

Correlation for the third virial coefficient for non-polar and polar compounds using a cubic equation of state

E.M. Beshler¹ and J. Lielmezs

*Department of Chemical Engineering, The University of British Columbia,
Vancouver, B.C. (Canada)*

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Abstract

A correlation for the prediction of the third virial coefficient of non-polar and polar compounds has been developed by using a cubic equation of state. It requires a knowledge of the critical pressure, the critical temperature and the Pitzer acentric factor ω , and, in addition for polar compounds, a knowledge of the dipole moment and of an empirical substance-dependent factor x . Predictions agree very well with the experimental data for all the compounds considered. The results also compare well with the values obtained by means of the De Santis and Grandes, and the Orbey and Vera correlations, which were established to predict the third virial coefficient of non-polar gases.

LIST OF SYMBOLS

$a(T)$	function defined by eqn. (4a)
b	defined by eqn. (4b)
B, C, D	second, third and fourth virial coefficients respectively
c	power series coefficient defined by eqns. (13a)–(13c)
F	function defined by eqns. (7), (8) and (12)
N	number of data points
P	pressure
R	universal gas constant
T	temperature
u	constant, eqn. (2)
V	volume
w	constant, eqn. (2)
x	substance-dependent constant, defined by eqns. (8) and (12)

Correspondence to: J. Lielmezs, Department of Chemical Engineering, The University of British Columbia, Vancouver, B.C., Canada.

¹ Present address: El-Fateh University, Tripoli, Libya.

Greek letters

α	function defined by eqns. (4a) and (12)
μ	dipole moment
ω	Pitzer acentric factor
Ω_a, Ω_b	dimensionless entities

Subscripts

c	critical state
r	reduced state

INTRODUCTION AND THEORETICAL DEVELOPMENT

In the analysis of vapour–liquid equilibrium behaviour, the non-ideality of the vapour phase should be taken into account. For calculations to moderate pressures, the density is often less than two-thirds the critical density, and the virial equation of state expanded to include the third term may supply a good estimate of the vapour-phase fugacity coefficient.

This work provides a correlation for non-polar and polar fluids, based on the available critically selected experimental values of the third virial coefficient.

The expanded form of the pressure-explicit virial equation of state may be written

$$P = \frac{RT}{V} \left(1 + \frac{B}{V} + \frac{C}{V^2} + \frac{D}{V^3} + \dots \right) \quad (1)$$

where B is the second, C the third, D the fourth, etc. virial coefficient.

There are several correlations for estimating the third virial coefficient of non-polar compounds: those of Chueh and Prausnitz [1], Pope et al. [2], De Santis and Grande [3] and Orbey and Vera [4]. This work, using the recently proposed F function of Shaw and Lielmezs [5] and Mak and Lielmezs [6] via the Peng–Robinson equation [7], presents a correlation for estimating the third virial coefficient for both non-polar and polar fluids.

We write the generalized form of the cubic equation of state of Schmidt and Wenzel [8] as

$$P = \frac{RT}{V-b} - \frac{a(T)}{V^2 + ubV + b^2} \quad (2)$$

If eqn. (2) is expanded in inverse molar volume, similarly to eqn. (1), the second and third virial coefficients are given as

$$B = b - \frac{a(T)}{RT} \quad (3a)$$

$$C = b(b + ua(T)/RT) \quad (3b)$$

where $a(T)$ and b are defined as

$$a(T) = a(T_c)\alpha(T) = \Omega_a \left(\frac{R^2 T_c^2}{P_c} \right) \alpha(T) \quad (4a)$$

$$b = b(T_c) = \Omega_b \frac{RT_c}{P_c} \quad (4b)$$

The proposed correlation for the third virial coefficient has been developed from eqn. (3b) by using the Peng–Robinson equation of state reproduced from eqn. (2) by setting $u = 2$ and $w = -1$, i.e.

