

the disperse size distribution.

Results of the 37 mass transfer runs made are presented in Figure 2. The nonuniform size distributions have been accounted for by the hydraulic mean diameter of Equation (1). From the figure, there appears to be no systematic deviation among the data for the different size distributions. Furthermore, the data have asymptotic forms corresponding to $N_{Sh} \rightarrow bN_{Pe}^{1/3}$ for small N_{Re} and $N_{Sh} \rightarrow cN_{Re}^{2/3} N_{Sc}^{1/3}$ for large N_{Re} . These asymptotic relations and the interpolation formula shown in Figure 2 were noted earlier by Karabelas et al. (1971) in the case of a uniform particle size distribution. Since the hydraulic mean diameter of Equation (1) remains valid in the limiting case of uniform particle size, one should expect the correlation shown in Figure 2 to agree with previous work. That this situation prevails may be seen upon comparison with results of Wilson and Geankoplis (1966), which are quite representative of earlier investigations:

$$\epsilon N_{Sh} N_{Sc}^{-1/3} = 1.09 N_{Re}^{1/3} \quad 0.0016 < N_{Re} < 55 \quad (2)$$

$$\epsilon N_{Sh} N_{Sc}^{-1/3} = 0.25 N_{Re}^{0.69} \quad 55 < N_{Re} < 1500 \quad (3)$$

Equations (2) and (3) are in essential agreement with the correlation in Figure 2. From the above, it is concluded that one can obtain reliable results by employing the hydraulic mean diameter when computing fluid-particle transfer coefficients in nonuniform packing.

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NOTATION

d_i = diameter of sphere in size group i , cm
 D_p = mean particle diameter (equation 1), cm

D = molecular diffusivity, cm^2/s
 G = superficial mass velocity, $\text{g}/\text{cm}^2\text{-s}$
 k_L = mass transfer coefficient, cm/s
 L = bed depth, cm
 n_i = number of particles in size group i
 N_{Re} = Reynolds number, UD_p/ν
 N_{Pe} = Peclet number, UD_p/D
 N_{Sh} = Sherwood number, $D_p k_L/D$
 N_{Sc} = Schmidt number, ν/D
 U = superficial velocity, cm/s
 ϵ = porosity
 ρ = density, g/cc
 ν = kinematic viscosity, cm^2/s

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Generalized Equations of State for Compressed Liquids—Application of Pitzer's Correlation

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Volumetric properties of compressed liquids are often required in engineering calculations. The three-parameter generalized correlation of Pitzer et al. (1955) only covers the region for $0.8 \leq T_r \leq 1.0$ and $0 \leq P_r \leq 9$. Lu et al. (1973) recently extended the correlation to the $0.5 \leq T_r \leq 0.8$ and $0 \leq P_r \leq 9.0$ region.

These correlations are all in tabular forms. This makes it difficult for computer applications where large storage areas and iterative procedures would be necessary to use the correlation. The purpose of this investigation is to find

a suitable analytic equation for the compressed liquid region.

DEFINITIONS

Pitzer (1955) introduced a third parameter called the *acentric factor* to extend the applicability of the theorem of corresponding state to normal fluids.

The acentric factor is defined as

$$\omega = -\log \frac{P_s}{P_c} - 1.000 \quad (1)$$

where P_s = vapor pressure at $T_r = 0.70$. This form is chosen to make $\omega = 0$ for the simple fluids Ar, Kr, and Xe with simple spherical molecules. Hence acentric factor is a factor that measures the deviation of the intermolecular potential function from that of the simple spherical molecules.

The compressibility factor may be written as

$$Z = Z(P_r, T_r, \omega) \quad (2)$$

It is assumed that a linear equation in ω is adequate in representing most volumetric data.

$$Z = Z^{(0)}(P_r, T_r) + \omega \cdot Z^{(1)}(P_r, T_r) \quad (3)$$

ANALYTICAL EQUATIONS BASED ON TABULATED VALUES OF PITZER AND LU

258 pairs of $Z^{(0)}$, $Z^{(1)}$ values are taken from the tabulations of Pitzer et al. (1955) and Lu et al. (1973). A linear regression method is used to fit the tabulated values to the proposed equations.

