A Four-Parameter Corresponding States Theory: Saturated Liquid Densities of Anormal Fluids

A fourth parameter to be used in conjunction with Pitzer's acentric factor is proposed to correlate the thermophysical properties of anormal fluids. It is based on the saturated liquid density at a reduced temperature of 0.7. A correlation for saturated liquid densities of anormal fluids is presented to cover a reduced temperature range from 0.42 to 1.0. For four hundred and fifty-six data points, covering twenty-one compounds, calculated average and average absolute deviation in predicted saturated liquid densities are −0.05 and 1.11%, respectively.

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SCOPE

Corresponding states theory plays a vital role in estimating thermophysical properties of polar as well as nonpolar substances. The three-parameter corresponding states theory presented by Pitzer has been highly successful in applications to normal fluids. For other fluids, termed here as anormal fluids, generalized correlations for saturated liquid density based on a fourth parameter are difficult to use if accurate vapor pressure data are not available.

The objective of this study is to present a new development of a fourth parameter which can be evaluated easily and accurately.

CONCLUSIONS AND SIGNIFICANCE

A new fourth parameter ψ, based on a saturated liquid density value, is defined which accounts for deviations in the behavior of anormal substances from those of normal substances in the sense of Pitzer's acentric factor.

The saturated liquid density correlation covering a range of reduced temperature from 0.42 to 1.0 is presented. In its application to twenty one anormal substances, covering four hundred and fifty six data points, average and average absolute deviation of −0.05 and 1.11%, respectively, were observed. The correlation is simple to use either for computerization or desk calculations since it is linear in the acentric factor as well as the new fourth parameter.

A universal characteristic of all fluids, normal or anormal, has been found to be that they exhibit a minimum in logarithm of the dimensionless volume U at a reduced temperature of 0.82. Physical significance of this characteristic is unclear.

A preliminary relationship between the new fourth parameter and that of Halm and Stiel is presented which allows prediction of vapor pressures of anormal fluids.

The corresponding states theory has been invaluable in estimating thermophysical properties of substances. Its evolution from two parameters, as suggested by Van der Waals (1873), to third and fourth parameters is described thoroughly by Reid and Sherwood (1966) and Leland and Chappelear (1968). The most widely used third parameters are acentric factor ω, as proposed by Pitzer et al. (1955), and the critical compressibility factor Z_c, as developed by Lydersen et al. (1955). Pitzer's acentric factor has been very successful in accurately correlating thermodynamic properties of nonpolar and slightly polar compounds. These fluids were characterized by Pitzer as normal fluids. This classification is followed here with the extension that all other fluids are anormal fluids which do not follow Pitzer's corresponding states theory. Thus, class of anormal fluids will primarily consist of polar compounds except a few which follow Pitzer's corresponding states theory.

Generalized correlations based on the critical compressibility factor cover all fluids, with a slight loss of accuracy for nonpolar compounds as concluded by Hougen et al. (1959) and Halm and Stiel (1970).

To extend the acentric factor corresponding states theory approach to anormal fluids, Pitzer (1955) had suggested a need of a fourth parameter. Accordingly, Halm and Stiel (1967, 1970) have presented a formulation of the fourth parameter χ and its application in correlating accurately several thermodynamic properties. The parameter χ is evaluated from experimental vapor pressure data at a reduced temperature of 0.6. For several anormal fluids, accurate vapor pressure data up to boiling point are given by Boublik (1973) and Lange (1967). Usually,