$$P = \frac{RT}{V-b} - \frac{a(T)}{V^2 + 2bV - b^2} \quad (5)$$

The third virial coefficient (eqn. (3b)) in terms of the Peng–Robinson equation (eqn. (5)) can be now written as

$$C = b^2 + 2ba(T)/RT \quad (6)$$

If we put eqns. (4a) and (4b) into eqn. (6), and then multiply throughout by $(P_c/RT_c)^2$, we obtain in terms of the Peng–Robinson equation of state an expression for the third virial coefficient given as

$$C \left(\frac{P_c}{RT_c} \right)^2 = \Omega_b^2 + 2\Omega_a\Omega_b F \quad (7)$$

where $F = \alpha(T_r)/T_r$ is the F function, and Ω_a and Ω_b assume the values for the Peng–Robinson equation of state, i.e. $\Omega_a = 0.45724$ and $\Omega_b = 0.07780$. To account empirically for the non-additivity contributions to the third virial coefficient [1,3,4,9], we follow the approaches of Mak and Lielmezs [10] and Romero and Lielmezs [11] and express the total F function (eqn. (7)) in terms of three parameters: the reduced temperature T_r , the Pitzer acentric factor ω , and the reduced dipole moment μ_r raised to an empirical substance-dependent power x , i.e.

$$F = F(T_r, \omega, \mu_r^x) \quad (8)$$

where

$$\mu_r = \frac{10^5 \mu^2 P_c}{T_c^2} \quad (9)$$

Defining

$$C_r = C(P_c/RT_c)^2 \quad (10)$$

we can rewrite eqn. (7) as

$$C_r = \Omega_b^2 + 2\Omega_a\Omega_b F(T_r, \omega, \mu_r^x) \quad (11)$$

Following Romero and Lielmezs [11], the three-parameter division of the F function (eqn. (11)) designates the following contributions: first, the contribution of simple fluids, the T_r contribution; second, the correction for the deviation of normal fluids from simple fluids, the ω and T_r contributions; and third, the behaviour of polar fluids, the T_r and μ_r^x contributions. Hence the total F function contribution may be written as the sum of the above three separate terms:

$$F(T_r, \omega, \mu_r^x) = F^{(0)}(T_r) + \omega F^{(1)}(T_r) + \mu_r^x F^{(2)}(T_r) \quad (12)$$

It has been shown by Lielmezs and co-workers [5,6,10,11] that each of the three F subfunctions may be expanded as follows:

$$F^{(0)}(T_r) = \sum_{i=1}^1 c_{i0} T_r^{(1-i)} \quad (13a)$$

$$F^{(1)}(T_r) = \sum_{i=1}^m c_{i1} T_r^{(1-i)} \quad (13b)$$

$$F^{(2)}(T_r) = \sum_{i=1}^n c_{i2} T_r^{(1-i)} \quad (13c)$$

As noted above, eqn. (13a) represents the $F^{(0)}$ function of simple fluids. To determine the numerical values of the coefficients c_{i0} and the number of terms appearing in eqn. (12), the general criterion of minimum variance of the curve fit was used (Fig. 1a). Once an expression for the $F^{(0)}$ subfunction (eqn. (13a)) was obtained, the $F^{(1)}$ subfunction (coefficients c_{i1} , eqn. (13b)) was calculated from

$$F^{(1)} = \frac{F - F^{(0)}}{\omega} \quad (14)$$

where the F function in turn is calculated from eqn. (11). To obtain the number of terms appearing in eqns. (13), again the general criterion of minimum variance of the curve fit was used (Fig. 1b).

