	$P_R = 8.0$	$P_R = 10.0$	$P_R = 15$	$P_R = 20$	$P_R = 30$	$P_R = 40$	$P_R = 50$
0.750	2.543	2.571	2.598							
0.800	2.449	2.481	2.514	2.558	2.604	2.698				
0.850	2.350	2.388	2.423	2.478	2.533	2.633	2.712	2.853		
0.900	2.247	2.291	2.332	2.397	2.458	2.568	2.654	2.797	2.910	
0.950	2.134	2.189	2.238	2.316	2.382	2.503	2.594	2.744	2.858	2.959
0.975	2.078	2.136	2.190	2.275	2.341	2.472	2.565	2.717	2.833	2.934
1.000	2.022	2.088	2.142	2.233	2.305	2.439	2.537	2.691	2.811	2.909
1.05	1.896	1.975	2.042	2.148	2.225	2.374	2.479	2.641	2.763	2.865
1.10	1,760	1.864	1.941	2.062	2.146	2.311	2.424	2.593	2.718	2.821
1.20	1.479	1.623	1.733	1.888	1.992	2.284	2.313	2.500	2.631	2.738
1.40	1.021	1.218	1.364	1.570	1.717	1.959	2.112	2.329	2.476	2.592
1.60	0.782	0.956	1.095	1.315	1.485	1.758	1.932	2.172	2.334	2.469
1.80	0.640	0.784	0.913	1.128	1.299	1.586	1.777	2.028	2.206	2.359
2.00	0.552	0.673	0.795	0.997	1.158	1.445	1.650	1.91	2.100	2.265
2.50	0.421	0.519	0.605	0.768	0.903	1.20	1.41	1.71	1.89	2.07
3.00	0.347	0.427	0.502	0.640	0.764	1.02	1.23	1.52	1.72	1.91
4.00	0.258	0.318	0.378	0.483	0.583	0.788	0.983	1.26	1.46	1.64
5.00	0.209	0.256	0.306	0.394	0.474	0.648	0.825	1.07	1.28	1.45
6.00	0.175	0.215	0.258	0.332	0.400	0.551	0.708	0.942	1.14	1.40
8.00	0.133	0.163	0.195	0.252	0.306	0.427	0.561	0.755	0.930	1.08
10.0	0.107	0.132	0.158	0.204	0.248	0.348	0.461	0.636	0.789	0.923
15.0	0.0722	0.0892	0.108	0.139	0.172	0.242	0.327	0.455	0.574	0.684
20.0	0.0543	0.0678	0.0822	0.106	0.131	0.186	0.251	0.357	0.456	0.545

GENERALIZED APPLICATION OF REDUCED DENSITY CORRELATION

The dipole moment of carbon dioxide, as presented by Wesson (*64*) from nine references, ranges from 0 to 0.303. The average dipole moment from this source is μ = 0.13 debye units. Because of this small order of magnitude of the dipole moment, carbon dioxide can be considered to be essentially a nonpolar compound. Under these conditions, the *P-V-T* behavior of carbon dioxide should be applicable to nonpolar substances having comparable critical compressibility factors, $z_c = 0.275$.

To test this conslusion, densities for seven substances having z_c values from 0.273 to 0.277 have been calculated from Figures 3 and 4 and compared with the experimental values found in the literature. The nonpolar substances, acetylene (*53*), propane (*56*), propylene (*12*), *n*-butane (*52*), and chlorine (*66*) have been considered in this study as well as dichlorodifluoromethane, Freon 12 (*8*), and methyl chloride (*58*) which possess dipole momnets of 0.51 and 1.97 debye units, respectively. In these comparisons density values have been considered for the saturated liquid and vapor states as well as for regions away from the saturated envelope. For chlorine, propane, propylene, Freon 12, and methyl chloride the densities calculated from Figures 3 and 4 for the liquid states produced an average deviation of 1.32% for 20 experimental values with a maximum deviation 0. 3.78% for methyl chloride. For the saturated vapor, experimental density values for chlorine, propane, propylene, and Freon 12 deviated by 4.13% from the calculated values of 21 points. However, methyl chloride exhibited excessive deviations for the saturated vapor state. This may be explained from the highly polar nature of methyl chloride. Calculated densities for 77 values, away from the region of saturated conditions, produced deviations of 1.43% for all seven compounds examined. The significant differences encountered with methyl chloride in its saturated state exist also for the gaseous state, which amounted to 2.42% for 14 values. As a result of this study, it is reasonable to expect that polar compounds will not follow a corresponding states behavior comparable to nonpolar substances having similar critical compressibility factors.

NOMENCLATURE

P = pressure, atm.
P_c = critical pressure, atm.
P_R = reduced pressure, P/P_c
T = temperature, ° K.
T_c = critical temperature, ° K.
T_R = reduced temperature, T/T_c
z = compressibility factor
ρ = density, grams per cc.
ρ_c = critical density, grams per cc.
ρ_R = reduced density, ρ/ρ_c

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