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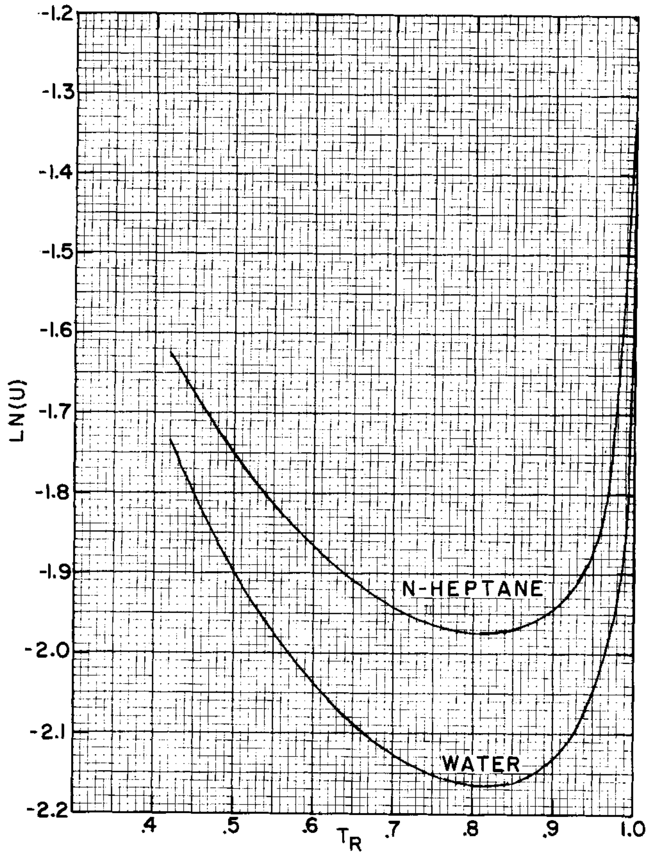

Fig. 1. Plot of ln(*U*) vs. T_r for *n*-heptane and water.

this range of vapor pressure data allows accurate evaluation of both ω and χ. But, if vapor pressure data are scattered, which is often the case at low reduced temperatures, evaluation of χ becomes difficult.

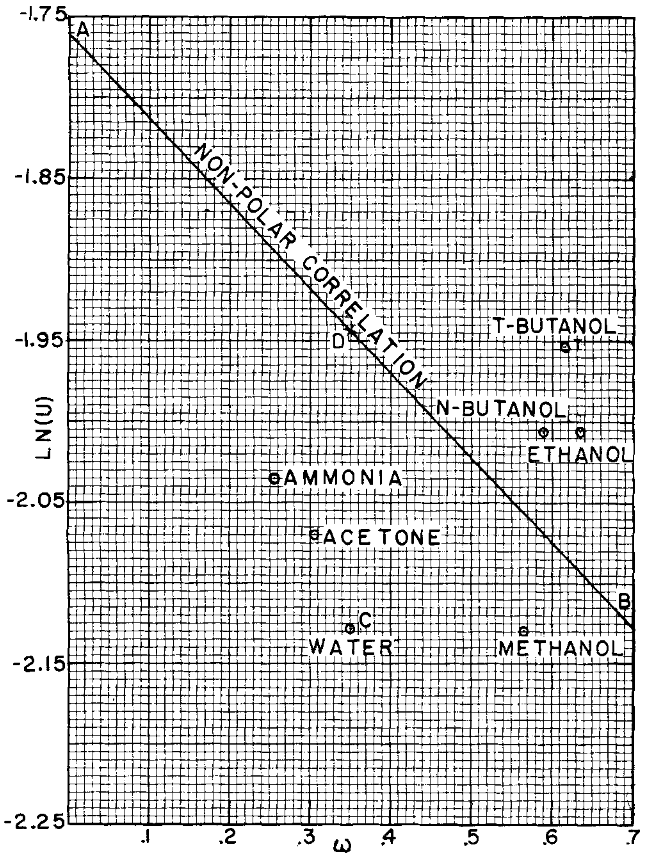

Fig. 2. Plot of ln(*U*) vs. ω at T_r = 0.7.

