

# **THERMODYNAMIC PROPERTIES OF HIGHLY NONSPHERICAL FLUIDS<sup>1</sup>**

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## **ABSTRACT**

The thermodynamic properties can be represented and predicted from the extended corresponding-states principle that remarkably improves the three-parameter corresponding-states principle of Pitzer et al. The extended corresponding-states principle is a four-parameter method in which the additive corresponding-states parameter is defined in terms of deviation of the critical compression factor of a real molecule from that of spherical molecule argon. The extended corresponding-states principle that is applicable to substances whose force fields deviate strongly from spherical symmetry requires only substance-dependent critical parameters and acentric factor. With revised to the method of Pitzer et al and of Lee and Kesler to extend highly nonspherical molecules, satisfactory results are obtained from the triple point to the highest temperatures and pressures over which experimental data exist for simple, nonpolar, polar, hydro-bonding and associating compounds. It has been shown that the extended corresponding-states principle is substantially more general and better than that obtainable from any available methods for the description and prediction of the entire surface properties of a highly nonspherical fluid.

**KEY WORDS:** compressibility factor; corresponding-states principle; critical parameters; enthalpy; entropy; fugacity; heat capacity; liquid; thermodynamic properties; vapor.

# 1. INTRODUCTION

## 1.1 Background

There has been interest in reduced equation of state for the purpose of generalizing the volumetric behavior of pure substances. Van der Waals was one of the first to recognize the utility of such reduced expression. With increasing application of the thermodynamic properties of pure substances at elevated pressures, there has been a revival of interest in generalizing methods for predicting their properties. Accurate generalized procedures for the calculation of thermodynamic properties of fluids and mixtures are required for the design of chemical processes. The two-parameter corresponding-states method applies accurately only to spherical molecules argon, krypton, and xenon. Suitable procedures are also available for nonpolar fluids. For polar fluids, the available equation of state are limited to specific fluids and ranges of conditions, or the procedures have not been presented in form to enable the calculation of derived thermodynamic properties.

Of the many predictive approaches that have been proposed, the corresponding-states principle has proven to be the most powerful framework as proposed by Leland and Chappelear [1]. The corresponding-states principle was the most useful and accurate derivative of the work of van der Waals on his famous equation of state [2]. The general corresponding-states principle has proven to be much better than has often been thought of in the past as stated by Huber and Hanley [3]. The methods based on the principle are theoretical and predictive, rather than empirical and correlative. The corresponding-states principle has a firm basis in statistical mechanics and kinetic theory, and has a great range and high accuracy. The corresponding-states principle should not only be able to represent data to a reasonable degree but, more importantly it does what a correlation cannot do, *i. e.*, accurately predicts the properties beyond the range of existing data as described by Huber and Hanley [3]. The simple general corresponding-states principle that accurately extends the theory of Pitzer *et al* for normal molecules to highly nonspherical substances was recently developed [4-8]. In this study, the extended corresponding-states method has been presented for the accurate calculation of the compressibility factor and derived thermodynamic properties of polar fluids for wide ranges of temperature and pressure.

## 1.2 Prior Work and Present Need

The early work is illustrative of the application of reduced variables in generalizing the properties of pure substances [9-11]. Newton [12] summarized the behavior of pure substances in regard to their agreement with the corresponding-states principle. Keyes [13] set forth some of the relationships of a reduced equation of state and made available appropriate coefficients for a number of pure substances. The molecular conditions for strict conformity with this principle have been fairly well established [14,15]. Meissner and Seferian [16] made a significant contribution to the correlation of the volumetric behavior of gases by the use of compressibility factor at the critical state as a correlating parameter. They obtained significant in terms of a universal function of the reduced pressure, temperature, and compressibility factor at the critical state. Cook and Rowlinson [17] carried out an exsensive evaluation of the deviation of pure substances from this principle. It appears that corresponding-states principle as originally proposed by van der Waals yields uncertainties of several per cent even for gases. In order to extend the corresponding-states principle to molecular fluids, it was necessary to take into account the non-spherical nature of real molecules through the acentric factor of Pitzer *et al.* [18]. Opfell *et al.* [19] gave an application of Benedict equation to the theory of corresponding states of Pitzer *et al.* Curl and Pitzer [20] represented volumetric and thermodynamic properties of fluids, enthalpy, free energy and entropy. The Pitzer method works well for a wide range of nonpolar substances but its predictive capability for polar and associating molecules is generally poor.

The use of four parameters for polar fluids corresponds to an intermolecular potential function in which dispersion and dipole-dipole interaction effects are accounted for. To extend the corresponding-states principle of Pitzer *et al.* to highly nonspherical substances, the recently developed extended corresponding-states parameter,  $\theta = (Z_c - 0.29)^2$ , is introduced to describe the deviation of the critical compression factor of a real nonspherical molecule from spherical molecules [4-8], where

$Z_c = p_c / R\rho_c T_c$  is the critical compression factor. The corresponding-states parameter  $\theta$  is obtained from the parameters of the behavior in the critical state, which are effected by the dipole and quadrupole moments. The extended corresponding-states parameter  $\theta$  reflects the physical behavior displayed by the dipole and quadrupole moments and measures the behavior of the deviation of a highly nonspherical molecule from that of the spherical molecule argon, as the similar definition of acentric factor. The relation between the critical compression factor and acentric factor,  $Z_c = 0.29 - 0.08\omega$  [18], is only approximate for nonpolar substances and can not be applicable to highly nonspherical substances. The extended corresponding-states parameter  $\theta$  significantly improves to reflect the behavior of polar substances.

The generalized relationships of Pitzer *et al.* [18] and of Lee and Kesler [21] for the thermodynamic properties of nonpolar fluids have been combined with accurate equation of state for polar substances which is utilized as the reference substances for reduced temperatures from 0.5 to 4 and reduced pressures to 10. The input data required are critical temperature, critical pressure, critical density, and acentric factor. Tabulated values of the polar fluid correction terms as well as simple and normal fluids are presented for the compressibility factor, fugacity coefficient, enthalpy, entropy, and heat capacity departures for the complete range investigated.

