INTERMOLECULAR INTERACTIONS AND P-V-T BEHAVIOUR OF GASES

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ABSTRACT

Intermolecular interactions represented by refractive index-density and dielectric constant-density functions are examined for their applicability in the prediction of PVT behaviour of gases. Molar polarization at the critical temperature arrived through such considerations is shown to be a property characterizing the behaviour of all classes of substances.

Key words: Intermolecular interactions; Molar polarization; Theorem of corresponding states; Compressibility factors; PVT behaviour of fluids.

1. INTRODUCTION

Investigation such as those of Ashton and Guggenheim [1] and Orcutt and Cole [2] have shown that the following relationships are valid for real gases within the accuracy of experimental measurements:

$$\frac{n^2 - 1}{n^2 + 2} V = P^0 \left(1 + \frac{B'}{V} + \frac{C'}{V^2} + \ldots\right) \tag{1}$$

$$\frac{\epsilon - 1}{\epsilon + 2} V = E \left(1 + \frac{B^*}{V} + \frac{C^*}{V^2} + \ldots\right) \tag{2}$$

Comparison of Equations (1) and (2) with the virial Equation of state:

$$\frac{PV}{T} = R\left(1 + \frac{B}{V} + \frac{C}{V^2} + \ldots\right) \tag{3}$$

shows that the coefficients of any of them can be calculated from the other using equations of the following type obtained by slight rearrangement of Equations (1) to (3).

$$\frac{RT}{P} = \frac{P^0 (n^2 + 2)}{(n^2 - 1)} = \frac{E (\epsilon + 2)}{(\epsilon - 1)} \tag{4}$$

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$$\frac{BRT}{P} = \frac{B'P^0(n^2+2)}{(n^2-1)} = \frac{B^*E(\epsilon+2)}{(\epsilon-1)} \tag{5}$$

$$\frac{CRT}{P} = \frac{C'P^0(n^2+2)}{(n^2-1)} = \frac{C^*E(\epsilon+2)}{(\epsilon-1)} \tag{6}$$

and so on.

Before attempting an equation of state employing the set of relations given by Equations (4) to (6), making use of all accurate data (of all the properties concerned), it was thought fit to consider whether the information can be used to arrive at a simpler method of engineering applicability. With this in view, the possibility of proposing an improved form of the theorem of corresponding state based on Equations (1) to (3) is investigated.

2. THEOREM OF CORRESPONDING STATES

Several [3-6] critical reviews, including some by the authors of the present work [5, 6], are available on the theorem of corresponding states and its applicability. These reviews show the inadequacy of the two parameter charts in representing the PVT behaviour of real gases and indicate that more parameters are necessary to characterize all classes of substances. A three parameter theorem of corresponding states is a compromise between simplification and practical utility. Among the third parameters the extensively used ones are: acentric factor ω proposed by Pitzer [7], Riedel's [8] parameter Y_c, and critical compressibility factor Z_c of Lydersen [9]. These parameters are described in detail in Reid and Sherwood [10]. Of these three third parameters defined to account for the non-ideality of substances, two involve pressure and temperature but not volume. Lydersen's parameter takes into account pressure, volume and temperature at the critical point. However, none of these parameters accounts for the other properties of molecules such as polarity, structure, etc. It was therefore thought that a better representation of PVT can be obtained by choosing a parameter which not only involves P and T in some form but also accounts for volume, polarity and structure.

The analogy of Equations (1) and (2) with Equation (3) prompts one to investigate whether the LHS of Equations (1) and (2) can be used as a third parameter. By definition the LHS of (*a*) Equation (1) is molar refraction and (*b*) Equation (2) is molar polarization. For non-polar substances molar refraction and molar polarization are identical. Therefore Equations (1) and (2) can be considered to be representing molar polarization for non-polar and polar substances respectively. The value of molar polarization can be obtained from the measurements of refractive index or dielectric constant as indicated by Equations (1) and (2). Values of this property are not available at the critical temperature for many substances. However, this is not a disadvantage in our case as we can readily make use of the relation giving its temperature dependence:

$$P_M = R_M + \frac{4\pi N\mu^2}{9kT} \tag{7}$$

The little data available in literature shows that neither R_M nor μ is a strong function of temperature. Therefore, P_M can be calculatted from R_M and μ values available in literature without regard to their temperature dependence. The fact that all the third parameters proposed earlier are connected by simple relations makes one believe that there is no additional advantage in choosing any temperature other than critical temperature in defining the third parameter. So the third parameter chosen in the present work is molar polarization at the critical temperature defined as:

$$P_{MC} = R_M + \frac{4\pi N\mu^2}{9kT_C} \tag{8}$$

The parameters in Equation (8) for different substances are of such magnitude that they can be employed in the correlation without making any effort to reduce their numerical values. The values of these parameters can be obtained from data tabulated in standard references. This parameter satisfies the critieria of what one might call a good third parameter because:

(i) It involves the properties, molar refraction (R_M) and dipole moment (μ) which are determined easily experimentally. In the absence of data molar refraction can be calculated to good accuracy from atomic refractions.

(ii) Dipole moment of the substance (present in the definition of the parameter) accounts for the electrical characteristics of the molecules while the contribution from the constituent atoms of the molecules is taken into account by molar refraction. Some consideration to the structure of the molecules also is given as for example the same value is not taken for atomic refraction for oxygen in carbonyl group, ether group and hydroxyl group. Thus, consideration is given to electrical nature, constitution and structure of the molecules.

(iii) The parameter varies considerably from substance to substance.

(iv) Critical volume has no direct bearing on the value of the parameter.

(v) No extrapolation other than the straight use of Equation (7) is involved in determining the parameter.

(vi) No PVT data of the substance are involved in the parameter except for T_C. T_C is either known or can be estimated with good accuracy.

3. COMPRESSIBILITY FACTOR CORRELATION

To test the validity of the third parameter proposed, generalized charts and tables have been constructed using experimental data of six substances —ammonia, ethylene, hydrogen, *n*-heptane, methane and sulphur dioxide. Some preliminary results of this work have been presented earlier [6]. Details are given in this paper. Compressibility factors of different substances under the same conditions of T_R and P_R, when plotted against P_{MC}, gave linear relations so that compressibility factors could be expressed as

$$Z = Z^{(0)} + P_{MC} Z^{(1)} \tag{9}$$

The functions $Z^{(0)}$ and $Z^{(1)}$, obtained in the range $T_R = 0 \cdot 5$–$4 \cdot 0$ and P_R upto $9 \cdot 0$, are plotted as a function of P_R and T_R in Figs. 1 and 2, and Tables I and III represent them. Polynomial expressions for $Z^{(0)}$ and $Z^{(1)}$ are given in Tables II and IV. The values of $Z^{(0)}$ and $Z^{(1)}$ are used to calculate the compressibility factors of 25 substances. The results of comparison of the calculated values with experimental data are summarized in Tables V and VI.

4. DISCUSSION

The first method of ascertaining which parameter is superior, is by comparison of the results obtained in using each parameter for conformity with the principle of theorem of corresponding states. This can be carried out by applying the various methods to substances which have nearly the same value of the parameter in each case. Ideally this comparison should show zero deviation from experimental data. Among those compared the parameter that gives minimum per cent average absolute deviation affords a better agreement with the principle of the theorem of corresponding states.

Nitrous oxide and carbon monoxide (with $P_{MC} = 7 \cdot 65$ and $7 \cdot 18$ respectively) can be considered as substances with the same P_{MC}. The per cent average absolute deviation in using the present method for all the points of these substances is $1 \cdot 0$. Hydrogen sulphide and ethane (with ω of $0 \cdot 100$ and $0 \cdot 105$ respectively) can be considered to be substances with nearly the same ω. The per cent average absolute deviation in using Pitzer's method for all the points of these substances is 2·0. Methane and nitrogen (with Z_C of 0·290 and 0·291 respectively) can be considered as substances with the