Table 1. Generalized Functions in Equation (4)

TR	$(\ln U_p)^0$	$(\ln U_{np})^1$	$(\ln U_p)^2$
0.420	−1.3807	−0.5753	1.1722
0.440	−1.4211	−0.5700	1.1526
0.460	−1.4604	−0.5630	1.1325
0.480	−1.4983	−0.5555	1.1130
0.500	−1.5343	−0.5485	1.0949
0.520	−1.5682	−0.5425	1.0784
0.540	−1.5997	−0.5376	1.0638
0.560	−1.6286	−0.5340	1.0512
0.580	−1.6549	−0.5315	1.0404
0.600	−1.6785	−0.5298	1.0313
0.620	−1.6995	−0.5288	1.0238
0.640	−1.7181	−0.5280	1.0177
0.660	−1.7343	−0.5273	1.0128
0.680	−1.7483	−0.5265	1.0090
0.700	−1.7605	−0.5254	1.0061
0.720	−1.7709	−0.5240	1.0040
0.740	−1.7797	−0.5222	1.0024
0.760	−1.7870	−0.5201	1.0014
0.780	−1.7928	−0.5179	1.0008
0.800	−1.7968	−0.5156	1.0005
0.820	−1.7988	−0.5133	1.0000
0.840	−1.7981	−0.5112	1.0000
0.860	−1.7940	−0.5090	1.0000
0.880	−1.7852	−0.5067	1.0000
0.900	−1.7701	−0.5036	1.0000
0.920	−1.7469	−0.4991	1.0000
0.940	−1.7130	−0.4920	1.0000
0.960	−1.6654	−0.4805	1.0000
0.980	−1.6004	−0.4626	1.0000
0.982	−1.5940	−0.4590	0.9990
0.984	−1.5840	−0.4510	0.9970
0.986	−1.5730	−0.4410	0.9950
0.988	−1.5620	−0.4280	0.9930
0.990	−1.5470	−0.4120	0.9870
0.992	−1.5310	−0.3920	0.9800
0.994	−1.5020	−0.3670	0.9700
0.996	−1.4570	−0.3370	0.9550
0.998	−1.3500	−0.3020	0.9350
0.999	−1.3000	−0.2830	0.9050
1.000	−1.2480	−0.2629	0.7000

Accurate saturated liquid density data are usually available. Therefore, in the alternate approach presented here, a new fourth parameter is defined, based on liquid density data. The generalized saturated liquid density correlation for anormal fluids presented here is easy to use since it is linear in both the acentric factor and the new fourth parameter ψ.

CORRELATION

An acentric factor correlation of saturated liquid densities of normal fluids has been presented by Bhirud (1978),[*] which has the following form:

$$\ln U_{np} = (\ln U_{np})^0 + \omega(\ln U_{np})^1 \qquad (1)$$

and

$$U_{np} = \left(\frac{P_c}{\rho_{sl}RT}\right)_{np} \qquad (2)$$

The difference between a normal fluid and an anormal fluid can be seen in the case of *n*-heptane and water which have just about the same acentric factor value. For these compounds, the quantity ln(*U*) is plotted vs. T_r in Figure 1. Note that, the curve for water lies consistently below that for *n*-heptane throughout the entire temperature range. Therefore, in principle, the deviation in ln(*U*) of water from *n*-heptane at any temperature can be used in defining a fourth parameter. To be consistent with Pitzer's choice of temperature, a fourth parameter ψ is de-

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[*] The paper referred to above is scheduled for publication in the *AIChE J.*, 24, No. 6 (1978).