## 2. PRESENT METHOD

The corresponding-states principle is the most effective and widely used to represent and predict a property. While the basic corresponding-states principle has a rigorous basis for spherical fluids by van der Waals, the compressibility factor  $Z$  (or an another property) is correlated with the reduced temperature  $T_r = T/T_c$ , reduced density  $\rho_r = \rho/\rho_c$  or reduced pressure  $p_r = p/p_c$  with  $T_c$ ,  $p_c$ , and  $\rho_c$  the critical parameters to reduce temperature  $T$ , pressure  $p$ , and density  $\rho$ ,

$$Z = f_{vdW}(T_r, p_r) = Z^{(0)}(T_r, p_r) \quad (1)$$

Many suggestions have been offered to retain the general concept yet allow increase in accuracy and applicability to the two-parameter corresponding-states principle. In general, the more successful modifications have involved the inclusion of an additional third parameter into the function expressed by Eq. 1. The third parameter is the acentric factor, proposed by Pitzer *et al.*, which is a measure of the nonsphericity of a molecule's force field; *e.g.*, a value of  $\omega = 0$  denotes rare-gas spherical molecules. The expansion is employed:

$$Z = f_{\text{Pitzer}}(T_r, p_r, \omega) = Z^{(0)}(T_r, p_r) + \omega Z^{(1)}(T_r, p_r) \quad (2)$$

where  $Z^{(0)}$  is for spherical molecules, and the  $Z^{(1)}$  term is a deviation function for weakly nonspherical fluids, usually normal fluids. Pitzer et al. [18] described  $Z^{(0)}$  and  $Z^{(1)}$  as function of  $T_r$  and  $p_r$ . Lee and Kesler [21] revised the method using the more recent experimental data. The corresponding-states principle of Pitzer *et al* and of Lee and Kesler excludes systems containing highly polar and exhibiting specific interactions like associating and hydrogen bonding.

Accurate general relations for polar fluids should include four characteristic parameters for each substance to account for size and polarity effects by means of the acentric factor and polarity effect to extend the three-parameter corresponding-states principle applicable to polar molecules. Wu and Stiel [22] proposed a fourth parameter as the polarity factor obtained from an experimental pressure-density-temperature data point. Wilding and Rowley [23] developed the radius of gyration and a liquid density at a known condition.

To extend the corresponding-states principle of Pitzer et al to highly nonspherical substances for the representation of a property, the compressibility factor of a highly nonspherical fluid at a reduced temperature and pressure is determined by the extended corresponding-states principle as follows:

$$Z = f(T_r, p_r, \omega, \theta) = Z^{(0)}(T_r, p_r) + \omega Z^{(1)}(T_r, p_r) + \theta Z^{(2)}(T_r, p_r) \quad (3)$$

Where  $Z^{(2)}$  term is a deviation function for the strongly nonspherical fluid. With

$$Z^{(1)} = \frac{(Z_2 - Z_0)\theta_1 - (Z_1 - Z_0)\theta_2}{\omega_2\theta_1 - \omega_1\theta_2} \quad (4)$$

$$Z^{(2)} = \frac{(Z_1 - Z_0)\omega_2 - (Z_2 - Z_0)\omega_1}{\omega_2\theta_1 - \omega_1\theta_2} \quad (5)$$

$Z_0$  is the compressibility factor of the spherical fluid argon,  $Z_1$ ,  $\omega_1$ ,  $\theta_1$  is the compressibility factor of a weakly non-spherical fluid, and  $Z_2$ ,  $\omega_2$ ,  $\theta_2$  the compressibility factor of the strongly polar fluid. The procedure for the derived thermodynamic properties can be similarly obtained according to the corresponding-states principle. The spherical, weakly, and highly nonspherical parts of thermodynamic properties given in Tables I to III are fitted from the recent equations of state. These substances with highly accurate equations of state are argon [24], weakly nonspherical fluids nitrogen [25], methane [26], ethane [27], carbon dioxide [28], chlorodifluoromethane [29], 1,1,1,2-tetrafluoroethane [30], and strongly nonspherical polar fluids, difluoromethane [31], 1,1,1-trifluoroethane [32], 1,1-difluoroethane [33], ammonia [34], water [35]. These high-accuracy equations of state for these representative substances are used to determine  $Z^{(0)}$ ,  $Z^{(1)}$ , and  $Z^{(2)}$ , and other derived properties over the entire fluid region. The complete thermodynamic properties are obtained from the equations

$$\frac{H^0 - H}{RT_c} = \frac{H^0 - H}{RT_c} \Big|^{(0)} + \omega \frac{H^0 - H}{RT_c} \Big|^{(1)} + \theta \frac{H^0 - H}{RT_c} \Big|^{(2)} \quad (6)$$

$$\frac{S^0 - S}{RT_c} = \frac{S^0 - S}{RT_c} \Big|^{(0)} + \omega \frac{S^0 - S}{RT_c} \Big|^{(1)} + \theta \frac{S^0 - S}{RT_c} \Big|^{(2)} \quad (7)$$

$$\log(f/p) = \log(f/p) \Big|^{(0)} + \omega \log(f/p) \Big|^{(1)} + \theta \log(f/p) \Big|^{(2)} \quad (8)$$

$$\frac{C_p^0 - C_p}{RT_c} = \frac{C_p^0 - C_p}{RT_c} \left|^{(0)} + \omega \frac{C_p^0 - C_p}{RT_c} \right|^{(1)} + \theta \frac{C_p^0 - C_p}{RT_c} \left|^{(2)}\right. \quad (9)$$

### 3. COMPARISONS AND RESULTS

Comparisons between calculated and the highly accurate equations of state for the thermodynamic properties indicate that highly accurate results for the polar fluids are obtained as well as the simple and nonpolar substances. Comparison results for the compressibility factor, enthalpy, entropy, fugacity, heat capacity are given in Tables IV to V, respectively. It has been shown that the percent deviations of the compressibility factor along the critical isotherm for the vapor and liquid regions at pressures up to 10 from the theory of Pitzer et al. and the extended theory, respectively. These results have indicated that the extended theory may well yield a complete description of all classes of molecules at least one order of magnitude more accurate than is obtained from the Pitzer theory of corresponding states.