$$\frac{V^{(0)}}{V_{ci}} = f_1(T_r) \cdot T_r + f_2(T_r) \cdot T_r P_r \quad (4)$$

$$\frac{V^{(1)}}{V_{ci}} = f_3(T_r) \cdot T_r + f_4(T_r) \cdot T_r P_r + f_5(T_r) \cdot T_r P_r^2 \quad (5)$$

where

$$Z^{(0)} = \frac{PV^{(0)}}{RT} = \frac{V^{(0)}}{RT_c} \cdot \left(\frac{P_r}{T_r} \right) = \frac{V^{(0)}}{V_{ci}} \left(\frac{P_r}{T_r} \right)$$

and

$$Z^{(1)} = \frac{V^{(1)}}{V_{ci}} \left(\frac{P_r}{T_r} \right)$$

V_{ci} ($\equiv RT_c/P_c$) is the ideal critical volume as defined by Su (1946). The data-fitting procedure is carried out using $V^{(0)}/V_{ci}$, $V^{(1)}/V_{ci}$ as the dependent variables because the variation of $Z^{(0)}$, $Z^{(1)}$ is large while $V^{(0)}/V_{ci}$, $V^{(1)}/V_{ci}$ stays relatively constant throughout the region where $0.5 \leq T_r \leq 0.99$, $0 \leq P_r \leq 9.0$. All $f_i(T_r)$ functions can be properly represented by a quadratic function of T_r where $i = 1, 2, 3, 4, 5$. We find

$$Z^{(0)} = (0.46578 - 0.74371 T_r + 0.46265 T_r^2) P_r - (0.00721 - 0.02606 T_r + 0.02610 T_r^2) P_r^2 \quad (6)$$

$$Z^{(1)} = -(0.31771 - 0.61168 T_r + 0.40349 T_r^2) P_r + (0.05867 - 0.17141 T_r + 0.13623 T_r^2) P_r^2 - (0.00684 - 0.01951 T_r + 0.01446 T_r^2) P_r^3 \quad (7)$$

Eight-hundred forty-two experimental volumetric data points of 27 substances are found from the literature. They include 405 points which are chosen to reproduce the data used in Table 3 of the Lu et al. (1973) paper.

The available volumetric data are compared with the Z values calculated using Equation (3), Equation (6), and Equation (7). The overall average deviation is 2.4% and the maximum deviation is 17%. Large deviations of 10% are likely to occur near critical region when $T_r \approx 0.99$ and $P_r \approx 1.0$. This is not unexpected since the curvature is greater at the critical region than other regions. If we exclude this small region then the maximum deviation is only 11.5%. The maximum average deviation is 10.3% for 1-pentene. The tabular correlation of Lu et al. (1973) shows an average deviation of 11.3% for the same substance.

A comparison is also made between the formulated ana-

lytical equations and the tabulation of Lydersen, Greenkorn, and Hougen (1955). Lydersen et al. (1955) utilized critical compressibility Z_c as a third parameter which can be converted to acentric factor ω by using the approximation

$$Z_c = \frac{1}{1.28\omega + 3.41} \quad (8)$$

as suggested by Hougen et al. (1959). They compare favorably with the analytical equations of $Z^{(0)}$, $Z^{(1)}$. The overall average deviation is 2.1%.

ANALYTICAL EQUATIONS BASED ON ORIGINAL LITERATURE DATA OF 27 SUBSTANCES

Since $Z^{(0)}$, $Z^{(1)}$ values can be represented in analytical form, we proceed to improve the tabulated values by modifying Equation (6) and Equation (7) using the literature volumetric data of 27 different substances. Again linear regression method is used to improve $Z^{(0)}$ and $Z^{(1)}$ alternatively. First $Z^{(1)}$ is represented by Equation (7), then $Z^{(0)}$ can be re-evaluated. Second, new $Z^{(0)}$ equation is used to obtain a new equation for $Z^{(1)}$. More iterations are carried out until the overall average deviation reaches a minimum.

The improved set of equations are

$$Z^{(0)} = (0.46407 - 0.73221 T_r + 0.45256 T_r^2) P_r - (0.00871 - 0.02939 T_r + 0.02775 T_r^2) P_r^2 \quad (9)$$

$$Z^{(1)} = -(0.02676 + 0.28376 T_r - 0.28340 T_r^2) P_r - (0.10209 - 0.32504 T_r + 0.25376 T_r^2) P_r^2 + (0.00919 - 0.03016 T_r + 0.02485 T_r^2) P_r^3 \quad (10)$$

In these equations, the pressure range is extended to $P_r = 10$. A total of 867 literature data points are used. The overall average deviation is 1.9% and the maximum deviation is 12.2% which occurs at $T_r = 0.98$, $P_r = 0.9$. (See Table 1.) If we exclude this small region, the maximum deviation is 10%. The critical region volumetric behavior is slightly better represented in this newly-derived form than the Equation (6) and (7) derived using Pitzer's (1955) and Lu's (1973) tabulations. The average deviation for 1-pentene is 9.8%. Comparing with the Lydersen correlation (1957), the overall average deviation is 2.5%; the maximum deviation is 9%.

