Vapour pressures up to their critical temperatures of normal alkanes and 1-alkanols

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Abstract - Values for the critical temperatures and critical pressures, and coefficients of equations for the vapour pressures of normal alkanes and 1-alkanols are given. They are based on surveys of the available data and estimates of the critical properties of the higher members. The resultant vapour-pressure curves show a progressive change in shape and there are corresponding changes in the coefficients of the equations.

INTRODUCTION

ESDU International publishes Data Items providing reliable numbers for use by engineers and designers, and this paper describes the work underlying current issues of Items on the vapour pressures of alkanes and alkanols. These Items (ref. 1,2) contain comprehensive lists of references to the relevant experimental work. Values have been selected for the critical temperatures and pressures, and equations are proposed for representation of the vapour pressures. The coefficients of these equations fall into a pattern reflecting the characteristic shapes of vapour-pressure curves. The form of equation used for representation of vapour pressures requires values for the critical temperature and pressure but the critical points of the higher members are experimentally inaccessible, and for these compounds it was necessary to estimate values for the critical temperature and pressure.

THE VAPOUR-PRESSURE CURVE

Figure 1 shows the vapour pressures of benzene and 1-propanol as deviations $\ln(p/p')$ from the line given by the reduced equation,

$$\ln(p'/p_c) = B(T_c/T - 1) = B(1/T_r - 1),\qquad(1)$$

where p, p', p_c, T, T_c, and $T_r = T/T_c$ are respectively vapour pressure,
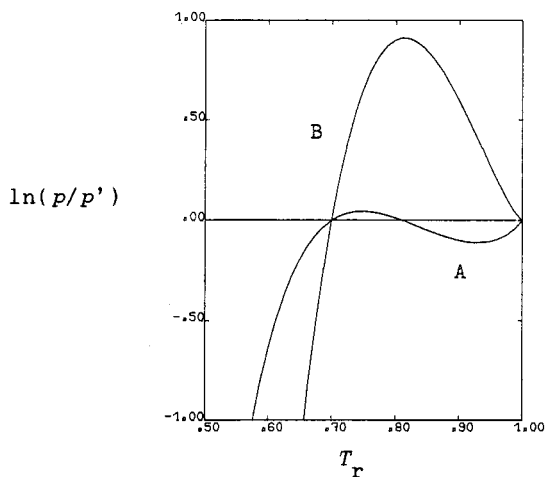

$\ln(p/p')$

Fig. 1 Vapour pressures of benzene (A) and 1-propanol (B).

pressure calculated from equation (1), critical pressure, temperature,
critical temperature, and reduced temperature. Equation (1) is fitted to
observed values at a reduced temperature 0.7, this temperature being
convenient as that at which the acentric factor ω is defined (ref. 3). Such a
plot is more sensitive than a direct plot of vapour pressures in showing the
characteristic shape of the curves that we believe to be universal (ref. 4).
The key points about the curve for the non-associated benzene are that the
slope at the critical temperature is greater than B, the slope of the line of
equation (1), and that there is a reversal of curvature at a reduced
temperature about 0.8. Mills (ref. 5) found indications of this change in
curvature from an analysis of the 19th century measurements by Young, and it
is implicit in the equation proposed by Cox (ref. 6), and in the method
used by Kay for presentation of his experimental results (ref. 7,8), but its
existence was first stated explicitly by Thodos (ref. 9). The slope of the
curve for the associated substance 1-propanol is less than B and the curve
lies entirely above the line of equation (1) between a reduced temperature
0.7 and 1; reversal of the curvature is close to the critical temperature and
is barely demonstrable experimentally, and with some vapour-pressure
formulations will not be apparent.

The reversal in curvature corresponds to the minimum in $\Delta H/\Delta Z$ noted by
Waring (ref. 10), where ΔH is the enthalpy of vaporization and ΔZ is the
compression factor pV/RT. Waring based his work on experimental values of ΔH
and ΔZ, but we can also obtain the ratio of the two quantities by
differentiation of a vapour-pressure equation because

$$\Delta H/\Delta Z = RT^2 \mathrm{dln}p/\mathrm{d}T, \tag{2}$$

and it appears from vapour-pressure studies (ref. 4,11) that, certainly for
non-associated substances and highly probably for associated substances,
there is always a minimum in $\Delta H/\Delta Z$.