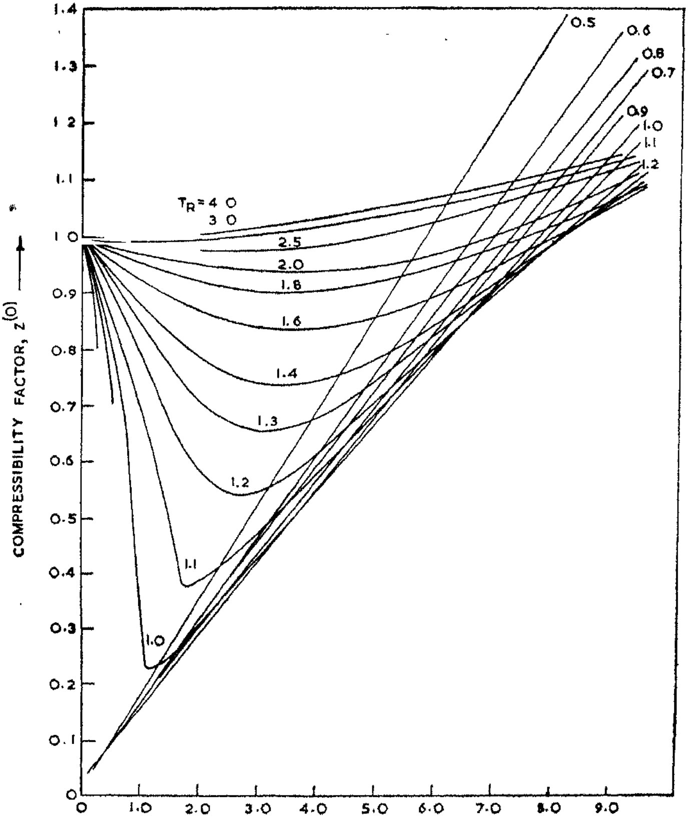

FIG. 1. $Z^{(0)}$ as a function of P_R and T_R.

TABLE I*

Compressibility factor $Z^{(0)}$ *in the relation* $Z = Z^{(0)} + P_{Rc}Z^{(1)}$

T_R \ P_R	0·75	1·25	1·50	3·00	5·00	7·00	8·00
0·60	0·1216	0·1949	0·2355	0·4729	0·7781	1·0782	1·2249
0·80	0·1267	0·2022	0·2383	0·4626	0·7488	1·0751	1·1560
0·90	0·1299	0·2005	0·2439	0·4588	0·7206	0·9697	1·0930
1·10	0·7787	0·5837	0·4724	0·4738	0·6750	0·9210	1·0250
1·30	0·8800	0·8037	0·7658	0·6533	0·7353	0·9147	0·9929
1·60	0·9402	0·9001	0·8910	0·8421	0·8587	0·9528	1·0090
1·80	0·9600	0·9414	0·9325	0·9036	0·9248	0·9801	1·0302
2·50	0·9782	0·9726	0·9734	0·9901	1·0335	1·0690	1·0969
3·00	0·9810	0·9729	0·9829	1·0015	1·0391	1·0792	1·1075

Tables I–VI are representative tables. Detailed tables can be obtanied from the methods

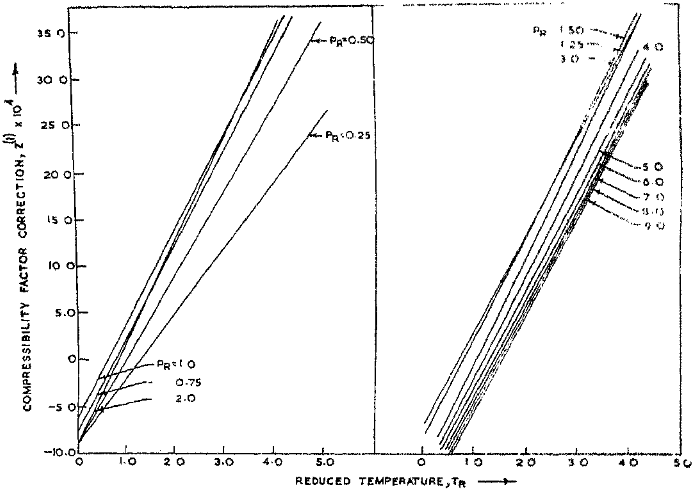

FIG. 2. $Z^{(1)}$ as a function of T_R and P_R.