### 4. CONCLUSIONS

The relationships of this study enable the accurate calculation of compressibility factors and derived thermodynamic properties of polar fluids in the gaseous and liquid regions for wide ranges of temperature and pressure. It is shown that the compressibility factor of a polar fluid can be expressed as a linear relationship in the third and fourth parameters and the compressibility factors of a simple fluid, nonpolar and polar reference fluids at the reduced temperature and pressure. Similar linear relationships are also presented for derived thermodynamic properties, including fugacity coefficients and enthalpy, entropy, and heat capacity departures. These linear relationships are applicable for all the fluids in the gaseous region and in the liquid region for polar substances. The results can be extended for substances with larger parameters in the liquid region by the inclusion of quadratic terms in the acentric factor. The relationships presented here can also be utilized for the calculation of thermodynamic properties of polar mixtures at elevated temperatures and pressures by the use of effective parameters.

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Table I. Values of  $Z^{(0)}$ 

$T_r \diagup p_r$	0.01	0.05	0.1	0.2	0.4	0.6	0.8	1
0.55	0.9817	0.0099	0.0198	0.0396	0.0791	0.1184	0.1576	0.1967
0.6	0.9857	0.0094	0.0188	0.0375	0.0749	0.1121	0.1491	0.186
0.65	0.9886	0.009	0.018	0.0359	0.0716	0.107	0.1424	0.1775
0.7	0.9908	0.9521	0.0174	0.0347	0.0691	0.1033	0.1372	0.171
0.75	0.9924	0.9608	0.9183	0.0338	0.0674	0.1006	0.1335	0.1662
0.8	0.9937	0.9676	0.9331	0.8557	0.0665	0.0992	0.1314	0.1633
0.85	0.9947	0.9729	0.9444	0.8824	0.0669	0.0993	0.1312	0.1626
0.9	0.9955	0.9771	0.9533	0.9024	0.7811	0.1022	0.1339	0.1649
0.93	0.9959	0.9792	0.9577	0.9121	0.807	0.6639	0.1383	0.1688
0.95	0.9961	0.9804	0.9603	0.9179	0.8216	0.6981	0.1441	0.1734
0.97	0.9964	0.9816	0.9627	0.9231	0.8346	0.7256	0.5604	0.1813
0.98	0.9965	0.9822	0.9639	0.9256	0.8406	0.7376	0.5922	0.1878
0.99	0.9966	0.9827	0.9649	0.9279	0.8462	0.7487	0.6176	0.1989
1	0.9967	0.9832	0.966	0.9302	0.8515	0.7589	0.6391	0.2893
1.01	0.9968	0.9837	0.967	0.9323	0.8566	0.7685	0.6578	0.4767
1.02	0.9969	0.9842	0.968	0.9344	0.8614	0.7774	0.6745	0.5247
1.05	0.9971	0.9855	0.9707	0.9401	0.8745	0.8011	0.7157	0.6096
1.1	0.9975	0.9874	0.9745	0.9483	0.8929	0.8327	0.7665	0.692
1.15	0.9978	0.989	0.9778	0.9551	0.9077	0.8575	0.804	0.7466
1.2	0.9981	0.9903	0.9806	0.9608	0.92	0.8775	0.8331	0.7869
1.3	0.9985	0.9925	0.985	0.9698	0.939	0.9076	0.8758	0.8436
1.4	0.9988	0.9941	0.9883	0.9766	0.953	0.9293	0.9057	0.8823
1.5	0.9991	0.9954	0.9908	0.9816	0.9633	0.9451	0.9272	0.9097
1.6	0.9993	0.9963	0.9927	0.9855	0.9712	0.9571	0.9434	0.9301
1.7	0.9994	0.9971	0.9942	0.9885	0.9773	0.9664	0.9559	0.9457
1.8	0.9995	0.9977	0.9954	0.991	0.9822	0.9737	0.9656	0.9579
1.9	0.9996	0.9982	0.9964	0.9929	0.9861	0.9796	0.9734	0.9676
2	0.9997	0.9986	0.9972	0.9945	0.9893	0.9843	0.9797	0.9754
2.2	0.9998	0.9992	0.9984	0.9969	0.994	0.9914	0.989	0.9869
2.4	0.9999	0.9996	0.9993	0.9986	0.9973	0.9963	0.9954	0.9948
2.6	1	0.9999	0.9998	0.9997	0.9995	0.9997	1	1
2.8	1	1	1	1.0006	1.0011	1.0021	1.0029	1.0038
3	1	1	1.0006	1.0013	1.0025	1.0037	1.0053	1.0069
3.5	1	1.0006	1.001	1.002	1.0042	1.0065	1.0087	1.011
4	1	1.001	1.001	1.0026	1.0052	1.0075	1.0101	1.0127

Table I. Values of  $Z^{(0)}$  (continued)