The equation we have used for representation of the vapour-pressure curve
is the four-constant form of that proposed by Wagner (ref. 12-14),

$$\ln(p/p_c) = (a\,\tau + b\,\tau^{1.5} + c\,\tau^{2.5} + d\,\tau^5)/T_r, \tag{3}$$

where $\tau = 1 - T_r$.

Unlike those in some vapour-pressure equations the series in equation (3)
appears always to be highly convergent when the equation is fitted to a
long-range set of good data. The first term converts to an equation of form
similar to equation (1) and taken alone provides a first approximation of the
final result in the calculation of vapour pressures. The coefficient of the
first term is equal to $-\mathrm{dln}p/\mathrm{dln}T$ at the critical temperature and it is
therefore equal in magnitude to the quantity used by Riedel in his analysis
of vapour pressures, α_c (ref. 15). Riedel found that α_c for the organic
compounds he studied ranged between 6 and 8, and this, therefore, indicates
the values we may expect to find for the first coefficient. The remaining
terms of equation (3) are correcting terms that introduce curvature to the
straight line given by the first term, each one, except in some instances at
very low reduced temperatures, having a smaller effect than its predecessor.
The last term does not have a significant effect on the calculated vapour
pressure at reduced temperatures greater than 0.7. A fifth term with a higher
power should have an even smaller effect and should, we believe, only be
introduced for the most accurate long-range sets of observations. Generally
the effect of the fourth term on the temperature of the minimum in $\Delta H/\Delta Z$ is
small and the reduced temperature of the minimum is given approximately
(within 0.01) by

$$T_r = 1 + 0.2b/c. \tag{4}$$

Equation (4) shows, alternatively, that the ratio b/c depends on the
reduced temperature of the minimum in $\Delta H/\Delta Z$.

What has just been written must, however, be qualified in respect of
simple gases and the lower alkanes because for these substances the
fourth coefficient has a relatively high value in comparison with its
predecessors, and for these substances the fourth coefficient must be brought
into the calculation of the $\Delta H/\Delta Z$ minimum because equation (4) gives too low
a value - a point not noted in an earlier statement of the equation (ref.
16).

CRITICAL TEMPERATURES AND PRESSURES

Figure 2 shows the experimental values of critical temperature for the
n-alkanes and 1-alkanols plotted against carbon number, n. For the lower
members the preferred values depend on assessment of the reliability of
experimental observations but even for the alkanes the chain cannot be
lengthened very much before decomposition interferes with experiment, and the
observed values for compounds above decane may be doubtful. For the higher
members above hexadecane the critical point is experimentally inaccessible
with techniques presently available. For these compounds the critical
temperature and pressure are not measureable quantities, but they are of
importance for application of the principle of corresponding states, and the
form of equation (3) requires values for them for its fitting and evaluation;
they must, therefore, be estimated. For the intermediate compounds smoothed
or estimated values that are within the probable error limits of observations
may be considered more reliable than those observed. For most other series
decomposition sets in with a much smaller molecule, and for the alkanols it
begins to become significant at pentanol. Figure 3 shows the critical
pressures of the two series, and for this property the reliable experimental
basis fails at an even smaller carbon number than it does for critical
temperature. Clearly the observed critical pressure of tetradecane is one we
must doubt, and shall adjust, whatever method of estimation is adopted for
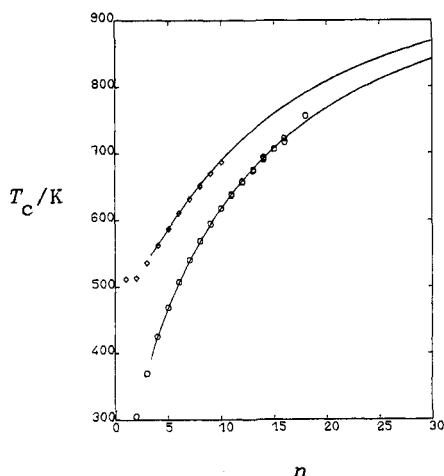the higher members of the series.