TABLE II

Expressions for $Z^{(0)}$

$$Z^{(0)} = A + BT_R + CT_R^2 + DT_R^3 + ET_R^4$$

P_R	T_R range	A	B	C	D	E	% Av. Abs. Devn.. in Z^0
0·75	0·50–0·90	0·8290	−2·8682	0·3795	1·6347	0·0	0·21
	1·00–2·00	−9·4326	25·5027	−23·7337	9·8494	−1·5292	0·35
	2·00–4·00	0·8957	0·0491	−0·0068	0·0	0·0	0·11
1·25	0·50–1·00	0·6868	−1·5699	1·4318	−0·3078	0·0	2·71
	1·00–2·00	−21·1658	54·0986	−49·9862	20·5490	−3·1555	0·26
	2·00–4·00	0·8436	0·0719	−0·0087	0·0	0·0	0·19
1·50	0·50–1·00	1·3821	−4·4234	5·4939	−2·2020	0·0	1·68
	1·10–2·00	−24·3259	60·5678	−54·9041	22·1802	3·3556	0·28
	2·00–4·00	0·7835	0·1180	−0·0142	0·0	0·0	0·22
3·00	0·50–1·00	2·6185	−8·3414	10·5442	−4·3763	0·0	1·51
	1·10–2·00	8·2133	−24·0155	26·1006	−11·8286	−1·9393	0·85
	2·00–4·00	0·6155	0·1723	−0·0229	0·0	0·0	0·39
5·00	0·50–1·00	4·1860	−13·2975	16·9825	−7·1967	0·0	1·57
	1·10–2·00	2·0199	−3·9997	3·9632	−1·5442	0·2162	0·39
	2·00–4·00	0·6843	0·2062	−0·0284	0·0	0·0	0·61
7·00	0·50–1·00	5·5182	−17·3159	22·1539	−19·4255	0·0	2·51
	1·10–2·00	4·6807	−9·3553	8·6636	−3·4937	0·5273	0·21
	2·00–4·00	0·7756	0·1705	−0·0225	0·0	0·0	0·40
8·00	0·50–1·00	5·8852	−17·7891	22·2610	−9·3266	0·0	1·33
	1·10–2·00	5·2207	−10·1638	9·0231	−3·5207	0·5152	0·07
	2·00–4·00	0·7798	0·1936	−0·0276	0·0	0·0	0·30

TABLE III

Compressibility factor correction

$Z^{(1)} \times 10^4$ *of the relation* $Z = Z^{(0)} + P_{35} Z^{(1)}$

T_R \ P_R	0·75	1·25	1·50	3·00	5·00	7·00	8·00
0·60	— 1·75	— 0·80	— 1·55	— 4·50	— 6·60	— 8·05	— 8·60
0·80	+ 0·25	+ 1·20	+ 0·55	+ 2·30	— 4·60	— 6·05	— 6·60
0·90	+ 1·25	+ 2·20	+ 1·60	— 1·20	— 3·60	5·05	5·60
1·10	+ 3·25	+ 4·20	+ 3·70	+ 1·00	— 1·60	— 3·05	3·60
1·30	+ 5·25	+ 6·20	+ 5·80	+ 3·20	+ 0·40	— 1·05	— 1·60
1·60	+ 8·25	+ 9·20	+ 8·95	+ 6·50	+ 3·40	+ 1·95	+ 1·40
1·80	+10·25	+11·20	+11·05	+ 8·50	+ 5·40	+ 3·95	+ 3·40
2·50	+17·25	+18·20	+18·40	+16·40	+12·40	+10·95	+10·40
3·00	+22·25	+23·20	+23·65	+21·90	+17·40	+15·95	+15·40

TABLE IV

Expressions for $Z^{(1)}$

$Z^{(1)} = (A + BT_R) \times 10^{-4}$ T_R *range* = 0·50 *to* 4·00

P_R	A	B
0·75	— 7·75	10·00
1·25	— 6·80	10·00
1·50	— 7·85	10·50
3·00	—11·10	11·00
5·00	—12·60	10·00
7·00	—14·05	10·00
8·00	—14·60	10·00

same Z_C. The per cent average absolute deviation in using Lydersen's method for all the points of these substances is 1·6. Thus the present third parameter affords a better agreement with the principle of the theorem of corresponding states.