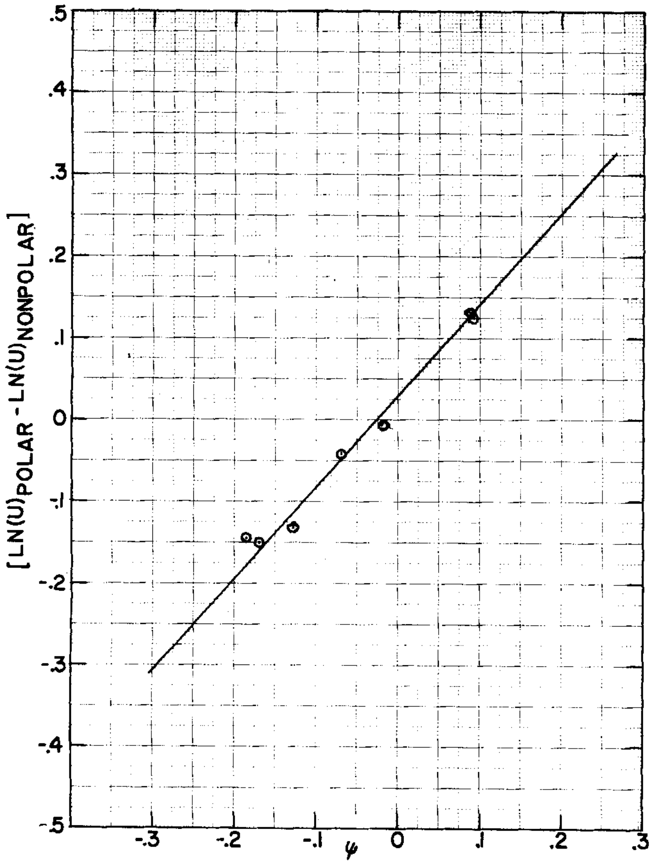

Fig. 3. Plot of $[\ln(U)_{\text{polar}} - \ln(U)_{\text{nonpolar}}]$ vs. ψ at $T_r = 0.5$.

fined at the reduced temperature of 0.7 as follows:

$$\psi = \{\ln Up - \ln U_{np}\}_{T_r=0.7} \qquad (3)$$

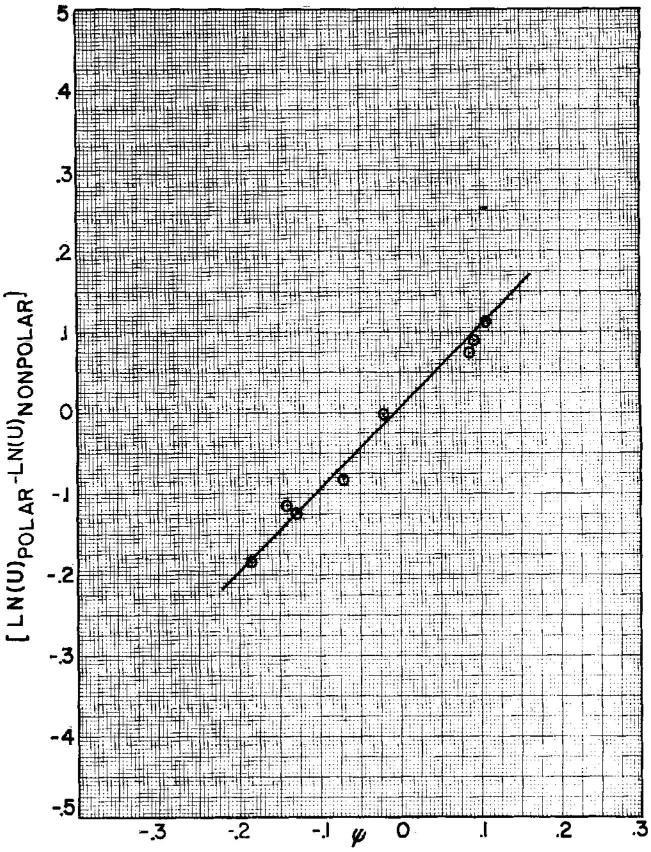

Fig. 4. Plot of $[\ln(U)_{\text{polar}} - \ln(U)_{\text{nonpolar}}]$ vs. ψ at $T_r = 0.9$.