Fig. 2. Critical temperatures of
n-alkanes (A) and 1-alkanols (B);
curve for alkanes from
equation (6); curve for
1-alkanols related to that for
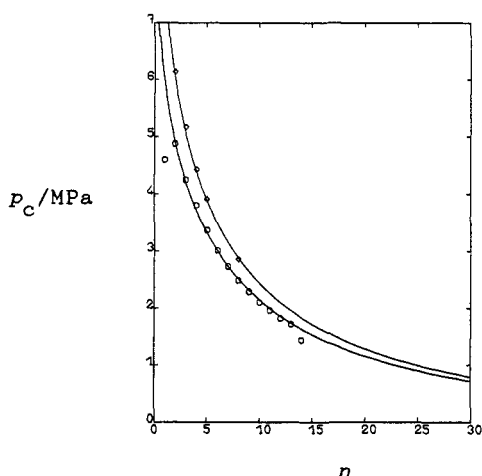alkanes by equation (9).

Fig. 3. Critical pressures of
n-alkanes (A) and 1-alkanols (B);
curve for alkanes from equation (12)
with power 0.5 for n; curve for
1-alkanols related to that for
alkanes by equation (11).

CRITICAL TEMPERATURES AND PRESSURES OF ALKANES

Many methods have been proposed for the estimation of critical properties.
The discussion here in relation to the critical temperatures of alkanes will
be confined to the methods proposed by Riedel (ref. 17) and developed by
Ambrose (ref. 18-21), and one proposed by Kreglewski (ref. 22) and recently
discussed by Tsonopoulos (ref. 23).

Riedel found that the quantity $X = T_b/(T_c - T_b)$, where T_b is the normal boiling
point, varies linearly with carbon number for the n-alkanes. In accordance
with the numbers proposed by Ambrose, if *n* is the carbon number,

$$X = 1.242 + 0.134n. \tag{5}$$

We used equation (5) to calculate the critical temperatures of the higher
members. Equation (5) is an empirical correlation without theoretical basis
for its extrapolation beyond the range of experimental observations but it

has a practical justification in that no correlation fits the observed values better. Kreglewski (ref. 22) proposed (with slightly different coefficients) the equation,

$$\ln(960 - T_c/K) = 6.8162 - 0.2115n^{2/3}, \qquad (6)$$

which implies that the critical temperature approaches a limiting value 960 K for an infinite chain length, and Tsonopoulos has shown (ref. 23) that although equation (5) and equation (6) diverge at higher carbon numbers they give substantially the same values for alkanes up to eicosane, the upper bound for consideration in this work. The curve plotted for the alkanes in Fig. 2 is that arising from equation (6).

For the correlation of critical pressures Ambrose (ref. 20,21,24) revised the equation used by Lydersen (ref.25) and found that

$$(p_c/MPa)^{1/2} (M/kg\ mol^{-1})^{-1/2} = 0.0339 + 0.0226n, \qquad (7)$$

where M is the molar mass. We calculated the critical pressures of the higher alkanes from equation (7).

VAPOUR PRESSURES OF ALKANES

Reference 1 includes a comprehensive list of references to the vapour pressure data for these compounds. The data are generally of good quality over long ranges for the alkanes up to octane, and it was not surprising that their equations conformed to the expected conditions with coefficients of the sizes we expected. For the higher members the data do not extend up to the critical point and for these compounds we are in effect interpolating between measurements around atmospheric pressure and a critical point that for some compounds is itself slightly uncertain. The equations for these compounds did not always conform at the first fitting, and to make them do so the values for the critical temperature and pressure were adjusted slightly. In order not to give a false impression of accuracy we have not specified in Table 1 the values of critical temperature more closely than 1 K, and of critical pressure more closely than 0.005 MPa for the alkanes above dodecane.

TABLE 1. n-Alkanes - critical temperatures and pressures and coefficients of equation (3)