The numerical values of coefficients c_{i2} (eqn. (13c)) were determined by means of multiple linear regression methods, letting $x = 1$, i.e. setting first

$$F^{(2)}(T_r, \omega, \mu_r^{x=1}) = \frac{F_i - (F_i^{(0)} - \omega_i F_i^{(1)})}{\mu_{r_i}^{x=1}} \quad (15)$$

Then, using the c_{i2} coefficient value computed using eqn. (15), the optimum value of the exponent x (Table 1) was determined in each case for each compound in such a way as to minimize the root-mean-square-percentage (RMS%) error in the calculated value of the third virial coefficient. As a measure of x stability, the x range over which there is a 10% or less variation (increase) in RMS% error with respect to the optimum (least) x , is given in Table 1. The minimum number of terms appearing in eqn. (13c)

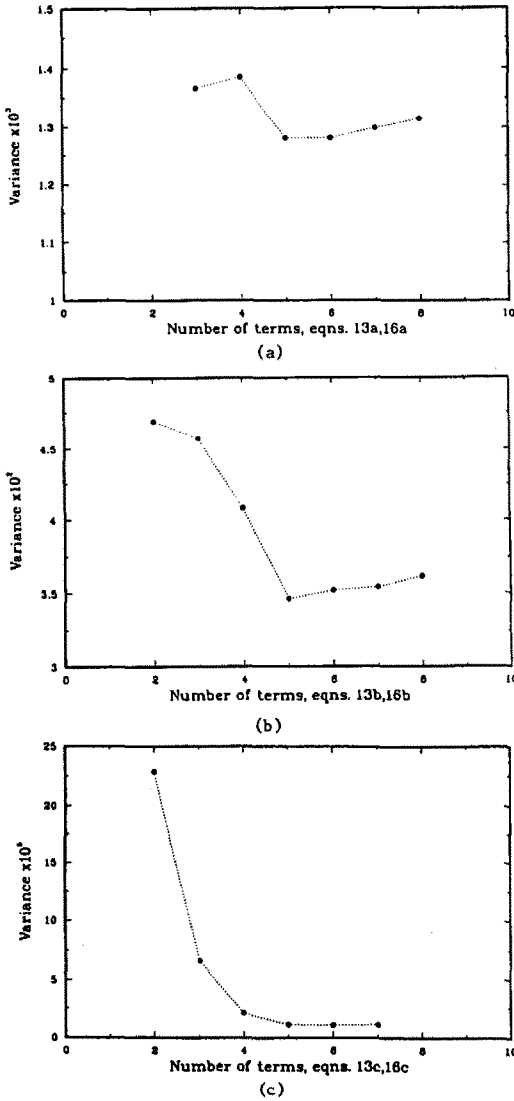


Fig. 1. Plots of the variance against the number of terms to determine the minimum number of terms in the equations: (a) eqns. (13a) and (16a); (b) eqns. (13b) and (16b); (c) eqns. (13c) and (16c).

was once again obtained from the general criterion of minimum variance of the curve fit of the $F^{(2)}$ subfunction (Fig. 1c).

The final proposed expressions in the expanded coefficient form for the $F^{(0)}$ (eqn. (13a)), $F^{(1)}$ (eqn. (13b)) and $F^{(2)}$ (eqn. (13c)) subfunctions are

$$\begin{aligned}
 F^{(0)}(T_r) = & -0.01175 - 0.80483/T_r + 7.29366/T_r^2 - 16.98304/T_r^3 \\
 & + 16.86138/T_r^4 - 5.94613/T_r^5
 \end{aligned}
 \tag{16a}$$

TABLE 1
Reduced third virial coefficient C_3 , absolute average deviation (AAD%)^a and RMS% error^b value comparison