Hsi and Lu (1974) had proposed two 20 constants analytical equations to represent the volumetric behavior of $Z^{(0)}$ and $Z^{(1)}$ in the range of $0 \leq P_r \leq 9.0$ and $0.50 \leq T_r \leq 0.80$. These equations claim to represent 418 literature volumetric data points with an overall average deviation of 1.51%, and a maximum deviation of 6.8% excluding 1-pentene. Equation (9) and Equation (10) reproduce the volumetric behavior of 554 literature data points in the same temperature and pressure region with an overall average deviation of 1.69%, and a maximum deviation of 6.6% excluding 1-pentene. Equations (9) and (10) are superior than Hsi and Lu's equation (1974), however, because they have a substantially larger range of applicability and they employ fewer numerical constants.

If no acentric factor value of a given substance under consideration is available, and no reliable estimate may be obtained, then another approximation using only P_r and T_r is recommended:

$$Z = (0.46711 - 0.83113 T_r + 0.54721 T_r^2) P_r - (0.06685 - 0.19271 T_r + 0.14200 T_r^2) P_r^2 + (0.00442 - 0.01307 T_r + 0.00969 T_r^2) P_r^3 \quad (11)$$

This equation predicts the 867 volumetric data points with

TABLE 1. DEVIATION OF ANALYTICAL EQUATIONS BASED ON LITERATURE DATA OF 27 SUBSTANCES

Compound	Reduced pressure		Reduced temp.		No. of points	Acent. factor	Avg. dev.	Max. dev.	Reference
Methane	0.32	0.60	0.79	0.79	2	0.013	0.694	0.903	Vennix et al. (1970)
	0.42	5.10	0.83	0.99	10	0.013	4.385	12.193	
	0.18	6.83	0.60	0.69	17	0.013	0.750	1.700	
<i>n</i> -butane	0.11	9.08	0.73	0.73	21	0.201	0.513	0.703	Olds et al. (1944)
	0.27	9.08	0.81	0.97	50	0.201	1.422	3.037	
<i>i</i> -butane	0.19	7.56	0.76	0.76	12	0.192	0.395	0.650	Gonzalez and Lee (1966)
	0.57	9.45	0.84	0.93	17	0.192	1.235	2.387	
Neopentane	0.22	9.70	0.72	0.79	25	0.195	1.439	3.728	Gonzalez and Lee (1968)
	1.08	9.70	0.87	0.95	23	0.195	2.318	4.880	
<i>n</i> -hexane	0.19	8.66	0.73	0.78	14	0.290	0.843	1.980	Kelso and Felsing (1940)
	0.45	6.90	0.83	0.93	12	0.290	1.744	3.727	
2-methylpentane	0.19	8.66	0.75	0.80	14	0.295	0.746	1.812	Kelso and Felsing (1940)
	0.45	8.66	0.85	1.00	16	0.295	2.119	7.455	
3-methylpentane	0.18	8.41	0.69	0.79	27	0.277	1.217	2.199	Day and Felsing (1951)
	0.28	8.41	0.84	0.99	16	0.277	1.860	12.211	
2,2-Dimethylbutane	0.33	9.78	0.76	0.76	11	0.266	3.504	4.336	Felsing and Watson (1943b)
	0.33	9.78	0.81	0.97	21	0.266	2.887	6.102	
<i>n</i> -heptane	1.88	7.51	0.51	0.78	15	0.359	2.127	3.103	Dolittle (1963)
	1.88	7.51	0.88	0.97	4	0.359	1.858	3.877	
<i>n</i> -octane	0.20	8.12	0.66	0.79	24	0.395	0.893	1.935	Felsing and Watson (1942)
	0.41	8.12	0.83	0.96	18	0.395	1.937	5.151	
2,2,4-Trimethylpentane	0.02	9.74	0.51	0.63	50	0.395	1.419	3.457	Benson and Winnick (1971)
	0.20	9.84	0.69	0.78	25	0.310	2.485	3.797	
2,2,3,3-Tetramethylbutane	0.39	9.84	0.82	0.96	14	0.310	2.296	5.065	Felsing et al. (1947)
	0.41	8.16	0.70	0.80	24	0.377	1.681	5.105	
Cyclohexane	0.41	6.12	0.87	1.00	10	0.377	3.893	10.042	Reamer and Sage (1957)
	0.09	8.98	0.56	0.74	23	0.203	5.696	6.652	
Propene	0.36	9.10	0.80	0.92	15	0.203	5.377	7.521	Sage and Lacey (1955)
	0.30	8.99	0.76	0.76	13	0.143	1.371	1.617	
1-Butene	0.60	8.99	0.85	0.94	12	0.143	1.533	3.264	Sage and Lacey (1955)
	0.34	8.57	0.74	0.74	12	0.203	0.210	0.355	
1-Pentene	0.34	8.57	0.82	0.98	16	0.203	1.366	5.507	Day and Felsing (1951)
	0.14	7.82	0.75	0.79	20	0.218	9.828	10.238	
Argon	0.31	6.25	0.60	0.80	16	0.0	0.653	1.481	Michel et al. (1958), Din (1962a)
	0.63	6.25	0.86	1.00	14	0.0	2.824	7.124	
Nitrogen	0.22	5.84	0.60	0.72	14	0.0	0.733	1.683	Van Itterbeek et al. (1963)
	0.15	8.94	0.71	0.79	17	0.040	0.705	2.693	
Hydrogen sulfide	0.45	8.94	0.87	0.99	18	0.040	4.196	10.299	Sage and Lacey (1955)
	0.15	7.65	0.74	0.74	17	0.100	1.146	1.938	
Carbon dioxide	0.31	7.65	0.83	0.92	15	0.100	1.446	2.882	Jenkin (1920)
	0.15	1.08	0.78	0.80	13	0.225	2.813	3.179	
Propane	0.26	1.26	0.83	1.00	12	0.225	3.093	7.444	Din (1962a)
	0.02	1.43	0.62	0.68	11	0.152	1.279	2.043	
<i>n</i> -nonane	0.60	9.03	0.52	0.69	41	0.444	1.115	2.291	Sage and Lacey (1955)
Oxygen	0.04	2.88	0.48	0.58	23	0.017	0.323	0.848	Van Itterbeek et al (1960)
Krypton	0.01	0.09	0.55	0.69	7	0.0	1.104	1.324	Gladun and Menzel (1970)
<i>n</i> -pentane	0.20	8.17	0.66	0.70	14	0.252	0.166	0.274	Lee and Ellington (1965a)
<i>n</i> -decane	0.66	9.86	0.50	0.66	32	0.487	1.081	2.565	Lee and Ellington (1965b)
Overall	0.01	9.86	0.48	1.00	867		1.934	12.211	