alkane	T_c/K	p_c/MPa	a	b	c	d	ω
methane	190.551	4.5992	-6.02242	1.26652	-0.5707	-1.366	0.011
ethane	305.33	4.8714	-6.47500	1.41071	-1.1440	-1.859	0.099
propane	369.85	4.247	-6.76368	1.55481	-1.5872	-2.024	0.152
butane	425.25	3.792	-7.01763	1.67770	-1.9739	-2.172	0.199
pentane	469.80	3.375	-7.30698	1.75845	-2.1629	-2.913	0.251
hexane	507.90	3.035	-7.53998	1.83759	-2.5438	-3.163	0.299
heptane	540.15	2.735	-7.77404	1.85614	-2.8298	-3.507	0.350
octane	568.95	2.490	-8.04937	2.03865	-3.3120	-3.648	0.397
nonane	594.90	2.290	-8.32886	2.25707	-3.8257	-3.732	0.443
decane	617.65	2.105	-8.60643	2.44659	-4.2925	-3.908	0.490
undecane	638.85	1.955	-8.85076	2.60205	-4.7305	-4.081	0.533
dodecane	658.65	1.83	-9.08593	2.77846	-5.1985	-4.173	0.573
tridecane	676	1.71	-9.32959	2.89925	-5.5550	-4.470	0.618
tetradecane	693	1.61	-9.54470	3.06637	-6.0070	-4.530	0.654
pentadecane	708	1.515	-9.80239	3.29217	-6.5317	-4.584	0.696
hexadecane	722	1.435	-10.03664	3.41426	-6.8627	-4.863	0.737
heptadecane	735	1.37	-10.23600	3.54177	-7.1898	-5.000	0.77
octadecane	746	1.30	-10.47230	3.69655	-7.5779	-5.109	0.8??
nonadecane	758	1.23	-10.68217	3.98054	-8.3030	-4.995	0.8₄4
eicosane	769	1.16	-10.97958	4.25588	-8.9573	-5.043	0.891

Coefficients of the equations are in Table 1 but their remarkably regular stepwise change with carbon number is more easily seen from the plot in Fig. 4. Even the last coefficient, for which we had not previously found such regularity (ref. 26), behaves reasonably. Figure 4 also includes a plot of ω calculated from the equations. Figure 5 shows curves for methane, ethane,

butane, hexane, octane, decane, tetradecane, and octadecane plotted in the
same way as Fig. 1. Figure 6 shows plots of $\Delta H/\Delta Z$ for the same compounds
calculated from the equations with the minima for all the compounds from
methane to eicosane. The minimum advances from a reduced temperature about
0.7 for methane to about 0.9 for octane and the higher alkanes.

All the equations fit what we believe to be good experimental observations,
but for some of the higher members we have placed greater weight on the
coefficient pattern of the equations than on what we believe to be less
reliable observations. The equations have a reasonable but not perfect fit to
the very low-pressure values obtained by King and Al-Najjar for hexane to
decane, and dodecane, tetradecane, and hexadecane by extrapolations based on
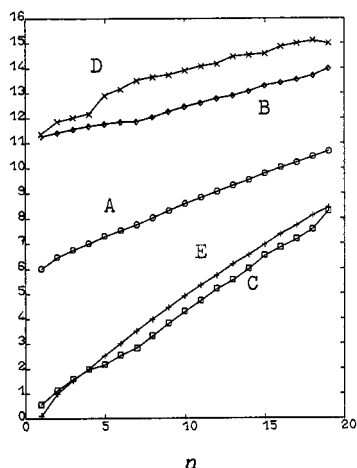heat capacity measurements (ref. 27).

Fig. 4. n-Alkanes - coefficients
of equation (3) and ω.
A, $-a$; B, $10+b$; C, $-c$; D, $10-d$;
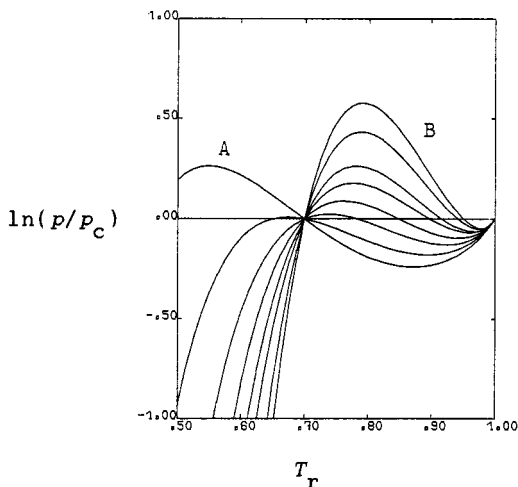E, 10ω.

Fig. 5. Vapour pressures of methane (A),
ethane, butane, hexane, octane,
decane, tetradecane, and
octadecane (B).

Equation (3) was used for the alkanes in ref. 1 with the powers 3 and 6
for the two last terms instead of 2.5 and 5, and the coefficients in Table 1
therefore differ from those in ref. 1. We made the change from one form to
the other as a result of the work reported in ref. 14 in which it was
concluded that the coefficients of the (2.5,5) form of equation (3)
generally fell into a more consistent pattern than those of the (3,6) form.
However, the coefficients in ref. 1 also vary regularly; there are no
significant differences between the fits of the two forms of the equation to
the observed values.