$T_r \diagup p_r$	1.2	1.5	2	3	5	7	10
0.55	0.2356	0.2938	0.3902	0.581	0.9552	1.3209	1.8569
0.6	0.2227	0.2776	0.3684	0.5476	0.8983	1.2403	1.7392
0.65	0.2125	0.2646	0.3508	0.5207	0.8516	1.1732	1.6413
0.7	0.2046	0.2545	0.337	0.4989	0.8132	1.117	1.5586
0.75	0.1986	0.2468	0.3261	0.4814	0.7813	1.0703	1.4886
0.8	0.1949	0.2417	0.3185	0.4682	0.7556	1.0314	1.4287
0.85	0.1935	0.2393	0.314	0.4587	0.735	0.9988	1.3779
0.9	0.1953	0.2401	0.3129	0.453	0.719	0.9719	1.3346
0.93	0.1987	0.2428	0.3142	0.4516	0.7115	0.9582	1.3115
0.95	0.2026	0.2458	0.3161	0.4514	0.7073	0.95	1.2975
0.97	0.2086	0.2503	0.3191	0.4521	0.7038	0.9426	1.2843
0.98	0.2129	0.2533	0.321	0.4526	0.7023	0.9391	1.2775
0.99	0.2184	0.2569	0.3233	0.4534	0.701	0.9357	1.2713
1	0.2262	0.2613	0.3259	0.4545	0.6998	0.9326	1.2652
1.01	0.2378	0.2667	0.329	0.4557	0.6987	0.9295	1.2597
1.02	0.2581	0.2735	0.3326	0.4572	0.6978	0.9268	1.2539
1.05	0.4562	0.3086	0.3472	0.4632	0.6962	0.9191	1.2385
1.1	0.6065	0.4618	0.3915	0.4792	0.6966	0.9094	1.2156
1.15	0.685	0.587	0.4712	0.5045	0.701	0.9032	1.1962
1.2	0.7389	0.6654	0.5613	0.5399	0.7094	0.9	1.1806
1.3	0.8114	0.7639	0.693	0.6309	0.7369	0.9021	1.157
1.4	0.8593	0.8259	0.7763	0.7186	0.7751	0.913	1.1421
1.5	0.8927	0.8684	0.8325	0.7872	0.8174	0.9302	1.1352
1.6	0.9173	0.8993	0.8728	0.8391	0.8581	0.9506	1.1331
1.7	0.936	0.9226	0.9031	0.8786	0.8942	0.9718	1.134
1.8	0.9506	0.9405	0.9263	0.9093	0.9249	0.9921	1.1376
1.9	0.9621	0.9547	0.9445	0.9334	0.9506	1.0103	1.1422
2	0.9714	0.966	0.9591	0.9527	0.972	1.0266	1.1473
2.2	0.985	0.9827	0.9802	0.9809	1.0042	1.0531	1.1563
2.4	0.9943	0.994	0.9946	1	1.0268	1.0722	1.1636
2.6	1.0007	1.0019	1.0045	1.0128	1.0417	1.0851	1.1687
2.8	1.0054	1.0076	1.0116	1.0223	1.0529	1.0947	1.1722
3	1.0088	1.0113	1.0163	1.0288	1.0605	1.1008	1.1733
3.5	1.0136	1.017	1.0239	1.0377	1.0705	1.1086	1.1724
4	1.0156	1.0199	1.0267	1.0416	1.0741	1.1093	1.1671

Table II. Values of  $Z^{(1)}$ 

$T_r \diagup p_r$	0.01	0.05	0.1	0.2	0.4	0.6	0.8	1
0.55	0.0044	-0.0059	-0.0117	-0.0234	-0.0465	-0.0692	-0.0916	-0.1132
0.6	-0.0314	-0.0054	-0.0109	-0.0215	-0.0425	-0.0632	-0.0835	-0.1036
0.65	-0.0197	-0.005	-0.01	-0.0199	-0.0392	-0.058	-0.0766	-0.0949
0.7	-0.0134	-0.0691	-0.0094	-0.0186	-0.0367	-0.0546	-0.0715	-0.0885
0.75	-0.0074	-0.0375	-0.0795	-0.0168	-0.0325	-0.0476	-0.0626	-0.0768
0.8	-0.0051	-0.0255	-0.0521	-0.1166	-0.0313	-0.0456	-0.0593	-0.0724
0.85	-0.0034	-0.0174	-0.0346	-0.0737	-0.031	-0.0446	-0.0575	-0.0692
0.9	-0.0026	-0.0116	-0.0232	-0.0473	-0.1081	-0.0456	-0.0567	-0.067
0.93	-0.0019	-0.0091	-0.0182	-0.0362	-0.0769	-0.149	-0.0578	-0.0664
0.95	-0.0015	-0.0075	-0.0152	-0.0302	-0.0616	-0.1051	-0.0609	-0.0661
0.97	-0.0012	-0.0063	-0.0125	-0.0249	-0.0489	-0.0766	-0.1404	-0.0665
0.98	-0.0014	-0.0061	-0.0113	-0.0226	-0.0438	-0.0656	-0.1	-0.0671
0.99	-0.0014	-0.0052	-0.0103	-0.0203	-0.039	-0.0566	-0.0752	-0.0682
1	-0.0011	-0.0046	-0.0094	-0.0182	-0.0344	-0.0484	-0.0585	-0.0693
1.01	-0.0009	-0.0044	-0.0085	-0.0161	-0.0304	-0.0411	-0.0458	-0.0214
1.02	-0.0008	-0.0038	-0.0077	-0.0145	-0.0264	-0.0345	-0.0353	-0.0092
1.05	-0.0003	-0.0029	-0.0053	-0.0098	-0.0168	-0.019	-0.0135	0.0111
1.1	-0.0004	-0.0015	-0.0024	-0.0043	-0.005	-0.0011	0.0089	0.0305
1.15	0.0001	-0.0007	-0.0008	-0.0001	0.0032	0.0103	0.0227	0.0421
1.2	-0.0002	0.0004	0.0009	0.0027	0.0088	0.018	0.0315	0.0499
1.3	0.0002	0.001	0.0027	0.0062	0.015	0.0267	0.0405	0.0576
1.4	0.0002	0.0016	0.003	0.0069	0.0165	0.0281	0.0412	0.0566
1.5	0.0002	0.0018	0.0036	0.0077	0.0175	0.0287	0.0417	0.0556
1.6	0.0003	0.0019	0.0037	0.0079	0.0175	0.0283	0.0407	0.0536
1.7	0.0004	0.0019	0.0039	0.0078	0.0168	0.0271	0.0384	0.0509
1.8	0.0005	0.0017	0.0035	0.0071	0.0164	0.0261	0.0364	0.0474
1.9	0.0003	0.0015	0.0034	0.0071	0.0152	0.0243	0.034	0.044
2	0.0004	0.0014	0.0032	0.0068	0.014	0.0225	0.0317	0.0414
2.2	0.0003	0.0014	0.003	0.0058	0.0123	0.0197	0.0274	0.0359
2.4	0.0003	0.0012	0.002	0.0043	0.0109	0.0157	0.0227	0.0295
2.6	0	0.0004	0.003	0.0028	0.0089	0.0128	0.0198	0.0268
2.8	0	0	0.0015	0.0045	0.0092	0.0117	0.0179	0.0238
3	0	0.0025	0.002	0.0018	0.0084	0.0127	0.0169	0.0198
3.5	0	0.001	0.0025	0.0025	0.0052	0.0084	0.0112	0.0159
4	0	-0.002	0.0025	-0.0005	0.0028	0.0069	0.0101	0.0131

