Second virial coefficients of normal alkanes, linear 1-alkanols and their binaries

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Abstract - The extensive 1986 analysis of Dymond et al. for normal
alkanes is expanded to include recent data on butane, pentane, and
hexane. It now appears that the Hayden-O'Connell or the Tsonopoulos
correlation may be used for $T < 0.65\ T_c$, while the Tsonopoulos
correlation is better for $T > 0.65\ T_c$. The reanalysis also suggests
that the original McCann-Danner group-contribution method should be
used in place of the modifications proposed by Dymond et al.
Consideration is also given to extrapolation to nonane and heavier
normal alkanes. Preliminary recommendations are made for the second
virial coefficients of methanol, ethanol, 1-propanol, and 1-butanol.
Analysis of the IUPAC database with the correlation of Tsonopoulos
suggests that the 1974 recommendations need to be modified only for
methanol: the value $\underline{b} = 0.064$ should replace the earlier choice ($\underline{b}$ =
0.056). Extrapolation to pentanol and heavier linear 1-alkanols is
at present uncertain. The 1979 recommendations of Tsonopoulos for
the second virial cross-coefficients of alkane/alkane and
alkane/1-alkanol binaries are generally supported by recent data.
However, more, high-quality data are needed for C_4+ linear 1-alkanols
and their binaries to develop an improved corresponding-states or a
group-contribution method.

INTRODUCTION

Dymond et al. (ref. 1) recommended second virial coefficient values for the first eight
normal alkanes, following a critical review of all available experimental data. In
addition, these values were used to determine the relative accuracies of various
widely-used correlation methods, as well as of the McCann-Danner (ref. 2) group-
contribution method.

The 1986 review is expanded here by considering recent data on $\underline{n}$-butane, $\underline{n}$-pentane, and
$\underline{n}$-hexane. In addition, for the extrapolation to nonane and heavier normal alkanes, the
available data for critical constants are considered, with special emphasis on the T_c
values, for which the recent DECHEMA recommendations (ref. 3) differ significantly from
those of Ambrose and Teja's data (ref. 4).

Preliminary recommendations are made for the second virial coefficients of the C_1 - C_4
linear 1-alkanols. In addition, the IUPAC database is analyzed with the Tsonopoulos
correlation (ref. 5). Extrapolation to pentanol and heavier linear 1-alkanols is also
considered.

Finally, the Tsonopoulos correlation is also used to analyze second virial cross-
coefficients for alkane/alkane and alkane/1-alkanol binaries. Such data can be
correlated by using a characteristic binary constant k_{ij}, which corrects for the
deviation of T_{cij} from the geometric mean.

NORMAL ALKANES

The 1986 review (ref. 1) is expanded and modified on the basis of new data and a
reevaluation of the entire database.

New data

Table 1 presents new B data for butane (ref. 6), pentane (ref. 7), and hexane (ref. 8).
These data are plotted as solid points in Figs. 1-3. The values calculated for butane
and pentane from speed of sound measurements (refs. 6,7) have an uncertainty which,

TABLE 1.   New B data on n-alkanes
            (additional to the database in ref. 1)

T (K)	B/cm^{3}mol^{-1}		
	butane[a]	pentane[b]	hexane[c]
250	-1081.5		
260	-979.5	-1639	
270	-892.4		
280	-817.6	-1366	
290	-752.6		
300	-695.9	-1156	
310	-646.0		
320	-601.8	-990	
352.95			-1205
373.25			-1020
393.75			-910

[a]ref. 6. [b]ref. 7. [c]ref. 8.

according to Eubank (ref. 9), may be as much as 10%. However, Kohler (ref. 10) considers
the values to have an accuracy of 20-30 cm^{3}mol^{-1}. The other B values (ref. 8) were
obtained from PVT measurements extrapolated to zero density. Uncertainties arising from
adsorption effects are claimed to be low.