PRINCIPLE OF CORRESPONDING STATES

We believe it has not previously been stated that the progressive change in
shape of the curves in Fig. 5 is consequent upon the principle of
corresponding states, and a similar figure may be obtained on the basis of
the equations given by Lee and Kesler (ref. 28). We have found for the
alkanes that we can improve on those equations by introducing an additional
term as follows

$$\ln p_r = \ln p_r^{(0)} + \omega \ln p_r^{(1)} + \omega^2 \ln p_r^{(2)}, \qquad (8)$$

where each of the terms is expressed in the form of equation (3), and the
values of the coefficients are:

	$\ln p_r^{(0)}$	$\ln p_r^{(1)}$	$\ln p_r^{(2)}$
a	-5.97616	-5.03365	-0.64771
b	1.29874	1.11505	2.41539
c	-0.60394	-5.41217	-4.26979
d	-1.06841	-7.46628	3.25259

Equation (8) allows us to calculate a series of equations with coefficients conforming exactly to a pattern and, except at low reduced temperatures for the lower members of the series, deterioration in the fit to observed values in comparison with the equations in Table 1 is not very great.
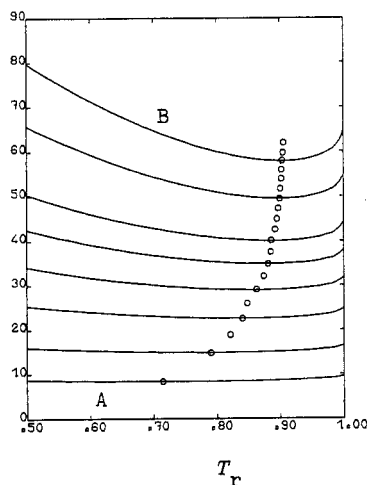

Fig. 6. $(\Delta H/kJ)(\Delta Z/mol)^{-1}$ for methane (A), ethane, butane, hexane, octane, decane, tetradecane, and octadecane (B) with minima for all n-alkanes to eicosane.
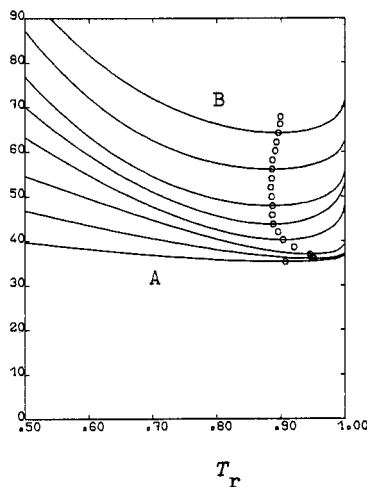
Fig. 7. $(\Delta H/kJ)(\Delta Z/mol)^{-1}$ for methanol (A), ethanol, 1-butanol, 1-hexanol, 1-octanol, 1-decanol, 1-tetradecanol, and 1-octadecanol (B) with minima for all 1-alkanols to eicosanol.

CRITICAL TEMPERATURES AND PRESSURES OF 1-ALKANOLS

The alkanols present a more difficult problem than the alkanes because of their special behaviour and because the data are much fewer. We have not found any incremental scheme satisfactory for correlation of their critical properties, and it seemed best to relate them to those of the alkanes. We assumed that as the chain length increased the effect of the -OH group would become progressively less and that the critical temperatures and critical pressures would converge on those of the alkanes. We calculated the critical temperatures of the higher 1-alkanols from those of the corresponding alkanes T_{alk} by

$$T_c/T_{alk} = 1 + 1.576/n^{1.14}. \tag{9}$$

Equation (9) is not very sensitive to change in the power of n, and that used arises from fitting to values for 1-butanol to 1-decanol (those for the higher members themselves being estimates). Anselme, Rosenthal and Teja (ref. 29) related critical temperatures of 1-alkanols to those of alkanes by the equation,

$$T_c = T_{alk} + 833.0959/(2.015476 + n). \tag{10}$$

Equation (10) gives values that approach those for the alkanes more quickly than do the values from equation (9). For example, for hexadecane the critical temperature is 722 K and for 1-hexadecanol the equation (9) value is 770 K whereas the equation (10) value is 768 K. For very long chains equation (9) converges on but never quite reaches the line for the alkanes while equation (10) gives a line parallel to the alkane line. At present the experimental evidence provides no reason to prefer one equation to the other. The line shown for 1-alkanols in Fig. 2 is derived from equation (9) in conjunction with equation (6).