Table II. Values of  $z^{(1)}$  (continued)

$T_r \diagup p_r$	1.2	1.5	2	3	5	7	10
0.55	-0.1351	-0.1671	-0.219	-0.317	-0.4972	-0.6559	-0.873
0.6	-0.123	-0.1517	-0.1984	-0.2866	-0.4435	-0.5831	-0.7712
0.65	-0.1127	-0.1384	-0.1804	-0.2588	-0.3985	-0.5211	-0.679
0.7	-0.1046	-0.1284	-0.1664	-0.2356	-0.3586	-0.4635	-0.6013
0.75	-0.0905	-0.1107	-0.1422	-0.2003	-0.3023	-0.3898	-0.5058
0.8	-0.0852	-0.103	-0.1317	-0.1833	-0.2723	-0.3488	-0.4459
0.85	-0.0807	-0.097	-0.1218	-0.1664	-0.2433	-0.3081	-0.3915
0.9	-0.0769	-0.0903	-0.1115	-0.1495	-0.2146	-0.2706	-0.3421
0.93	-0.074	-0.0861	-0.1046	-0.1384	-0.1973	-0.2477	-0.3132
0.95	-0.0725	-0.0827	-0.0991	-0.1304	-0.1853	-0.233	-0.2943
0.97	-0.0699	-0.0782	-0.093	-0.122	-0.1734	-0.2184	-0.2755
0.98	-0.0679	-0.0751	-0.0894	-0.1167	-0.167	-0.2109	-0.2675
0.99	-0.0649	-0.0714	-0.0851	-0.1122	-0.1608	-0.2032	-0.2575
1	-0.0597	-0.067	-0.0804	-0.1069	-0.1545	-0.1959	-0.2472
1.01	-0.0471	-0.0606	-0.0751	-0.1015	-0.148	-0.1881	-0.2402
1.02	-0.0021	-0.0509	-0.0689	-0.0953	-0.1413	-0.1808	-0.228
1.05	0.1032	0.0225	-0.0413	-0.0755	-0.121	-0.1577	-0.2037
1.1	0.0707	0.1656	0.0528	-0.0306	-0.0837	-0.1195	-0.1635
1.15	0.0711	0.1332	0.1623	0.0303	-0.0428	-0.0801	-0.1194
1.2	0.0738	0.1198	0.1842	0.0986	0.0011	-0.0401	-0.0822
1.3	0.0768	0.1102	0.1666	0.1893	0.0905	0.0385	-0.0042
1.4	0.0732	0.1007	0.1476	0.2061	0.1638	0.1123	0.0689
1.5	0.0707	0.0948	0.1358	0.2007	0.2102	0.1714	0.1299
1.6	0.0674	0.0888	0.1261	0.1897	0.2331	0.2148	0.1794
1.7	0.0633	0.083	0.1168	0.1782	0.2411	0.2439	0.2236
1.8	0.0589	0.0773	0.1085	0.1665	0.2406	0.259	0.2535
1.9	0.0551	0.0717	0.1005	0.1559	0.2355	0.2702	0.2772
2	0.0512	0.0669	0.0932	0.1455	0.2267	0.272	0.2915
2.2	0.0442	0.0577	0.0814	0.1297	0.2112	0.2658	0.3123
2.4	0.0397	0.0513	0.0699	0.1142	0.1923	0.2534	0.3161
2.6	0.0339	0.0434	0.0643	0.1035	0.1786	0.2398	0.3112
2.8	0.0305	0.0389	0.0554	0.0909	0.1614	0.2243	0.299
3	0.0238	0.0341	0.0516	0.0824	0.1479	0.2131	0.291
3.5	0.0185	0.0279	0.0382	0.0668	0.1244	0.181	0.2626
4	0.0145	0.0202	0.0305	0.0541	0.1041	0.1582	0.236

Table III. Values of  $Z^{(2)}$ 

$T_r \diagup p_r$	0.01	0.05	0.1	0.2	0.4	0.6	0.8	1
0.55	-20.1179	-0.2911	-0.5834	-1.1693	-2.3618	-3.5631	-4.7975	-6.0793
0.6	-1.44	-0.3006	-0.5772	-1.1823	-2.4081	-3.642	-4.8879	-6.1548
0.65	-0.6345	-0.3041	-0.6108	-1.2259	-2.4754	-3.7421	-5.0307	-6.3136
0.7	-0.2393	-2.8433	-0.6171	-1.2427	-2.5041	-3.7707	-5.0715	-6.362
0.75	-0.241	-1.8189	-5.7648	-1.2891	-2.7165	-4.1486	-5.5637	-7.0234
0.8	-0.1137	-0.8794	-2.5102	-10.1462	-2.7746	-4.1985	-5.6362	-7.0867
0.85	-0.0449	-0.3936	-1.1549	-3.8798	-2.8056	-4.2449	-5.689	-7.1695
0.9	0.0004	-0.1529	-0.467	-1.62	-7.3662	-4.2927	-5.8069	-7.3292
0.93	-0.0049	-0.0436	-0.2092	-0.8873	-4.0182	-13.8142	-5.9451	-7.476
0.95	0.0166	-0.0095	-0.1038	-0.5487	-2.6805	-7.6214	-6.061	-7.6433
0.97	0.0141	0.0219	-0.0073	-0.2797	-1.7732	-4.819	-13.5151	-7.9196
0.98	0.02	0.0529	0.0075	-0.1801	-1.3957	-3.8807	-8.8367	-8.1215
0.99	0.0318	0.0481	0.0337	-0.0989	-1.0693	-3.1189	-6.7392	-8.3859
1	0.0238	0.0486	0.0982	-0.0266	-0.8116	-2.4866	-5.3195	-10.577
1.01	0.0054	0.094	0.1058	0.0387	-0.5607	-1.9872	-4.2557	-7.2186
1.02	0.0114	0.071	0.1426	0.1016	-0.3804	-1.5664	-3.4475	-5.9707
1.05	-0.0059	0.1222	0.1732	0.2338	0.0476	-0.6168	-1.7468	-3.429
1.1	0.0303	0.1451	0.2255	0.4093	0.463	0.1942	-0.3595	-1.3271
1.15	0.0275	0.1731	0.2876	0.4804	0.6598	0.5873	0.2682	-0.3689
1.2	0.0513	0.1638	0.2741	0.5089	0.7431	0.7664	0.5321	0.0345
1.3	0.0308	0.1518	0.2679	0.4937	0.753	0.7879	0.6626	0.2574
1.4	0.0483	0.2004	0.4072	0.7644	1.257	1.5392	1.6129	1.4739
1.5	0.0524	0.1771	0.315	0.6055	1.0006	1.2134	1.2052	1.0698
1.6	0.0211	0.1319	0.2641	0.4714	0.7711	0.9041	0.8326	0.6593
1.7	0.0151	0.0912	0.2016	0.3837	0.6042	0.6542	0.5607	0.3023
1.8	-0.0066	0.0795	0.1707	0.3067	0.414	0.3848	0.2633	0.0298
1.9	0.017	0.076	0.1282	0.2073	0.2677	0.2116	0.0535	-0.2214
2	0.0151	0.0625	0.0894	0.1455	0.1771	0.0675	-0.1599	-0.4794
2.2	0.017	0.0177	0.0117	0.0316	-0.0386	-0.2094	-0.4742	-0.8292
2.4	0.0016	-0.0152	-0.0104	-0.0444	-0.0879	-0.2498	-0.6594	-0.8353
2.6	0	-0.0142	-0.2588	0.0255	-0.2188	-0.3691	-0.7813	-1.1636
2.8	0	0	0.0761	-0.3152	-0.3631	-0.5998	-0.88	-1.3542
3	0	-0.2366	-0.0786	0.0349	-0.612	-0.8137	-1.1141	-1.5054
3.5	0	-0.214	-0.2366	-0.2366	-0.5144	-0.9238	-1.2016	-1.6567
4	0	0.1773	-0.2366	-0.1538	-0.5896	-0.9999	-1.4357	-1.827