Compound	ω^c	μ^c	N	ΔT_r	AAD% ^d			RMS% E ^d			x	Data sources ^e
					I	II	III	I	II	III		
Argon			10	1.01-1.43	14.87	16.68	9.57	22.47	10.91	14.17		Onnes and Crommelin (1910) [13]
			7	1.98-2.97	13.26	19.25	12.78	15.16	23.57	14.57		Tanner and Masson (1930) [14]
			12	0.88-1.65	3.31	2.23	3.59	3.97	2.86	4.99		Michels et al. (1958) [15]
Xenon			4	0.95-1.81	7.84	2.40	1.99	8.45	3.14	2.51		Crain and Sonntag (1966) [16]
			13	1.01-1.98	14.69	11.15	8.41	16.00	13.13	10.47		Beattie et al. (1951) [17]
Krypton			7	0.94-1.46	16.09	11.23	7.86	19.51	13.98	10.43		Michels et al. (1954) [18]
			9	1.30-4.17	16.71	28.50	15.28	22.13	37.58	17.93		Whalley and Scheider (1954) [19]
			7	1.30-2.02	13.07	8.39	5.20	13.11	9.48	6.67		Trappeniers et al. (1966) [20]
Average								15.10	14.33	10.22		
Benzene	0.212		5	0.88-1.02	4.03	2.86	3.94	4.80	3.67	4.19		Connolly and Kandalic (1960) [21]
	0.398		5	0.87-1.01	4.57	5.39	3.23	5.16	7.08	3.88		Connolly and Kandalic (1960) [21]
Sulphur hexa-fluoride	0.286		6	0.96-1.27	18.83	9.81	10.33	19.23	10.82	11.09		Clegg et al. (1955) [22]
	0.039		11	1.01-2.32	18.06	13.33	0.26	23.77	19.74	14.49		Onnes and Van Urk (1924) [23]
Nitrogen			6	1.06-2.17	11.08	4.74	2.44	11.75	5.73	2.79		Canfield et al. (1963) [24]
			6	1.06-2.17	6.69	2.44	4.60	7.60	2.92	5.76		Hoover et al. (1964) [25]
			4	1.13-2.17	9.37	2.61	1.95	10.63	2.80	3.36		Crain and Sonntag (1966) [16]
Ethane			13	1.24-2.31	7.70	1.52	4.19	9.04	1.84	4.92		Roe (1972) [26]
	0.099		7	1.02-1.67	10.73	3.27	3.26	11.27	4.63	4.05		David et al. (1952) [27]
			16	0.89-2.04	14.42	7.11	3.71	18.27	10.67	4.87		Doulin and Harrison (1973) [28]
		7	0.89-1.38	5.55	2.14	2.83	6.17	2.47	2.89		Michels et al. (1954) [29]	

Methane	0.011	16	1.43–3.27	12.00	18.23	5.86	12.06	20.68	7.05	Douslin et al. (1964) [30]
		3	1.57–1.83	21.07	14.51	11.11	23.94	18.14	15.18	Lee and Edmister (1970) [31]
		7	1.43–2.22	6.01	10.20	10.76	7.33	11.06	10.98	Michels and Nederbragt (1935) [32]
		7	1.43–2.22	2.70	5.01	7.02	2.87	5.82	7.45	Michels and Nederbragt (1936) [33]
		7	1.43–2.22	5.38	9.53	9.14	7.99	11.64	10.36	Schamp et al. (1958) [34]
		10	0.83–1.53	5.16	1.82	4.16	6.17	2.52	4.71	Roe (1972) [26]
		7	0.97–1.50	2.60	3.25	3.05	3.25	3.94	4.74	Bird et al. (1950) [35]
Ethene	0.089	12	0.97–1.50	10.34	10.67	9.70	24.11	21.90	20.31	Butcher and Dadson (1964) [36]
		8	1.0–1.23	8.99	12.12	11.11	10.72	12.84	11.82	Trappeniens et al. (1975) [37]
		21	0.84–1.59	4.89	6.10	3.76	6.16	6.91	4.40	Douslin and Harrison (1976) [38]
		12	0.86–1.04	15.83	6.65	8.13	15.88	7.57	8.84	Lee (1976) [39]
Average						11.28	8.27	7.64		
Carbon monoxide	0.066	0.1	2.06–3.18			25.02			25.13	Michels et al. (1952) [40]
	0.144	0.4	0.82–1.16			13.76			15.17	Michels et al. (1953) [41]
Propene		9	0.89–1.43			29.24			33.54	Marcham et al. (1949) [42]
		4	0.81–1.23			20.38			21.84	Lange and Stein (1970) [43]
Trifluoromethane	0.260	1.6	0.87–1.34			47.10			48.61	Michels et al. (1952) [44]
Methyl fluoride	0.187	1.8	0.78–1.14			50.87			58.99	Mansoorian et al. (1977) [45]
Methyl chloride	0.153	1.9	0.59–0.83			66.92			74.78	Anderson et al. (1968) [46]
Acetone	0.304	2.9	0.83–1.12			75.56			78.59	Kelland and McLaurin (1969) [47]
Methanol	0.556	1.7	0.7–0.88			60.65			62.30	Moreland et al. (1967) [48]
n-Propanol	0.623	1.7							46.55	
Average										