We related the critical pressures of the 1-alkanols to those of the alkanes P_{alk} by the equation

$$p_c/p_{alk} = 1 + 0.284/n^{0.3},\tag{11}$$

where, again, the power of n was determined by fitting to the experimental observations.

VAPOUR PRESSURES OF 1-ALKANOLS

References to the experimental data to which the equations whose coefficients are in Table 2 were fitted are in ref. 2. Ambrose, Sprake and Townsend (ref. 30) found that the curves for 1-pentanol and 1-octanol differed in shape from those for the propyl and butyl alcohols, and it seems reasonable to suppose that if the critical properties of alkanols converge on those of the alkanes at high carbon number then the shapes of the vapour-pressure curves, though very different for the lower members, should converge as the carbon number

TABLE 2. 1-Alkanols - critical temperatures and pressures and coefficients of equation (3)

alkanol	T_c/K	p_c/MPa	a	b	c	d	ω
methanol	512.64	8.092	-8.63571	1.17982	-2.4790	-1.024	0.564
ethanol	513.92	6.132	-8.68587	1.17831	-4.8762	-1.588	0.643
1-propanol	536.78	5.168	-8.53706	1.96214	-7.6918	-2.945	0.620
1-butanol	563.05	4.424	-8.40615	2.23010	-8.2486	-0.711	0.591
1-pentanol	588.15	3.909	-8.98005	3.91624	-9.9081	-2.191	0.579
1-hexanol	610.7	3.470	-9.49034	5.13288	-10.5817	-5.154	0.575
1-heptanol	632.5	3.135	-9.68778	5.35716	-10.1672	-8.010	0.580
1-octanol	652.5	2.86	-10.01437	5.90629	-10.4026	-9.048	0.594
1-nonanol	671.5	2.63	-9.91542	5.13670	-8.8075	-12.497	0.610
1-decanol	689	2.41	-9.75478	4.18634	-7.0572	-15.980	0.629
1-undecanol	705	2.24	-9.85733	3.97841	-6.6002	-16.691	0.656
1-dodecanol	720	2.08	-9.91901	3.61884	-5.8537	-18.204	0.684
1-tridecanol	734	1.935	-9.99402	3.36986	-5.4865	-18.592	0.712
1-tetradecanol	747	1.81	-10.13519	3.27661	-5.3447	-18.711	0.744
1-pentadecanol	759	1.70	-10.32431	3.32013	-5.4784	-18.263	0.778
1-hexadecanol	770	1.61	-10.54087	3.47260	-6.0770	-15.939	0.892
1-heptadecano	780	1.50	-10.73125	3.55515	-6.3591	-15.696	0.853
1-octadecanol	790	1.44	-10.91637	3.57835	-6.6199	-15.060	0.892
1-nonadecanol	799	1.38	-11.22657	4.03454	-7.7867	-11.970	0.934
1-eiosanol	809	1.30	-11.23154	3.66900	-7.0775	-14.321	0.954

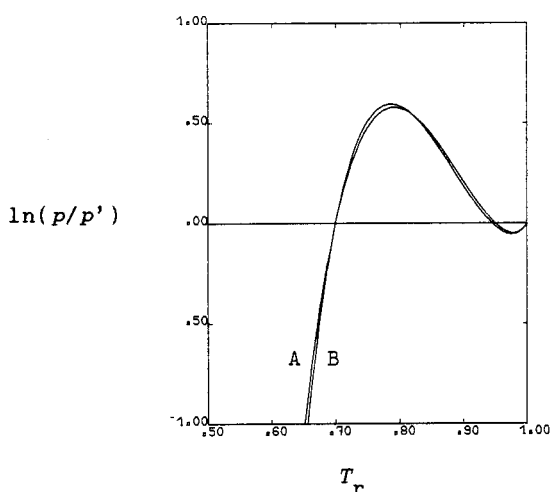$\ln(p/p')$

Fig. 8. Vapour pressures of octadecane (A) and 1-octadecanol (B).