Evaluation of the fourth parameter ψ for a few polar compounds used in developing this correlation can be seen in Figure 2, where $\ln(U)$ is plotted vs. acentric factor

Table 2. Summary of Deviations of Calculated Saturated Liquid Densities from

Compound	T_c, k	P_c, kPa	ω	ψ	T_r range	No. of points	Percent deviation*			
							Nonpolar		Polar	
							Avg.	Av. abs.	Avg.	Av. abs.
Water†	647.30	22 118.4	0.3480	−0.1845	0.42/1.00	42	−15.17	15.17	0.87	1.48
Ammonia†	405.60	11 297.8	0.2550	−0.1395	0.49/1.00	12	−12.12	12.12	0.32	1.16
Methanol†	512.58	8 099.3	0.5656	−0.0701	0.40/1.00	32	−5.63	5.63	0.16	1.11
Ethanol†	516.25	6 383.5	0.6341	0.0873	0.53/1.00	26	8.47	8.47	−1.13	1.56
N-propanol	536.71	5 169.7	0.6260	0.0929	0.47/1.00	26	9.79	9.79	−0.75	1.06
I-propanol	508.31	4 764.3	0.6630	0.0872	0.50/1.00	31	9.71	9.71	−0.18	0.90
N-butanol†	562.93	4 412.7	0.5893	0.0900	0.45/1.00	29	10.76	10.76	0.06	0.91
T-butanol†	506.15	3 971.9	0.6137	0.1066	0.60/1.00	19	11.16	11.16	−0.26	0.45
Acetone†	508.15	4 701.5	0.3057	−0.1265	0.41/1.00	49	−11.54	11.54	−0.36	1.00
Sulfur dioxide†	430.80	7 887.6	0.2460	−0.0181	0.46/1.00	25	−1.21	1.39	−0.25	1.06
Chlorine	417.00	7 701.4	0.0708	−0.0390	0.56/1.00	19	−3.61	3.61	−0.21	0.65
Carbon monoxide	132.92	3 499.1	0.0930	0.047	0.51/1.00	13	5.09	5.09	0.37	0.89
Carbonyl sulfide	375.00	5 874.3	0.0855	−0.1000	0.46/1.00	29	−7.77	7.83	1.04	1.63
Acetonitrile	548.00	4 833.2	0.3200	−0.4500	0.41/1.00	23	−35.72	35.72	0.55	2.26
Acetic acid	594.45	5 784.7	0.4340	−0.2115	0.49/0.96	15	−19.15	19.15	−0.40	0.53
Naphthalene	748.40	4 050.7	0.3024	−0.0430	0.66/0.79	12	−4.49	4.71	−0.37	1.43
Biphenyl	789.00	3 847.3	0.3643	0.1030	0.47/0.96	17	10.98	10.98	−0.42	1.13
Methyl chloride	416.25	6 679.6	0.1520	−0.0520	0.56/0.84	13	−4.99	4.99	−0.21	0.39
Ethyl chloride	460.40	5 267.6	0.1880	−0.0430	0.53/0.71	13	−4.01	4.01	−0.45	0.76
Dimethylformamide	647.20	4 423.0	0.3273	−0.2950	0.42/0.64	11	−25.37	25.37	0.37	0.39

† Compounds used in developing this correlation.

* Percent deviation = (Calculated density − Experimental density) × 100./Experimental density.