^a $AAD\% = \sum^N [|Exp. - Cal. | / Exp.] \times 100 / N$.

^b $RMS\% \text{ error} = \sum (\text{percentage error})^2 / N^{0.5}$; Percentage error = $[(Exp. - Calc.) / Exp.] \times 100$.

^c All physical property data from ref. 9.

^d I, De Santis and Grande (1979) [3]; II, Orbey and Vera (1983) [4]; III, this work.

^e All experimental data for third virial coefficient taken from ref. 12.

$$F^{(1)}(T_r) = 9.25492 - 65.50763/T_r + 162.02620/T_r^2 - 183.23773/T_r^3 + 96.85253/T_r^4 - 19.08807/T_r^5 \quad (16b)$$

$$F^{(2)}(T_r) = -1.30215 + 6.56985/T_r - 13.08120/T_r^2 + 12.85166/T_r^3 - 6.24687/T_r^4 + 1.20561/T_r^5 \quad (16c)$$

The $F^{(0)}(T_r)$ subfunction curve-fit final form (eqn. (16a)) was determined by means of linear least-squares regression methods from a set of 76 experimental data points representing argon, krypton and xenon, with a curve-fit variance of 0.001 280 78 and correlation coefficient $R = 0.9573$. The final expression for the coefficient of the $F^{(1)}(T_r)$ subfunction (eqn. (16b)) was obtained similarly from a set of 55 experimental data points representing ethane, *n*-octane, nitrogen, benzene and sulphur hexafluoride, with a variance of 0.035 194 34 and correlation coefficient $R = 0.8862$. The final form of the $F^{(2)}(T_r)$ subfunction (eqn. (16c)) was determined from a set of 22 experimental data points representing trifluoromethane, methyl chloride, methyl fluoride and acetone, with a variance of 0.000 010 67 and correlation coefficient $R = 0.9907$. The experimental data used are listed in Table 1.

DISCUSSION

The third virial coefficients (eqns. (3b), (6), (7), (11)) were calculated using the proposed method (eqns. (12)–(16c)) for 18 non-polar and polar compounds. Table 1 compares the experimental virial coefficients with the values obtained in this work and those calculated by means of the De Santis and Grande [3] and Orbey and Vera [4] correlations. For both, the simple and normal fluids results indicate that the proposed method (eqns. (12), (13a), (13b), (16a), (16b), see Table 1) has a marginal edge in the curve-fit accuracy over the De Santis and Grande (Table 1) and Orbey and Vera (Table 1) correlations. To demonstrate this, Fig. 2 shows the reduced third coefficient values as obtained by means of the proposed method, and by the De Santis and Grande, and the Orbey and Vera correlations, and the available experimental data plotted against the reduced temperature for the normal fluids ethane, methane, sulphur hexafluoride and benzene. As seen from Fig. 2, all the methods compare very well with the experimental data, the proposed method indicating higher accuracy in the high temperature (above the critical point) region. It should be noted that the correlations of De Santis and Grande [3] and Orbey and Vera [4] have been developed to predict the third virial coefficients for non-polar gases only. The correlation of De Santis and Grande [3] requires a knowledge of the dipole polarizability of the molecule, Bondi's molecular volume [49], the