The validity of the proposed third parameter can be established by applying the correlation developed in the present work to classes of substances for which superiority is claimed in using the other parameters. Pitzer's method has been designed for normal fluids, so it is expected to give good results for normal fluids. Likewise, the critical compressibility factor method of Lydersen and coworkers should give good results for substances with Z_C nearly equal to 0·27 by virtue of its development on the basis of averaged data of substances, with $Z_C = 0{\cdot}27$. Viswanath's charts (11) are superior because of the better results he finds for substances such as hydrogen and ammonia.

Methane, ethane, nitrogen, hydrogen, hydrogen sulphide and carbon monoxide can be considered as normal fluids ($\mu^4 P_C{}^2/T_C \leqslant 0{\cdot}5 \times 10^{-6}$). It can be seen from Table V that the present method gives a per cent average absolute deviation of the same order as Pitzer's method for nitrogen and carbon monoxide and better results for all the other substances. On an overall basis for this group, the per cent average absolute deviation in using the present method is 1·0 against 1·2 for Pitzer's method. Ethylene, nitrous oxide, sulphur dioxide and hydrogen sulphide are substances with Z_C approximately equal to 0·27. It can be seen from Table V that the present method gives better results compared to Lydersen's method for these substances individually. Collectively, the present method gives, for this group a per cent average absolute deviation of 1·5 as against 2·2 for Lydersen's method. For hydrogen and ammonia the present method gives results better than Viswanath's method. Collectively for all the points of these substances, the present method gives a per cent average absolute deviation of 4·4 compared to 6·0 in using Viswanath's method.

It can be seen from Table VI that the present method gives better results for all the isotherms compared except for $T_R = 2{\cdot}0$ isotherm.

On an overall basis the present method gives a per cent average absolute deviation of 1·8 compared to 2·2 for Pitzer's method, 2·6 for Lydersen's method and 2·4 for Viswanath's method (two parameter charts constructed based on the use of Su's ideal critical volume).

TABLE V

Summary of comparison of calculated and experimental compressibility factors for each substance

Substance	Reduced temperature range	Reduced pressure range	Per cent average absolute deviation**				Source of experimental data
			Present method	Pitzer's method	Lydersen's method	Viswanath's method	
Acetone ..	0·60	0·50–9·00	6·87 (13)	*	8·73 (13)	*	*a*
Ammonia ..	0·80–1·40	0·25–8·00	6·51 (55)	8·02 (55)	9·26 (55)	7·25 (46)	*b*
n-Butane ..	1·00–1·30	0·25–7·00	0·86 (44)	1·24 (44)	1·01 (44)	0·85 (44)	*c*
1-Butene ..	0·80–1·00	0·75–9·00	6·94 (27)	5·42 (27)	5·33 (27)	3·30 (8)	*d*
i-Butyl alcohol ..	0·50–0·60	0·25–9·00	2·78 (28)	*	3·46 (28)	*	*a*
Carbon monoxide ..	1·60–3·00	0·25–6·00	0·95 (40)	0·96 (40)	1·69 (40)	1·33 (40)	*e*
Chlorodifluoromethane ..	0·90–1·30	0·25–0·50	2·32 (9)	1·97 (9)	1·56 (9)	1·81 (8)	*f*
n-Decane ..	0·50–0·60	0·50–9·00	4·78 (22)	*	6·65 (22)	*	*a*
Diethyl ether ..	0·70–0·80	0·25–9·00	11·14 (23)	9·10 (11)	15·75 (23)	*	*a*
2, 2-Dimethyl butane	0·60–1·00	0·25–9·00	9·86 (55)	4·95 (31)	12·04 (55)	2·17 (14)	*a, f*
Ethane ..	1·00–1·60	0·25–6·00	0·54 (44)	1·00 (44)	1·61 (44)	0·74 (44)	*c*
Ethyl bromide ..	0·60–0·70	0·25–9·00	21·02 (28)	*	30·16 (28)	*	*a*