Consideration of these new data, together with the previously selected data sets
(ref. 1), has led to new recommendations for butane, pentane, and hexane. These are
given in Table 2 and are compared with the 1986 recommendations in Figs. 1 to 3, where
the vertical lines represent the estimated uncertainties in the recommended values. For
butane, the speed of sound values are less negative than the previously selected data at
low reduced temperatures, and the new recommendations take this into account. In the
case of pentane, little change to the earlier recommendations is considered necessary.
For hexane, the results of Spiske and Gaube (ref. 8) agree more closely with the less
negative sets of selected data (ref. 1) and with B values derived from Joule-Thomson
coefficient measurements (ref. 11), as shown in Fig. 3. Further support for these
changes to the earlier B recommendations (ref. 1), particularly for hexane, is given from
isothermal plots of B versus carbon chain length, for alkanes from butane to octane,
where the points are now found to lie on smooth curves.
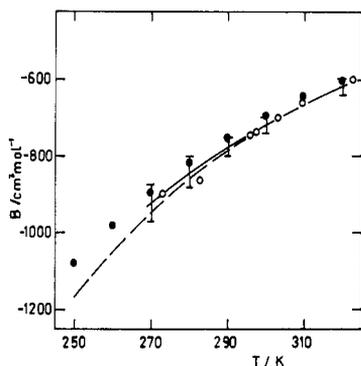
Fig. 1. Second virial
 coefficients of butane.
 - - 1986 recommendations
      (ref 1);
 ___ revised recommendations;
 o selected data (ref. 1);
 ● (ref. 6).
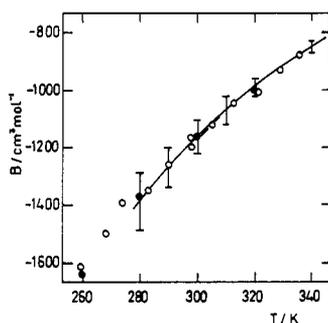
Fig. 2. Second virial
 coefficients of pentane.
 - - 1986 recommendations
      (ref 1);
 ___ revised recommendations;
 o selected data (ref. 1);
 ● (ref. 7).
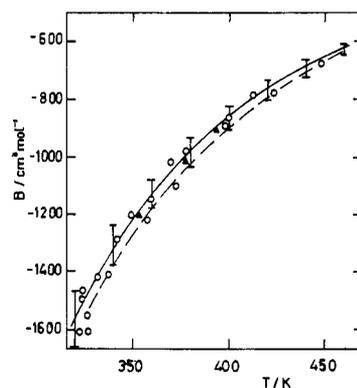
Fig. 3. Second virial
 coefficients of hexane.
 - - 1986 recommendations
      (ref. 1);
 ___ revised recommendations;
 o selected data (ref. 1):
 ▲ (ref. 8).

The new changes lead to the conclusion that the Tsonopoulos correlation (ref. 5) gives
the most satisfactory representation of all the recommended B data for n-alkanes.
Although at low temperatures the Hayden-O'Connell correlation (ref. 12) gives a slightly
better fit to the B values in the case of pentane, it gives poorer agreement with the B
data for butane over this range, and in general this method gives values that are not
sufficiently negative at higher temperatures. Furthermore, it is less clear whether the
McCann-Danner group-contribution method (ref. 2) should be used with the original or the
revised coefficients (ref. 1). It may be preferable to use the original set of
coefficients.

TABLE 2. Revised recommendations for n-alkanes

T/K	B/cm^{3}mol^{-1}		
	n-Butane	n-Pentane	n-Hexane
270	-925 ± 50		
280	-845 ± 40	[-1390 ± 100]	
290	-780 ± 30	[-1270 ± 70]	
300		-1165 ± 60	
310		-1070 ± 50	
320		-990 ± 30	-1570 ± 100
340			-1310 ± 70
360			-1130 ± 50
380			-985 ± 50
400			-860 ± 40
420			-770 ± 30
440			-690 ± 30
460			-620 ± 25
480			-560 ± 20
500			-510 ± 20
520			-460 ± 20
Notes:	270 K corresponds to T_r = 0.635	280 K corresponds to T_r = 0.60; 300 K corresponds to T_r = 0.64	320 K corresponds to T_r = 0.631