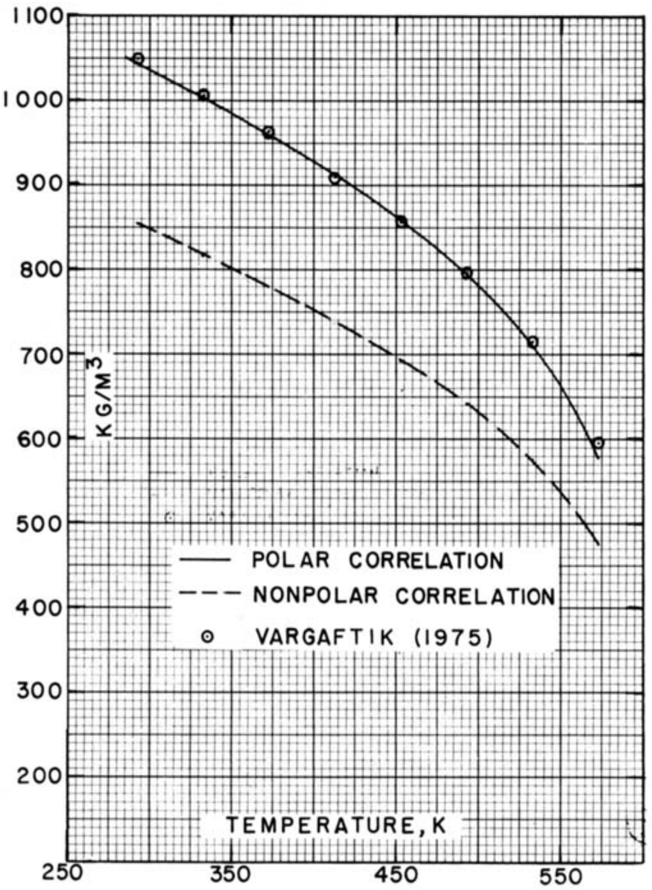

Fig. 5. Saturated liquid densities of acetic acid.

at the reduced temperature of 0.7, and line AB represents the nonpolar correlation as given by the Equation (1). The distance CD denotes a value of ψ of -0.1845 for water.

Literature Data

Max.	T_c Max.	Data source
−6.53	0.998	Electric Research Assoc. (1967)
3.96	0.986	Din (1962)
5.55	0.982	Eubank (1969), Zubarev et al. (1974)
−6.16	0.994	Wilhoit and Zwolinski (1973), Timmermans (1950, 1965)
−6.04	0.993	Vargaftik (1975)
−3.25	1.000	Timmermans (1950, 1965), Wilhoit and Zwolinski (1973)
−3.54	0.762	Timmermans (1950, 1965), Wilhoit and Zwolinski (1973), Kay and Donham (1955)
−1.53	1.000	Timmermans (1950, 1965), Wilhoit and Zwolinski (1973)
−4.31	1.000	Timmermans (1950, 1965), Lange (1969), Campbell and Chatterjee (1970),
−4.84	0.464	Perry et al. (1963)
−1.68	0.560	Perry et al. (1963)
−3.11	0.513	Vargaftik (1975)
5.41	0.956	Timmermans (1950, 1965)
5.83	0.989	Timmermans (1950, 1965)
−2.71	0.964	Vargaftik (1975)
−2.55	0.793	Vargaftik (1975)
−4.96	0.473	Vargaftik (1975), Perry et al. (1963)
−1.51	0.560	Perry et al. (1963)
−2.01	0.528	Perry et al. (1963)
0.90	0.640	DuPont (1963)

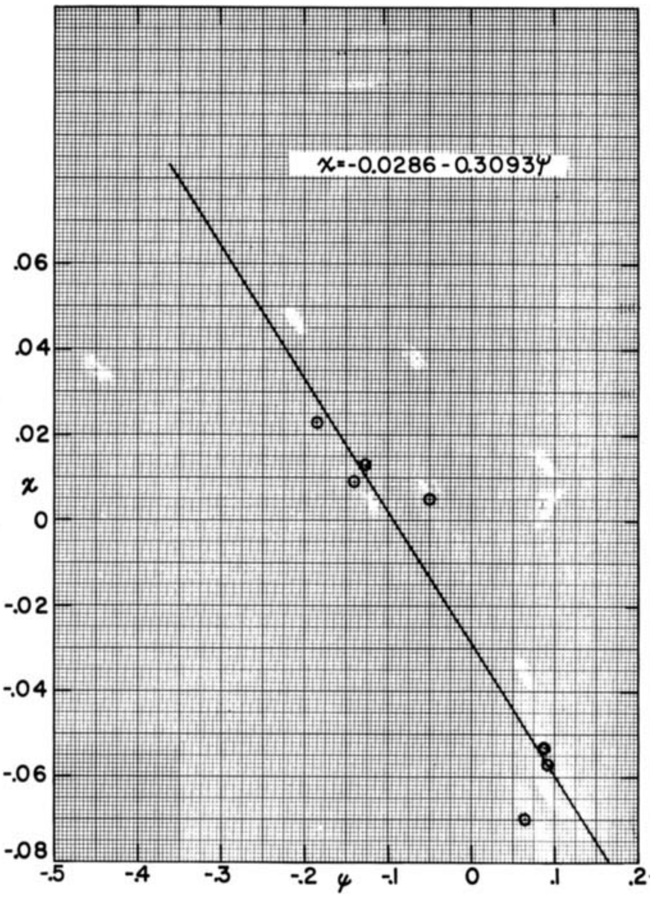

Fig. 6. Relationship between ψ and χ.