Ethylene	1·00–1·60	0·25–4·00	0·93 (46)	1·17 (46)	1·88 (46)	1·10 (46)	*c*
n-Heptane	0·50–0·90	0·25–8·00	0·75 (41)	5·86 (11)	3·82(41)	*	*h*
n-Hexane	0·80–0·90	0·75–8·00	0·47 (12)	1·56 (12)	0·89 (12)	*	*i*
Hydrogen	1·00–4·00	0·25–8·00	3·88 (89)	4·28 (89)	5·29 (89)	4·93 (89)	*j*
Hydrogen sulphide	0·80–1·20	0·25–6·00	2·39 (36)	3·17 (36)	2·44 (36)	1·50(26)	*k*
Methane	1·10–2·00	0·25–9·00	0·72 (66)	0·86 (66)	1·96 (66)	1·31 (66)	*l*
Neopentane	1·00–1·20	1·00–6·00	2·84 (24)	2·25 (24)	2·92 (24)	2·22 (24)	*m*
Nitrogen	1·00–2·00	0·25–3·00	0·74 (36)	0·53 (36)	0·96 (36)	0·66 (36)	*n*
Nitrous oxide	1·00–1·40	0·25–4·00	1·15 (38)	1·99 (38)	1·75 (38)	1·21 (38)	*o*
n-Octane	0·50–0·60	0·25–9·00	4·87 (28)	*	5·10 (28)	*	*a*
Propane	1·00–1·40	0·25–4·00	0·62 (49)	1·25 (49)	1·53 (49)	0·82 (49)	*c*
Steam	1·00–1·80	0·25–1·50	0·69 (36)	1·46 (36)	1·26 (36)	1·29 (36)	*c*
Sulphur dioxide	1·00–1·20	0·25–4·00	2·13 (26)	3·19 (26)	2·42 (26)	3·09 (26)	*p*
Overall	..	..	3·59 (915)	2·85 (730)	4·97 (915)	2·36 (640)	..
For the same points (using the present method)	..	..	..	2·25	3·59	1·79	..
For 640 common points	..	..	1·79	2·23	2·56	2·36	

** Numbers in the parenthesis of these columns refer to the number of points compared.

* The charts do not extend to these T_R values.

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TABLE VI

Summary of comparison of calculated and experimental compressibility factors for each isotherm

Isotherm (reduced temperature)	No. of points tested	Per cent average absolute deviation			
		Present method	Pitzer's method	Lydersen's method	Viswanath's method
0·50	49	3·06	*	4·14	*
0·60	88	12·02	*	13·08	*
0·70	48	12·98	*	17·79	*
0·80	53	5·31	7·15	7·91	*
0·90	37	5·85	6·01	5·79	*
1·00	104	2·46	3·51	3·12	3·07
1·10	116	1·38	2·20	1·93	1·80
1·20	110	1·21	1·51	2·12	1·51
1·30	96	1·58	1·78	2·50	2·21
1·40	52	1·78	1·97	4·04	2·94
1·60	55	1·36	1·41	1·96	1·62
1·80	42	1·42	1·32	1·59	1·81
2·00	37	4·69	4·50	4·20	4·72
2·50	11	1·93	2·96	2·01	2·60
3·00	11	1·74	1·98	2·74	2·08
4·00	6	0·27	0·88	4·40	1·50

* The charts do not extend to these T_R values.

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NOMENCLATURE

$B, C, \ldots$	Virial coefficients of the equation of state, functions of temperature (Equation 3).
$B', C', \ldots$	Virial coefficients of the refractive index equation (Equation 1).
$B^*, C^*, \ldots$	Virial coefficients of dielectric constant equation (Equation 2).
E	Temperature independent constant of Equation 2.
k	Boltzmann constant.
n	Refractive index.
N	Avagadro's number.
P	Pressure.
P^0	Temperature independent constant of Equation 1.
P_C	Critical pressure.
P_R	Reduced pressure.
P_M	Molar polarization.
P_{MC}	Molar polarization at the critical temperature.
R	Universal gas constant.
R_M	Molar refraction.
T	Temperature.
T_C	Critical temperature.
T_R	Reduced temperature.
V	Molar volume.
Y_C	Riedel's parameter.
Z	Compressibility factor.
Z_C	Critical compressibility factor, Lydersen's third parameter.
$Z^{(0)}$	Main part of the compressibility factor correlation, Equation 9.
$Z^{(1)}$	Correction term of the compressibility factor correlation, Equation 9.
ϵ	Dielectric constant.
ω	Acentric factor, Pitzer's third parameter.
π	Constant $= 22/7$.
μ	Dipole moment.