Extrapolation

Extrapolation beyond C$_8$ requires reliable values for T_c, P_c, and ω (acentric factor). Such values are available (ref. 4) up to C$_{18}$ for T_c, and up to C$_{13}$ for P_c. (The literature value for the P_c of n-tetradecane is clearly suspect.) At least up to C$_{13}$, therefore, it is possible to use vapor pressure data to determine the acentric factor of the alkane from

$$\omega = -\log_{10}(P_r^{s})_{T_r = 0.7} - 1.0 \qquad (1)$$

where P^s is the vapor pressure and r designates a reduced property (T_r = T/T_c; P_r = P/P_c).

It is important that reliable T_c, P_c, and ω values be used. This is emphasized because the recent, very extensive DECHEMA compilation of critical constants (ref. 3) recommends T_c values for C$_{10+}$ normal alkanes that are significantly different from the recommendations of Ambrose or the experimental results from Teja's group (see ref. 4). The discrepancy is clearly shown in Fig. 4, and it is our conclusion that the DECHEMA recommendations are erroneous.

LINEAR 1-ALKANOLS

Second virial coefficients for 1-alkanols have been tabulated in the compilation by Dymond and Smith (ref. 11), referred to hereafter as DS and, more recently, by Cholinski et al. (ref. 13), abbreviated as CSW. References to data are indicated by the author(s) name(s) and the number in the compilation. In the case of measurements which have not been included in these tabulations, the references are given in full. Recommendations are based on selected data sets. These are sets of measurements from groups which have made a careful study of PVT measurements and where the results cover a wide temperature range, where the analysis has been carried out using at least a three-term virial equation, and where consideration has been given to possible effects of adsorption.

Methanol

There are 23 sets of B data in the database for methanol. In addition to the data tabulated by DS and CSW, measurements of the second virial coefficient for methanol have been made by Eucken and Mayer (ref. 14), Russell and Maass (ref. 15), Weltner and Pitzer (ref. 16), Francis and Phutela (ref. 17), Bich et al. (ref. 18), Ohgaki et al. (ref. 19), and Gaube (ref. 20). The selected data sets are those of Kretschmer and Wiebe (DS- set 2), Kell and McLaurin (DS-6), Kudchadker and Eubank (DS-8), Zubarev and Bagdonas (CSW-14), Fischer et al. (CSW-8,9,10), and Bich et al. (ref. 18). There is close agreement between these sets of B values and the recommended values given in Table 3, which were obtained from a smooth curve through the points. These values agree to within the estimated uncertainties with predictions from the Gibbs energy equation of state recently derived

TABLE 3.   Recommended B values for 1-alkanols

	B/cm^{3}mol^{-1}			
T/K	Methanol	Ethanol	1-Propanol	1-Butanol
320	-1260 ± 60			
340	-850 ± 40	-1200 ± 100		
360	-635 ± 30	-870 ± 80		
380	-502 ± 25	-670 ± 50	-870 ± 50	-1215 ± 80
400	-406 ± 20	-535 ± 40	-705 ± 40	-990 ± 50
420	-330 ± 20	-435 ± 30	-595 ± 30	-795 ± 50
440	-275 ± 20	-365 ± 30	-516 ± 30	-615 ± 40
460	-233 ± 20	-314 ± 25	-446 ± 25	
480	-203 ± 15	-273 ± 20	-385 ± 20	
500	-179 ± 15	-240 ± 20	-337 ± 20	
520	-160 ± 15	-215 ± 15	-298 ± 20	
540	-143 ± 15	[-195 ± 15]	-267 ± 20	
560	-128 ± 15		-242 ± 20	
580	-116 ± 15			
600	-104 ± 15			

(ref. 21) from PVT data (including saturated vapor densities), gas phase specific enthalpy data, isobaric heat capacity measurements, and speed of sound data. It should be noted that the values derived from Joule-Thomson measurements (ref. 17) are less negative by about 60 cm^{3}mol^{-1} at 323 K. There is, however, some uncertainty in extrapolating these results to zero pressure.