At other reduced temperatures, the deviations of the anormal fluids from normal fluid behavior were plotted vs. ψ. Two such plots at reduced temperatures of 0.5 and 0.9 are given in Figures 3 and 4, respectively. These figures show a straight line dependency on ψ, with a small but significant intercept at the origin. This intercept being nonzero indicates that properties of the basis compound (which has an acentric factor value of zero) such as argon must be corrected slightly at those reduced temperatures in order to form a basis polar compound. Thus, the polar correlation for saturated liquid densities takes on the following form:

$$\ln U_p = (\ln U_p)^0 + \omega(\ln U_{np})^1 + \psi(\ln U_p)^2 \quad (4)$$

and

$$(\ln U_p)^0) = (\ln U_{np})^0 + \delta^0 \quad (5)$$

Generalized functions in Equation (4) are presented over a reduced temperature range of 0.42 to 1.0 in Table 1. Up to and including the reduced temperature of 0.98, these functions are described by the following equations:

$$\begin{aligned}(\ln U_p)^0 = {} & -0.40062 - 8.0006\, T_r + 49.3780\, T_r^2 \\ & - 170.6616\, T_r^3 + 287.6989\, T_r^4 \\ & - 232.5608\, T_r^5 + 73.03299\, T_r^6 \end{aligned} \quad (6)$$

$$\begin{aligned}(\ln U_{np})^1 = {} & 13.4412 - 135.74367\, T_r + 533.38\, T_r^2 \\ & - 1091.453\, T_r^3 + 1231.43\, T_r^4 \\ & - 728.22\, T_r^5 + 176.737\, T_r^6 \end{aligned} \quad (7)$$

$$\begin{aligned}(\ln U_p)^2 = {} & 3.30929/T_r - 4.11794/T_r^2 + 2.37746/T_r^3 \\ & - 0.628621/T_r^4 + 0.062072/T_r^5 \quad 0.82 < T_r \\ & = 1.0 \qquad 0.82 < T_r < 0.98 \end{aligned} \quad (8)$$

Beyond T_r of 0.98, linear interpolation between the tabulated values is used.

DISCUSSION

Literature values of saturated liquid densities of several organic and inorganic polar compounds were compared with those predicted by this correlation as well as the nonpolar correlation, and results are summarized in Table 2. Note that the three-parameter corresponding states theory gives large deviations for polar compounds which can only be corrected by utilization of the fourth parameter. For example, saturated liquid densities of acetic acid compared in Figure 5 were predicted with an average deviation of -19.2% by the three-parameter corresponding states theory compared to -0.5% for the four-parameter corresponding states theory.

For twenty one compounds in Table 3, with four hundred and fifty six data points, average and average absolute deviation in predicted saturated liquid densities were -0.05 and 1.00%, respectively.

In the development of a correlation for the saturated liquid densities of normal fluids as presented by Bhirud (1978), it was noted that the quantity $\ln(U)$ exhibited a minimum around reduced temperature of 0.82 for normal fluids. Exactly the same kind of behavior was observed in case of the polar compounds. Thus, this behavior seems to be universal for all fluids. Physical significance of the minimum is not clear so far.

The fourth parameter presented here and Stiel's fourth parameter are based on two different properties but employ the same technique, namely, the use of the deviation of anormal fluid behavior from the normal fluid behavior.

Therefore, a vapor pressure correlation based on this fourth parameter ψ can be obtained if a relation between ψ and Stiel's fourth parameter is available. For a few anormal fluids common to both correlations, the relationship between χ and ψ, as shown in Figure 6, is described by

$$\chi = -0.0286 - 0.3093\psi \qquad (9)$$

This relationship should be regarded as tentative, since data for only seven compounds were used in its development.