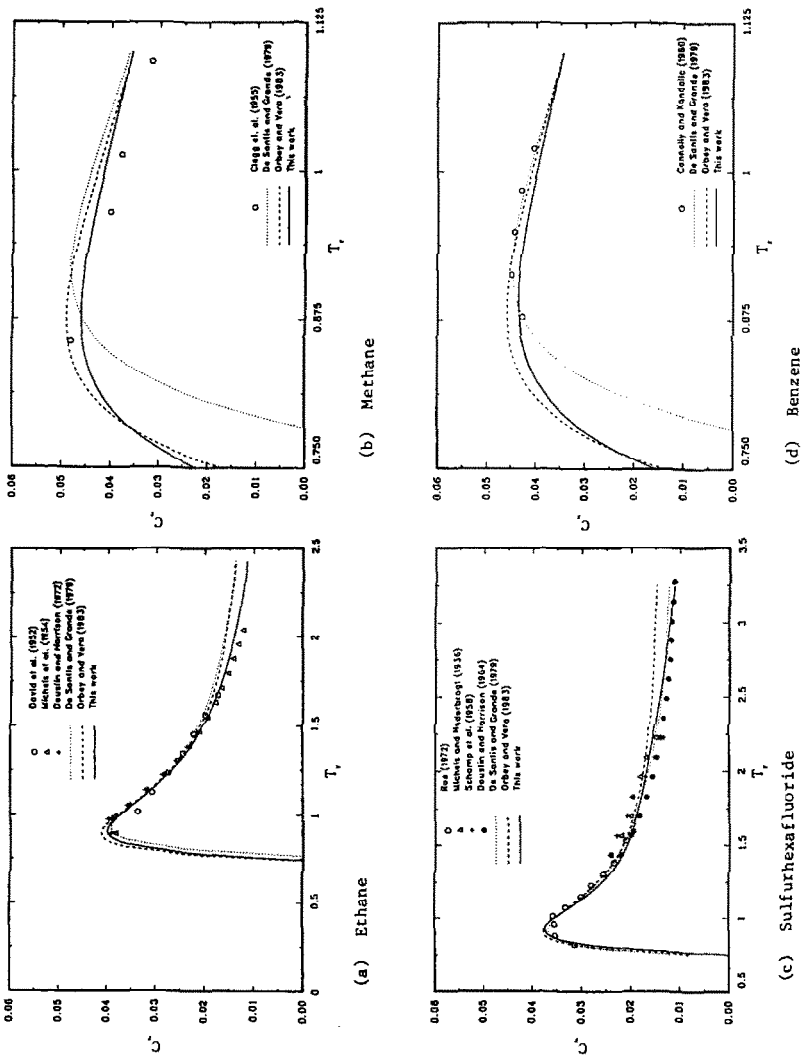


Fig. 2. Third virial coefficient C_3 of non-polar fluids versus reduced temperature T_r : (a) ethane; (b) methane; (c) sulphur hexafluoride; (d) benzene.