Table III. Values of  $Z^{(2)}$  (continued)

$T_r \diagup p_r$	1.2	1.5	2	3	5	7	10
0.55	-7.3247	-9.27	-12.5631	-19.5059	-34.2389	-50.2083	-75.1965
0.6	-7.4586	-9.404	-12.7406	-19.5632	-34.6311	-50.3988	-74.5969
0.65	-7.6448	-9.6548	-13.0439	-20.0341	-34.6966	-50.2335	-74.6378
0.7	-7.6843	-9.6712	-13.0331	-20.2392	-34.9503	-50.2936	-73.8662
0.75	-8.4747	-10.6556	-14.319	-21.8428	-37.0026	-52.7286	-76.1752
0.8	-8.5336	-10.7583	-14.4354	-21.8962	-36.9431	-52.3047	-75.4081
0.85	-8.5949	-10.8238	-14.5865	-22.1115	-37.0953	-52.2016	-74.7926
0.9	-8.8213	-11.0968	-14.8711	-22.3263	-37.2638	-52.0051	-74.049
0.93	-9.0533	-11.3205	-15.1159	-22.6065	-37.4024	-52.0458	-73.7563
0.95	-9.205	-11.5025	-15.3166	-22.7901	-37.5011	-52.0358	-73.6251
0.97	-9.4843	-11.7612	-15.5637	-22.9875	-37.6156	-52.0014	-73.4968
0.98	-9.6701	-11.9465	-15.695	-23.1648	-37.6929	-52.0132	-73.1111
0.99	-9.8891	-12.1314	-15.8538	-23.2497	-37.7757	-52.0343	-73.0529
1	-10.1663	-12.3137	-16.0275	-23.4044	-37.8333	-52.027	-73.0677
1.01	-10.5497	-12.5476	-16.1934	-23.5068	-37.8803	-52.0263	-72.9122
1.02	-10.2124	-12.8189	-16.3832	-23.6773	-37.9729	-52.0419	-73.1061
1.05	-5.9826	-12.1353	-16.8724	-24.0496	-38.1505	-52.0657	-72.6757
1.1	-2.8246	-6.7924	-15.3253	-24.4118	-38.4105	-51.9718	-71.887
1.15	-1.4114	-3.9758	-10.8485	-23.6153	-38.3612	-51.7683	-71.4476
1.2	-0.7409	-2.6345	-7.8991	-21.1401	-37.8612	-51.3312	-70.5077
1.3	-0.3295	-1.674	-5.0999	-15.3902	-35.1794	-49.4759	-68.6174
1.4	1.1519	0.227	-2.2932	-10.5222	-30.9122	-47.0648	-67.1772
1.5	0.7189	-0.0948	-2.2106	-8.8584	-26.8097	-43.3231	-63.7297
1.6	0.2944	-0.4385	-2.3841	-8.0928	-23.8677	-39.7857	-60.0567
1.7	-0.0456	-0.7938	-2.5952	-7.7648	-21.8514	-36.9019	-56.9232
1.8	-0.3324	-1.1105	-2.8249	-7.5732	-20.4779	-34.4402	-54.063
1.9	-0.6436	-1.3571	-3.0375	-7.5157	-19.5028	-32.75	-51.6115
2	-0.8456	-1.6202	-3.2139	-7.4937	-18.6733	-31.249	-49.2925
2.2	-1.2171	-1.96	-3.5468	-7.613	-17.689	-29.2099	-46.1379
2.4	-1.6073	-2.2854	-3.7647	-7.7364	-17.0996	-27.5355	-43.5875
2.6	-1.6848	-2.2934	-4.0119	-7.7033	-16.6344	-26.2525	-41.5511
2.8	-2.0274	-2.5985	-4.0018	-7.4521	-15.8557	-25.1994	-39.4548
3	-1.8022	-2.5915	-4.3074	-7.6	-15.5448	-24.6998	-38.1943
3.5	-2.0477	-2.9578	-4.294	-7.4055	-14.6989	-22.7323	-35.312
4	-2.199	-2.9667	-4.3028	-7.253	-14.0494	-21.3402	-32.8054

Table IV. Calculated deviation of compressibility factor from the corresponding-states theory of Pitzer et al.