In the case of acetone, Equation (9) yields a value of $\chi = 0.0105$. Using this value of χ with the acentric factor of 0.3057 in Halm and Stiel's vapor pressure correlation resulted, for thirteen points of Ambrose et al. (1974) covering a reduced temperature range of 0.5 to 0.7, in average and average absolute deviation of -0.12 and 0.25%, respectively, with maximum deviation of 0.8% only.

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NOTATION

P = pressure
R = gas constant
T = temperature
U = dimensionless volume = $(P_c V_{sl})/RT$
Z = compressibility factor

Greek Letters

δ^0 = generalized function
ω = Pitzer's acentric factor
ρ = density
ψ = fourth parameter
χ = Stiel's fourth parameter

Subscripts

c = quantity at the critical point
p = polar function
np = nonpolar function
r = reduced condition
sl = saturated liquid

Superscripts

0, 1, 2 = various generalized functions

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A Model For Moving-Bed Coal Gasification Reactors

A steady state model of moving-bed coal gasification reactors has been developed. Model predictions are in agreement with published commercial plant data for Lurgi pressurized gasification reactors and a pilot plant slagging gasifier. The dependence of reactor performance on operating variables has been studied for Illinois and Wyoming coals. For a given coal, maximum efficiency is determined by the coal-to-oxygen feed ratio. The location of the maximum temperature, which defines the combustion zone, is an important operating variable. Efficient operation of the dry ash reactor cannot be carried out below a critical feed gas temperature because of insufficient gasification and excessive carbon loss in the ash.

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Moving-bed coal gasification reactors are countercurrent devices in which a coal bed moves downward by gravity flow through an upward flowing gas stream. Steam and oxygen (or air) are fed at the bottom to provide the reactants for the combustion and gasification reactions. The pressurized Lurgi gasifier is commercially proven, while other moving-bed gasifiers, including a potentially high throughput and high temperature reactor that produces a liquid slag, are under development.

The composition and temperature of the product gas and the amount of unburned carbon in the ash largely determine the thermal efficiency of the process. The maximum temperature in the reactor determines the physical state of the ash (dry powder, clinker, slag) and hence the operability of a given reactor configuration. Composition and temperature depend on the properties of the coal being processed and on such operating parameters as feed rates, feed temperature, and reactor pressure. The throughput is limited by the maximum flow rate of steam and oxygen that does not cause entrainment and excessive pressure drop. The performance characteristics of moving-bed gasifiers do not seem to have been studied previously for wide ranges of coal properties, reactor configurations, and operating variables.

We describe here a steady state simulation of a moving-bed gasifier. The model describes the complex physical and chemical processes taking place in the multiphase moving bed, using mass and energy balances and information about rates of chemical reaction and physical transport processes. The use of such a model, following validation with plant data, enables the exploration and prediction of feasible and economically attractive ranges of design and operation in terms of the properties of the coal to be gasified.

CONCLUSIONS AND SIGNIFICANCE

The model has been compared to available commercial plant data for Lurgi reactors and to pilot plant experiments for a slagging reactor. Agreement with experiment is good, and the model has been used to predict the performance of moving-bed reactors for gasification of low activity Illinois and high activity Wyoming coals.

The maximum temperature in the bed is determined largely by the steam-to-oxygen ratio in the feed, while the efficiency at a given steam-to-oxygen ratio is determined by the carbon-to-oxygen feed ratio. In a dry ash reactor, the maximum temperature is limited by the ash fusion temperature, which requires a minimum steam-to-oxygen ratio of approximately seven. In a slagging reactor, the maximum temperature may be much higher, and the steam-to-oxygen ratio may be approximately one. The location of the maximum temperature, which defines the combustion zone, is an important operating variable. If the combustion zone is too low in the Lurgi reactor, then there will be insufficient combustion and an excess of unreacted carbon in the ash. If the zone is too high, there may be too much combustion coupled with insufficient

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