Ethanol

There are 10 sets of B data for ethanol. Along with the data given in DS and CSW, second virial coefficients have been reported by Francis and Phutela (ref. 17), Bich et al. (ref. 22), and Wilson et al. (ref. 23). The selected data sets are those of Kretschmer and Wiebe (DS-2) and Bich et al. (ref. 22). It should be noted that, at high temperatures (T > 493 K), Bich et al. (ref. 22) found evidence for decomposition. The results they obtained by direct calculation, assuming no decomposition, are shown as the dashed line in Fig. 5. The inflection and concave-down curvature are further evidence that these results are erroneous. The B values which they calculated taking decomposition into account become increasingly more negative than the other values at higher temperatures. Because of the uncertainties associated with decomposition, the recommended data in Table 3 are given at temperatures only up to 520 K. At lower temperatures, B values derived from isothermal Joule-Thomson coefficients (ref. 17) are less negative, but again there are uncertainties here arising from extrapolation to zero pressure. The agreement with the recommended values, taking account of the estimated uncertainties, can therefore be considered reasonable.
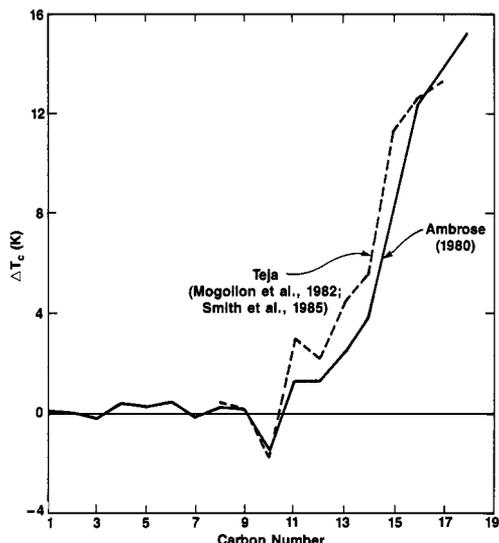

Fig. 4. Deviation of DECHEMA recommendations for T$_c$ of normal alkanes from Ambrose and Teja (see ref. 4).
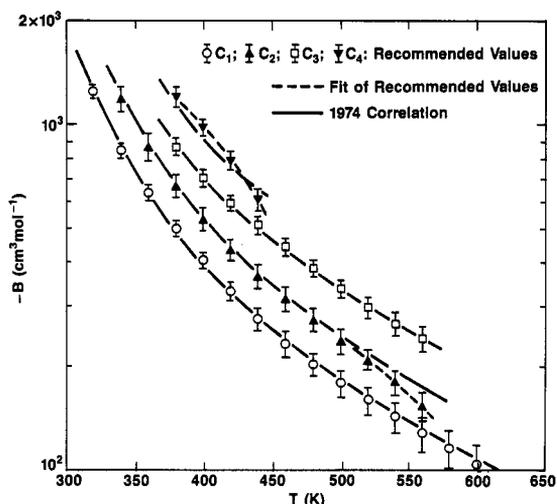
Fig. 5. Second virial coefficients of linear 1-alkanols.

1-Propanol

In addition to the two sets of data given by DS, measurements have been made by Markuzin and Sokolova (CSW-3) and more recently by Zawisza and Vejrosta (ref. 24). There is excellent agreement between this last set of measurements and those of Cox (DS-2), which are taken as the selected data sets, and this leads to the recommendations given in Table 3.

1-Butanol

Of the two data sets for this compound, only the values of Cox (DS-2) are selected. Recommendations based on these values are given in Table 3. However, a smooth line through these values exhibits the wrong curvature (dashed line in Fig. 5).

1-Pentanol

The only source of B data is the vapor pressure and enthalpy of vaporization data given by Counsell et al. (ref. 25). The uncertainty in any derived second virial coefficients would be too large to make any recommendation.

The C_1-C_4 1-alkanol B data have been analyzed with the correlation of Tsonopoulos, which is described in the next section.