overall average deviation of 5.2% and maximum deviation of 19% including the critical region.

CONCLUSIONS

In this investigation, Pitzer's correlation for compressed liquids is presented in analytical forms. This makes the computer application of this correlation much simpler.

Two sets of generalized equations of state representing $Z^{(0)}$ and $Z^{(1)}$ have been formulated. The first set of equations, Equation (6) and Equation (7), are derived for $Z^{(0)}$ and $Z^{(1)}$ separately over the region $0.5 \leq T_r \leq 0.99$ and $0 \leq P_r \leq 9.0$ using the tabulation of Pitzer et al. (1955) and Lu et al. (1973). The overall average deviation from literature data is 2.4%. The second set of equations, Equation (9) and Equation (10) extending the correlation to $P_r = 10.0$ are derived, using 867 litera-

ture volumetric data points of 27 different substances. They reproduce the volumetric data with overall average deviation of 1.9% and maximum deviation of 10% excluding a small region near critical point where deviations of 12% are observed. Both sets of equations compare favorably with the Lydersen correlation and the analytical equations as proposed by Hsi and Lu (1974).

For substances whose acentric factors are not known, a single equation, Equation (11), is formulated using only T_r and P_r as correlating parameters. This equation predicts volumetric behavior with 5% overall average deviation.

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Effect of Vertical Alignment on the Performance of Bubble and Foam Fractionation Columns

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In the course of recent research on bubble and foam fractionation (Valdes-Krieg et al., 1973, 1975), we have observed that the flow conditions and separations obtained in a countercurrent bubble fractionation column are sensitive to slight departures from true vertical alignment of the column. This phenomenon was observed to occur in a number of runs in which an anionic surfactant and copper ions were removed from a high-ionic-strength solution, which simulated a three-fold concentrated seawater brine. Surfactant removals were especially affected by column misalignment. We conducted a few experiments which would measure the effect of a controlled tilt in the column and compare its performance under aligned and misaligned conditions.

EXPERIMENT

A 6.95-cm diameter plexiglas column with a total length of 190.5 cm (foam and liquid pool combined) was used to conduct the experiments. Foam collapse was accomplished by means of a rotating perforated sleeve at the top of the column. The bottom of the column was provided with a fused-silica diffuser plate through which prehumidified air was sparged. Liquid entered the column 164 cm above the sparger plate through perforations in an annular distributor fed through a two-line manifold. The interface between aerated liquid (bubble regime) and foam was held 10 cm above the feed level. A similar device was used to withdraw the liquid effluent at the sparger level. The use of these distributors minimized liquid maldistribution at the inlet and outlet points, as well as bubble entrainment in the effluent. Axial liquid-phase concentration