$T_r \diagup p_r$	0.01	0.05	0.1	0.2	0.4	0.6	0.8	1
0.55	10.415	14.134	14.196	14.231	14.463	14.628	14.854	15.139
0.6	0.675	16.148	15.2	15.751	16.205	16.443	16.625	16.841
0.65	0.363	17.64	17.745	17.843	18.116	18.346	18.6	18.733
0.7	0.202	1.364	18.988	19.172	19.421	19.584	19.86	20.051
0.75	0.167	0.968	2.471	19.605	21.492	22.203	22.534	22.95
0.8	0.109	0.606	1.413	4.348	22.73	23.14	23.489	23.783
0.85	0.073	0.406	0.9	2.238	23.029	23.518	23.883	24.328
0.9	0.049	0.287	0.609	1.4	4.148	23	23.868	24.512
0.93	0.049	0.233	0.488	1.094	2.882	8.592	23.607	24.313
0.95	0.042	0.209	0.431	0.945	2.349	4.925	22.99	24.072
0.97	0.035	0.181	0.38	0.816	1.953	3.724	10.34	23.641
0.98	0.037	0.165	0.356	0.76	1.796	3.305	6.327	23.23
0.99	0.038	0.168	0.343	0.719	1.652	2.97	5.228	22.275
1	0.038	0.154	0.31	0.676	1.526	2.679	4.476	16.87
1.01	0.035	0.168	0.3	0.634	1.415	2.452	3.939	7.955
1.02	0.029	0.156	0.306	0.591	1.322	2.254	3.518	5.492
1.05	0.031	0.163	0.293	0.539	1.079	1.778	2.64	3.813
1.1	0.034	0.156	0.283	0.545	0.92	1.293	1.852	2.557
1.15	0.028	0.155	0.278	0.518	0.901	1.147	1.41	1.911
1.2	0.036	0.135	0.25	0.485	0.846	1.096	1.209	1.532
1.3	0.024	0.112	0.211	0.399	0.7	0.898	1.012	1.07
1.4	0.021	0.092	0.187	0.352	0.597	0.756	0.826	0.803
1.5	0.025	0.082	0.148	0.281	0.475	0.592	0.616	0.582
1.6	0.012	0.061	0.123	0.221	0.368	0.445	0.436	0.422
1.7	0.01	0.045	0.096	0.182	0.294	0.332	0.307	0.323
1.8	0.007	0.039	0.084	0.148	0.21	0.23	0.25	0.342
1.9	0.01	0.04	0.071	0.126	0.212	0.269	0.297	0.366
2	0.01	0.043	0.075	0.136	0.234	0.301	0.335	0.4
2.2	0.01	0.033	0.07	0.139	0.246	0.317	0.373	0.456
2.4	0.008	0.017	0.119	0.129	0.286	0.359	0.461	0.473
2.6	0	0.098	0.089	0.154	0.317	0.392	0.406	0.495
2.8	0	0.1	0.087	0.126	0.249	0.381	0.449	0.59
3	0	0.081	0.112	0.127	0.263	0.393	0.459	0.593
3.5	0	0.094	0.081	0.139	0.252	0.361	0.554	0.694
4	0	0.066	0.081	0.148	0.292	0.425	0.564	0.712

Table IV. Calculated deviation of compressibility factor from the corresponding-states theory of Pitzer et al.  
 (continued)

$T_r \diagup p_r$	1.2	1.5	2	3	5	7	10
0.55	15.264	15.578	15.995	16.925	18.475	19.949	21.702
0.6	17.076	17.323	17.792	18.516	20.48	21.878	23.441
0.65	18.994	19.322	19.779	20.607	22.076	23.5	25.244
0.7	20.233	20.48	20.879	22.149	23.714	25.029	26.567
0.75	23.157	23.529	23.929	24.925	26.189	27.518	28.882
0.8	24.034	24.486	24.987	25.896	27.263	28.519	29.966
0.85	24.418	24.945	25.735	26.852	28.314	29.52	30.871
0.9	24.905	25.544	26.333	27.413	29.119	30.237	31.616
0.93	25.084	25.689	26.583	27.833	29.485	30.689	32.063
0.95	24.85	25.643	26.645	27.975	29.675	30.908	32.317
0.97	24.682	25.587	26.704	28.044	29.847	31.072	32.549
0.98	24.503	25.533	26.64	28.181	29.926	31.155	32.525
0.99	24.121	25.383	26.607	28.154	29.99	31.246	32.624
1	23.567	25.131	26.536	28.172	30.029	31.31	32.778
1.01	22.428	24.764	26.369	28.097	30.05	31.356	32.823
1.02	17.532	24.196	26.194	28.097	30.08	31.411	32.985
1.05	6.174	16.953	24.8	27.674	30.03	31.5	33.093
1.1	3.48	5.992	16.557	25.898	29.628	31.368	33.086
1.15	2.557	3.926	8.127	21.901	28.592	30.922	33.053
1.2	2.032	3.025	5.534	16.376	26.88	30.114	32.607
1.3	1.44	2.14	3.666	8.432	21.931	27.413	31.299
1.4	1.053	1.611	2.787	5.781	16.888	24.005	29.321
1.5	0.818	1.309	2.303	4.757	13.231	20.663	26.986
1.6	0.655	1.079	1.972	4.126	10.948	17.897	24.65
1.7	0.525	0.923	1.721	3.673	9.52	15.839	22.701
1.8	0.494	0.788	1.534	3.332	8.601	14.256	21.026
1.9	0.526	0.8	1.385	3.07	7.983	13.201	19.686
2	0.557	0.82	1.355	2.978	7.493	12.343	18.478
2.2	0.605	0.861	1.368	2.99	6.934	11.223	16.905
2.4	0.69	0.94	1.461	3.004	6.629	10.421	15.771
2.6	0.73	0.989	1.545	2.991	6.38	9.842	14.932
2.8	0.812	1.041	1.544	2.89	6.068	9.412	14.148
3	0.783	0.993	1.682	2.937	5.945	9.223	13.7
3.5	0.824	1.149	1.671	2.878	5.622	8.515	12.747
4	0.855	1.164	1.685	2.831	5.405	8.051	11.957

Table V. Calculated deviation of compressibility factor from the extended corresponding-states theory