THE TSONOPOULOS B CORRELATION

In reduced form, the Tsonopoulos correlation (ref. 5) is written as

$$\frac{BP_c}{RT_c} = f^{(0)}(T_r) + \omega f^{(1)}(T_r) + f^{(2)}(T_r) \tag{2}$$

where

$$f^{(0)}(T_r) = 0.1445 - 0.330/T_r - 0.1385/T_r^2 - 0.0121/T_r^3 - 0.000607/T_r^8 \tag{3}$$

$$f^{(1)}(T_r) = 0.0637 + 0.331/T_r^2 - 0.423/T_r^3 - 0.008/T_r^8 \tag{4}$$

$$f^{(2)}(T_r) = a/T_r^6 - b/T_r^8 \tag{5}$$

$f^{(2)}(T_r)$ is the polar term. For non-hydrogen bonding polar compounds, such as ketones, it is only necessary to use a; that is, b = 0. Since the B of polar compounds is more negative than that of nonpolar compounds (for the same T_r and ω), it follows that $f^{(2)} < 0$ or a < 0. However, the $f^{(2)}$ of dimerizing compounds, such as alkanols, has a more complex temperature dependence: it is steeper at very low T_r (<0.6), but is flatter at $T_r > 0.8$. As a result, both parameters (which assume positive values) in Eq. 5 must be used.

Tsonopoulos set a = 0.0878 for all alkanols and then optimized the value of b, which was found to depend weakly on the reduced dipole moment of the alkanol. The results of the analysis of the IUPAC database with Eqs. 2-5 are summarized in Table 4 and in Fig. 5 (solid lines). The database is adequate only for methanol and ethanol, and only for methanol it is significantly different from that used in the 1974 correlation. A reanalysis of the data suggests that for methanol b = 0.064 gives a slightly better fit of the data than the 1974 value b = 0.056 (in both cases, a = 0.0878).

TABLE 4. Comparison of 1974 correlation with IUPAC database

Alkanol	No. Points	Average Absolute Deviation, cm^3mol^{-1}	Bias (EXP-CAL), cm^3mol^{-1}
Methanol	117	95.5	-77.1
Ethanol	65	137.6	-85.4
1-Propanol	17	127.2	59.6
1-Butanol	7	80.2	-48.3

Although the reanalysis still supports the 1974 observation that $\underline{b}$ is a weak function of the reduced dipole moment, this dependence can be confirmed only with reliable B data for C_4+ alkanols. Such data are still not available.

CROSS-COEFFICIENTS FOR MIXTURES

The second virial cross-coefficient, B_{ij}, has the same temperature dependence that B_{ii} and B_{jj} have, but the parameters to be used with Eqs. 2-5 are P_{cij}, T_{cij}, ω_{ij}, a_{ij}, and b_{ij}. The following mixing rules make it possible to relate these characteristic constants to pure-component parameters:

$$T_{cij} = (T_{ci}T_{cj})^{1/2} \quad (1 - k_{ij}) \tag{6}$$

$$P_{cij} = \frac{4T_{cij} \; (P_{ci}v_{ci}/T_{ci} + P_{cj}v_{cj}/T_{cj})}{(v_{ci}^{1/3} + v_{cj}^{1/3})^3} \tag{7}$$

$$\omega_{ij} = 0.5 \; (\omega_i + \omega_j) \tag{8}$$

where k_{ij} is a characteristic constant for each binary.

Equations 6-8 suffice for nonpolar/nonpolar binaries. For polar/nonpolar binaries, B_{ij} is assumed to have no polar term:

$$a_{ij} = 0 \tag{9}$$

$$b_{ij} = 0 \tag{10}$$

For polar/polar binaries, the polar contribution to B_{ij} is calculated by assuming that:

$$a_{ij} = 0.5 \; (a_i + a_j) \tag{11}$$

$$b_{ij} = 0.5 \; (b_i + b_j) \tag{12}$$

The most sensitive mixing rule is Eq. 6. T_{cij} can be assumed to be the geometric mean of T_{ci} and T_{cj} ($k_{ij} = 0$) only when i and j are very similar in size and chemical nature. Otherwise, in the absence of any specific chemical interaction between $\underline{i}$ and $\underline{j}$, k_{ij} should be positive and thus T_{cij} would be less than the geometric mean.