T_r

increases. As a result we do not at first sight discern regularity in the curves or in the coefficients of the equations for the 1-alkanols. However, regularity appears if we differentiate the equations to obtain $\Delta H/\Delta Z$ as in Fig. 7, which includes curves for methanol, ethanol, 1-butanol, 1-hexanol,

1-octanol, 1-decanol, 1-tetradecanol and 1-octadecanol, and the minima for all alkanols up to eicosanol. The position of the minimum, which, except for methanol, is very close to the critical temperature for the lower members, drops back and then rises for the higher members to a value similar to that of the corresponding alkane. The vapour-pressure curves themselves for ethanol to 1-butanol lie entirely above the equation (1) line at reduced temperatures between 0.7 and 1 but, beginning with pentanol, the curves drop below it, the inflection point drops to lower temperature, and the curves approach those of the corresponding alkanes. Figure 8 shows curves for 1-octadecanol and octadecane that are almost identical in the reduced form.

DISCUSSION

After most of the work described here was complete Tsonopoulos published an equation for the critical pressures of alkanes (ref. 23),

$$\ln(p_c/\text{kPa}) = 2.017 - 0.2743n^{2/3}. \tag{12}$$

This presupposes that the critical pressure for an infinite chain length is zero, and for $n = 100$ equation (12) gives a critical pressure 0.02 MPa whereas equation (7) gives a value 10 times as large. Differences between equations (7) and (12) become apparent at carbon numbers greater than 10 and if equation (12) is correct we have chosen critical pressures for the higher members that are too large.

The power 2/3 in equation (12) appears to derive from work by Kurata and Isida (ref. 31), who gave a theoretical justification that the effective length of a carbon chain varied with carbon number in this way. However, the expression given later by Nakanishi, Kurata and Tamura (ref. 32) for alkane critical pressures, again developed theoretically, was more complicated (having more than one term) and their values for the higher members were higher than we are proposing - in fact, their equation does not converge on a zero pressure. Equation (12) with a fitted value for the power 0.5 and coefficients 2.2526 and -0.4716 gives the curve shown for alkanes in Fig. 3. The curve for alkanols is derived from the same equation in conjunction with equation (11). Although the power 2/3 is satisfactory in equation (6) there seems to be no compelling reason for its use in equation (12) because, whatever theoretical reason may be given that the behaviour of long chains depends on the power 2/3, the equation itself has no theoretical basis. Nakanishi *et al.* themselves stated (ref. 32) of one of their equations that the error was always in one direction and that it might therefore be reduced by reducing the power of *n*, and "Such an adjustment .... seems to be natural from the theoretical point of view, for the flexibility of the molecular chain generally increases with *n* and shortens the effective chain length." They also stated that the effect did not become apparent until the chain exceeded 20 carbon atoms, but gave no reason for that choice; this work suggests that the effect becomes apparent at a chain length of 10. Up to that length no significant difference arises whether we use a power 2/3 or 0.5.

We now consider whether any conclusions may be drawn when the vapour-pressure data are combined with the critical properties, and for this purpose the best test substance is hexadecane because it is the highest member for which there are reliable measurements in the atmospheric range, those by Rossini's group (ref. 33). We found there was no difficulty in fitting these with a widely differing range of critical pressures - 1435 kPa, as we have chosen, or 1320 kPa, as given by equation (12) with the power 2/3. The implicit thesis of this paper and earlier work (ref. 14,26) is that the coefficients of equation (3) should fall into a pattern, and that if they do not the underlying data are in error. If the lower value for the critical pressure of hexadecane is used the pattern of coefficients of equation (3) is different from that reported previously for a variety of types of substance in ref. 26 and confirmed and extended in the present work on alkanes; in particular, the second coefficient is less than 1. This is lower than that for any other substance - the minimum value found has been 1.22 for nitrogen, and the range for the other well established non-associated compounds studied is from 1.22 to 2. These substances have values of ω up to 0.4, and there is a general tendency for the coefficient to increase as ω increases. With the critical pressures we have chosen for the higher alkanes ω rises to 0.9 and we find the general tendency is maintained, the second coefficient increasing further - up to 4. We think conformity to the established pattern of coefficients is a good reason for believing that the critical pressure of hexadecane must be

higher than 1320 kPa and, therefore, for preferring our value 1435 kPa and
the values for the other higher alkanes and alkanols calculated by means of
equation (7) and equation (11).

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