$T_r \diagup p_r$	0.01	0.05	0.1	0.2	0.4	0.6	0.8	1
0.55	3.057	3.257	3.238	3.257	3.216	3.172	3.134	3.129
0.6	0.279	2.758	2.932	2.835	2.776	2.743	2.75	2.712
0.65	0.19	2.287	2.295	2.252	2.229	2.208	2.199	2.154
0.7	0.137	0.539	1.877	1.843	1.817	1.827	1.766	1.787
0.75	0.102	0.452	0.71	2.145	1.763	1.73	1.716	1.684
0.8	0.078	0.36	0.673	0.784	1.765	1.743	1.699	1.747
0.85	0.061	0.297	0.567	1.009	1.803	1.788	1.695	1.743
0.9	0.049	0.245	0.477	0.907	1.419	1.849	1.791	1.775
0.93	0.047	0.221	0.429	0.828	1.477	1.089	1.848	1.723
0.95	0.043	0.206	0.402	0.783	1.438	1.732	1.904	1.775
0.97	0.037	0.187	0.378	0.734	1.365	1.828	1.671	1.793
0.98	0.038	0.179	0.358	0.707	1.338	1.817	1.88	1.837
0.99	0.03	0.181	0.352	0.69	1.304	1.8	2.053	1.95
1	0.036	0.167	0.337	0.668	1.264	1.764	2.092	1.546
1.01	0.037	0.163	0.328	0.646	1.236	1.734	2.108	3.655
1.02	0.029	0.164	0.314	0.62	1.201	1.697	2.084	2.27
1.05	0.03	0.144	0.286	0.569	1.094	1.567	1.968	2.26
1.1	0.027	0.126	0.246	0.487	0.939	1.356	1.725	2.037
1.15	0.019	0.108	0.204	0.411	0.807	1.16	1.499	1.779
1.2	0.019	0.088	0.175	0.354	0.689	0.998	1.283	1.544
1.3	0.013	0.062	0.127	0.248	0.494	0.726	0.95	1.151
1.4	0.011	0.062	0.114	0.247	0.491	0.726	0.962	1.194
1.5	0.012	0.044	0.074	0.156	0.328	0.508	0.683	0.858
1.6	0.01	0.027	0.053	0.099	0.202	0.331	0.462	0.609
1.7	0.008	0.026	0.048	0.084	0.171	0.251	0.337	0.405
1.8	0.006	0.023	0.048	0.079	0.146	0.212	0.28	0.35
1.9	0.008	0.031	0.056	0.1	0.179	0.242	0.29	0.33
2	0.008	0.035	0.064	0.118	0.212	0.293	0.355	0.395
2.2	0.008	0.031	0.069	0.135	0.25	0.342	0.431	0.477
2.4	0.008	0.017	0.12	0.134	0.297	0.389	0.541	0.549
2.6	0	0.099	0.1	0.151	0.343	0.437	0.473	0.636
2.8	0	0.1	0.086	0.165	0.288	0.454	0.54	0.649
3	0	0.099	0.096	0.123	0.304	0.394	0.459	0.665
3.5	0	0.072	0.068	0.168	0.304	0.341	0.476	0.582
4	0	0.097	0.082	0.167	0.275	0.288	0.396	0.504

Table V. Calculated deviation of compressibility factor from the extended corresponding-states theory  
(continued)

$T_r \diagup p_r$	1.2	1.5	2	3	5	7	10
0.55	3.078	3.052	2.987	2.888	2.711	2.566	2.403
0.6	2.668	2.649	2.621	2.566	2.342	2.246	2.166
0.65	2.173	2.13	2.121	2.053	1.989	1.905	1.849
0.7	1.807	1.78	1.772	1.663	1.624	1.579	1.609
0.75	1.692	1.644	1.614	1.567	1.466	1.378	1.27
0.8	1.705	1.695	1.64	1.566	1.445	1.344	1.255
0.85	1.704	1.683	1.635	1.558	1.431	1.328	1.22
0.9	1.709	1.68	1.639	1.551	1.419	1.317	1.204
0.93	1.756	1.706	1.597	1.515	1.402	1.306	1.202
0.95	1.721	1.673	1.582	1.491	1.387	1.297	1.221
0.97	1.727	1.615	1.544	1.482	1.365	1.288	1.214
0.98	1.722	1.617	1.524	1.453	1.371	1.286	1.196
0.99	1.72	1.6	1.496	1.412	1.356	1.279	1.157
1	1.713	1.574	1.463	1.416	1.335	1.278	1.209
1.01	1.771	1.547	1.431	1.385	1.326	1.279	1.172
1.02	1.525	1.498	1.378	1.341	1.304	1.267	1.187
1.05	2.752	1.334	1.267	1.23	1.253	1.24	1.241
1.1	2.233	2.278	1.181	1.006	1.128	1.178	1.114
1.15	2.008	2.156	2.178	0.942	0.994	1.115	1.191
1.2	1.766	1.987	1.897	1.153	0.946	1.045	1.124
1.3	1.333	1.565	1.756	1.423	0.93	0.945	1.13
1.4	1.407	1.683	2.017	1.948	1.137	0.949	1.025
1.5	1.03	1.28	1.608	1.815	1.347	1.049	1.215
1.6	0.739	0.951	1.257	1.601	1.325	1.164	1.321
1.7	0.512	0.696	0.967	1.355	1.224	1.223	1.514
1.8	0.404	0.485	0.733	1.144	1.218	1.182	1.573
1.9	0.354	0.434	0.541	0.951	1.196	1.156	1.715
2	0.433	0.475	0.493	0.78	1.142	1.069	1.668
2.2	0.592	0.663	0.682	0.601	1.037	1.123	1.668
2.4	0.628	0.634	0.762	0.743	0.916	1.219	1.599
2.6	0.645	0.749	0.842	0.825	0.843	1.256	1.446
2.8	0.701	0.731	0.856	0.867	0.883	1.345	1.555
3	0.703	0.776	0.818	0.949	0.849	1.286	1.759
3.5	0.595	0.708	0.808	0.941	0.914	1.575	2.206
4	0.611	0.632	0.829	0.869	1.083	1.774	2.679