Alkane/alkane binaries

As shown by Tsonopoulos (ref. 26), the optimum k_{ij}'s for alkane/alkane binaries are well represented by a relationship proposed by Chueh and Prausnitz (ref. 27):

$$k_{ij} = 1 - \left[\frac{2 \; (v_{ci} \; v_{cj})^{1/6}}{v_{ci}^{1/3} + v_{cj}^{1/3}} \right]^3 \tag{13}$$

The success of Eq. 13 was further confirmed by analyzing the extensive data of Wormald et al. (ref. 28). This is demonstrated in Fig. 6, a parity plot that also includes the data of D'Avila et al. (ref. 29) and of Kaul and Prausnitz (ref. 30). Generally, Eq. 13 reproduces the optimum k_{ij} values within ± 0.02.

Alkane/1-alkanol binaries

On the basis of limited data, Tsonopoulos (ref. 5) recommended an average value of $k_{ij} = 0.15$ for hydrocarbon/alkanol binaries. Additional data were analyzed in 1979 (ref. 26) and the average k_{ij} for 10 hydrocarbon/alkanol binaries was found to be 0.16 ± 0.03. However, some trends in the k_{ij} values were noted, as shown in Fig. 7, where k_{ij} has been plotted versus v_{ci}, the critical volume of the nonpolar component. These 1979 results have been confirmed by the data of Zawisza and Vejrosta (ref. 24) for $\underline{n}$-heptane/1-propanol (the inverse triangle in Fig. 7).

Although Fig. 7 presents a relatively clear picture for the k_{ij}'s of n-alkane/1-alkanol binaries, some questions remain. For example, the B_{ij} data of Lazalde-Crabtree et al. (ref. 31) for methane/methanol can be fitted only with temperature-dependent k_{ij}'s:

T/K	k_{ij}
227.6	-0.075
250.7	-0.023
273.2	0.073

Such temperature dependence would be consistent with formation of a methane/methanol complex, but that is unlikely. It should be noted that Lazalde-Crabtree's data for ethane/methanol are in good agreement with the k_{ij} plotted in Fig. 7 (0.12; the plotted value for the methane binary of methanol is 0.13).

The data of Rätzsch and Freydank (ref. 32) for n-pentane/methanol suggest a k_{ij} between 0.01 and 0.02. However, the same authors' data for ethylene/methanol are about 500 cm^3mol^{-1} more negative than those of Hemmaplardh and King (ref. 33). Finally, the single value at 308.15 K of Markuzin and Baidin (33) for n-hexane/ethanol requires a $k_{ij} = -0.1$.

In view of these questions, it is not possible to make a definitive statement on the B_{ij} of n-alkane/1-alkanol binaries. However, Fig. 7 may be closer to the "truth" and can be used to predict the k_{ij} of binaries of C_1-C_4 1-alkanols with n-alkanes--or nonpolar compounds, in general.
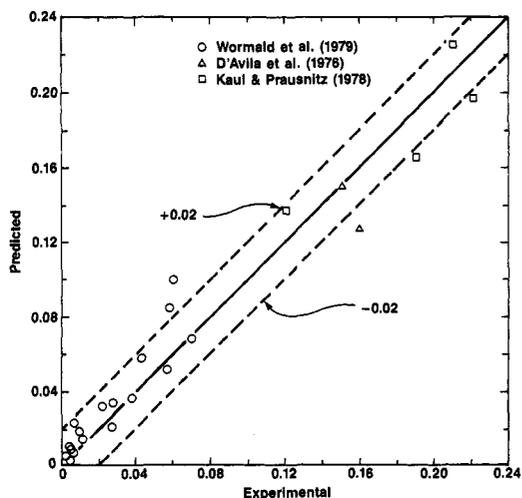