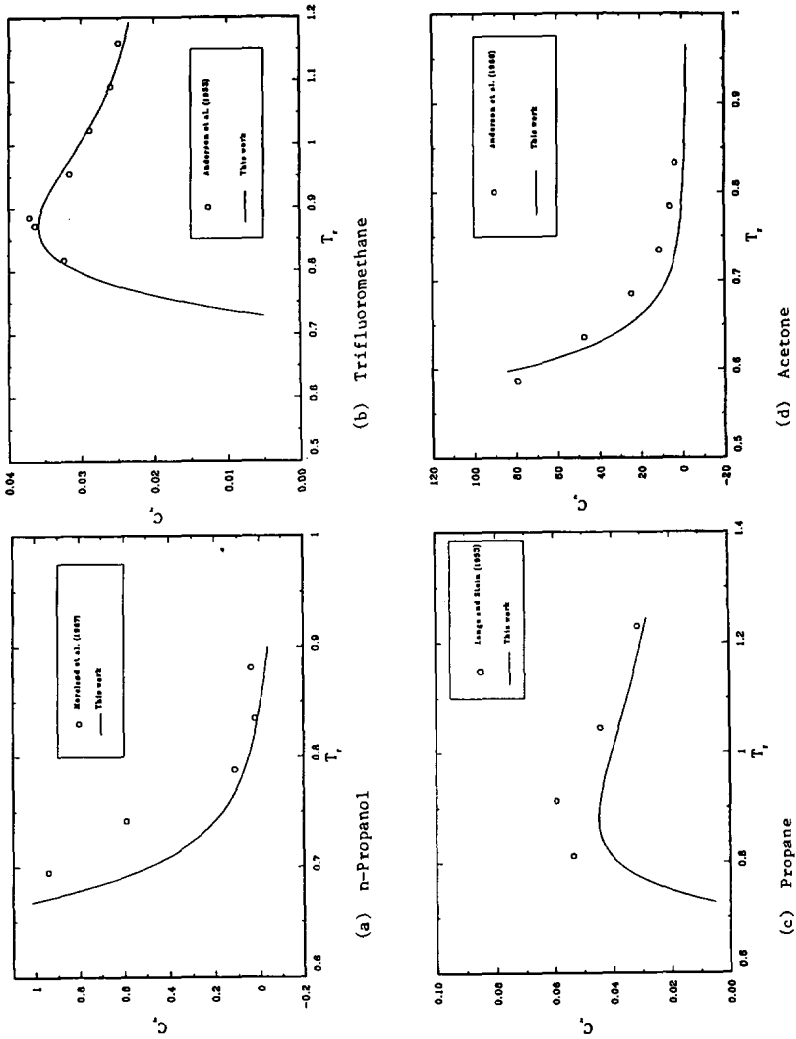


Fig. 3. Third virial coefficient C_r of polar fluids versus reduced temperature T_r : (a) *n*-propanol; (b) trifluoromethane; (c) propene; and (d) acetone.

critical volume, the critical temperature and Pitzer's acentric factor ω . The correlation of Orbey and Vera [4] uses only three characteristic parameters for pure compounds: critical pressure, critical temperature and Pitzer's acentric factor ω . However, the derivation of their proposed generalized empirical correlation is intricate. The proposed method for pure normal fluids (eqns. (12), (13a), (13b), (16a), (16b)) requires two substance-characteristic parameters: critical temperature and Pitzer's acentric factor ω . Equations (13a), (13b), (16a) and (16b) are simple linear polynomials describing the α term (eqns. (4a) and (7)) as a function of the variable T_r . It appears, therefore, that for non-polar fluids the proposed method is altogether simpler than the De Santis and Grande and the Orbey and Vera correlations (Table 1, Fig. 2). For pure polar fluids, the proposed method (eqns. (12), (13c), (16c)), in addition to the critical temperature and Pitzer's acentric factor ω , requires a knowledge of the reduced dipole moment μ_r , the critical pressure (eqn. (9)) and one curve-fitted substance-dependent constant, the optimum x coefficient value (eqns. (11), (12), see Table 1). The polar fluid contribution to the total F function (eqn. (12)) follows the F subfunction development of normal fluids and, similarly, is represented by a simple linear polynomial expansion in reduced temperature, T_r (eqns. (13c) and (16c)). Figure 3 shows the reduced third virial coefficient values as calculated by means of the proposed method (eqns. (12), (13c), (16c), see Table 1) and the available experimental data plotted against the reduced temperature T_r for the polar fluids *n*-propanol, trifluoromethane, propene and benzene. The predicted values appear to be within the range of the experimental accuracy for the third virial coefficients. Romero and Lielmezs [11] in their work on predicting the second virial coefficient for polar fluids have noted that the power series expansion in T_r for the given α function (eqns. (4a) and (7)) has a radius of convergence which extends from the point of expansion to the nearest singularity in the function, so that a power series expansion becomes useless beyond that point. Indeed, Romero and Lielmezs in their work for predicting the second virial coefficient [11] did set up experimentally tested curve-fitting ranges for the state parameter T_r . In the present work, however, owing to the uncertainties in the experimental information available, we were not able to present with certainty such temperature bounds. The results obtained and comparisons made here (Table 1, Figs. 1–3), however, strongly support the proposed method.

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