Fig. 6. Prediction of k_{ij} values for normal alkane/ alkane binaries with Eq. 13.
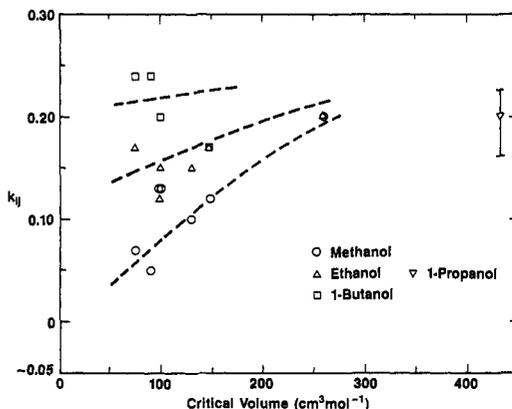
Fig. 7. Optimum k_{ij} values for nonpolar binaries of linear 1-alkanols.

CONCLUDING REMARKS

The 1986 review (ref. 1) and the additions made here for n-alkanes present a satisfactory picture for the first eight n-alkanes. Extrapolation to C_9+ n-alkanes with a corresponding-states correlation requires only T_c, P_c, and ω, which can be taken from Ambrose or the ongoing investigations of Teja's group (ref. 4). Similarly satisfactory is the situation of n-alkane/n-alkane binaries, for which the characteristic binary constant k_{ij} used in Tsonopoulos's correlation (ref. 5) can be predicted reliably with Eq. 13.

The situation is much less satisfactory for linear 1-alkanols and their binaries with n-alkanes. A sufficient database exists only for methanol and ethanol. More, high-quality data are needed at least for 1-butanol and all higher 1-alkanols. The correlation of Tsonopoulos (ref. 5) can be used to make limited extrapolations, but cannot be taken too far because of the limited database. An enlarged, reliable database will also make possible the development of a group-contribution method for 1-alkanols.

Finally, Fig. 7 presents a tentative correlation for the k_{ij} of n-alkane (more generally, any nonpolar compound)/1-alkanol binaries, but conflicts with other data must be resolved. That will require high-quality data on binaries of C_3+ n-alkanes with any 1-alkanol. Until these conflicts are resolved, any extrapolations would have to be considered highly uncertain. Very tentatively, an upper limit of $k_{ij} \approx 0.25$ is suggested for all binaries of 1-alkanols with C_8+ n-alkanes.

NOTATION

a, b	=	parameters of polar contribution term to B, $f^{(2)}$; see Eq. 5
B	=	second virial coefficient
$f^{(0)}$, $f^{(1)}$, $f^{(2)}$	=	dimensionless terms of Eq. 2
k_{ij}	=	characteristic binary constant; see Eq. 6
P	=	pressure
P^s	=	vapor pressure of pure component
R	=	gas constant
T	=	absolute temperature
v	=	molar volume
V	=	total volume
ω	=	acentric factor

Subscripts

c	=	critical property
i, j	=	property of component i, j
ij	=	characteristic property used in the calculation of the second virial cross-coefficient
r	=	reduced property

REFERENCES

1. J. H. Dymond, J. A. Cholinski, A. Szafranski, and D. Wyrzykowska-Stankiewicz, <u>Fluid Phase Equil.</u>, <u>27</u>, 1-13 (1986).
2. D. W. McCann and R. P. Danner, <u>Ind. Eng. Chem. Process Des. Dev.</u>, <u>23</u>, 529-533 (1984).
3. K. H. Simmrock, R. Janowsky, and A. Ohnsorge, <u>Critical Data of Pure Substances</u>, DECHEMA Chemistry Data Series, Vol. II, Frankfurt/Main (1986).
4. C. Tsonopoulos, <u>AIChE Journal</u>, <u>33</u>, 2080-2083 (1987).
5. C. Tsonopoulos, <u>AIChE Journal</u>, <u>20</u>, 263-272 (1974).
6. M. B. Ewing, A.R.H. Goodwin, M. L. McGlashan, and J. P. M. Trusler, <u>J. Chem. Thermodyn.</u>, <u>19</u>, 721-739 (1987).
7. M. B. Ewing, personal communication (1987).
8. J. Spiske and J. Gaube, <u>Chem. Eng. Technol.</u>, <u>10</u>, 143-150 (1987).
9. P. T. Eubank, personal communication (1988).
10. F. Kohler, personal communication (1988).
11. J. H. Dymond and E. B. Smith, <u>The Virial Coefficients of Pure Gases and Mixtures</u>, Clarendon Press, Oxford (1980).
12. J. G. Hayden and J. P. O'Connell, <u>Ind. Eng. Chem. Process Des. Dev.</u>, <u>14</u>, 209-216 (1975).
13. J. Cholinski, A. Szafranski, and D. Wyrzykowska-Stankiewicz, <u>Computer-aided Second Virial Coefficient Data for Organic Individual Compounds and Binary Systems</u>, PWN Polish Scientific Publishers, Warsaw (1986).
14. A. Eucken and L. Mayer, <u>Z. Phys. Chem.</u>, <u>B5</u>, 452-466 (1929).
15. J. Russell and O. Maass, <u>Can. J. Res.</u>, <u>5</u>, 436-441 (1931).
16. W. Weltner and K. S. Pitzer, <u>J. Amer. Chem. Soc.</u>, <u>77</u>, 2606-2610 (1951).
17. P. G. Francis and R. C. Phutela, <u>J. Chem. Thermodyn.</u>, <u>11</u>, 747-756 (1979).
18. E. Bich, R. Pietsch, and G. Opel, <u>Z. Phys. Chem., Leipzig</u>, <u>265</u>, 396-400 (1984).
19. K. Ohgaki, T. Hanafusa, Y. Kano, and T. Katayama, <u>J. Chem. Eng. Japan</u>, <u>17</u>, 329-331 (1984).
20. J. Gaube, personal communication (1988).
21. P. Topalis, R.J.B. Craven, K. M. de Reuck, and W. A. Wakeham, <u>Pure Appl. Chem.</u>, to be published.
22. E. Bich, M. Ramsdorf, and G. Opel, <u>Z. Phys. Chem., Leipzig</u>, <u>265</u>, 401-404 (1984).
23. K. S. Wilson, D. D. Lindley, W. B. Kay, and H. C. Hershey, <u>J. Chem. Eng. Data</u>, <u>29</u>, 243-245 (1984).
24. A. Zawisza and J. Vejrosta, <u>J. Chem. Thermodyn.</u>, <u>14</u>, 239-249 (1982).
25. J. F. Counsell, J. O. Fenwick, and E. B. Lees, <u>J. Chem. Thermodyn.</u>, <u>2</u>, 367-372 (1970).
26. C. Tsonopoulos, <u>Advan. Chem. Ser.</u>, No. 182, 143-162 (1979).
27. P. L. Chueh and J. M. Prausnitz, <u>AIChE Journal</u>, <u>13</u>, 1099-1107 (1967).
28. C. J. Wormald, E. J. Lewis, and D. J. Hutchings, <u>J. Chem. Thermodyn.</u>, <u>11</u>, 1-12 (1979).
29. S. G. D'Avila, B. K. Kaul, and J. M. Prausnitz, <u>J. Chem. Eng. Data</u>, <u>21</u>, 488-491 (1976).
30. B. K. Kaul and J. M. Prausnitz, <u>AIChE Journal</u>, <u>24</u>, 223-231 (1978).
31. H. Lazalde-Crabtree, G.J.F. Breedveld, and J. M. Prausnitz, <u>AIChE Journal</u>, <u>26</u>, 462-470 (1980).
32. M. Rätzsch and H. Freydank, <u>J. Chem. Thermodyn.</u>, <u>3</u>, 861-866 (1971).
33. B. Hemmaplardh and A. D. King, Jr., <u>J. Phys. Chem.</u>, <u>76</u>, 2170-2175 (1972).
34. N. P. Markuzin and V. N. Baidin, <u>Vestnik Leningrad Univ., Ser. Fiz. Khim.</u>, No. 2, 77 (1973).