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SOME REMARKS ON THE SOLUBILITY OF GASES IN LIQUID LONG-CHAIN *n*-ALKANES AND *n*-ALKAN-1-OLS

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Abstract

The Ostwald coefficients $L_{2,1}$ of He, Ne, Ar, Kr, Xe, N_2, O_2, CH_4, CF_4 and SF_6 dissolved in several homologous *n*-alkanes, *n*-C_lH_{2l+2}, $6\leq l\leq 16$, and *n*-alkan-1-ols, *n*-$C_lH_{2l+1}OH$, $1\leq l\leq 11$, were recently measured at 298.15 K and atmospheric pressure with a modified Ben-Naim/Baer apparatus. Altogether, 201 gas/liquid systems were investigated. For about half of these systems, both pressure control and measurement of the volume of gas dissolved in a given volume of solvent were achieved with a microprocessor. This improved medium-precision instrument ($\sigma\leq 0.5\%$ in most cases) combines easy handling with fully automated data retrieval, and is totally *mercury-free*. In this communication, the salient points of data reduction are presented, with the focus on the estimation of several important auxiliary quantities such as second virial coefficients and partial molar volumes at infinite dilution. The effect of the chain length l of the *n*-alkane or the *n*-alkan-1-ol upon solubility is demonstrated, and some correlations between $L_{2,1}$, or the derived Henry fugacities $H_{2,1}$, and appropriate molecular and/or bulk properties of the gases and the solvents are discussed.

Keywords: gas solubilities, *n*-alkanes, *n*-alkan-1-ols, non-electrolyte solutions, thermodynamics

Introduction

Membranes are perhaps the most ubiquitous cellular structures in living systems. They provide the cell with a permeability barrier and are involved in a large number of cellular functions [1, 2]. The conventional view of biological membranes is essentially based on the fluid-mosaic model of Singer and Nicolson [3]. It envisages the membrane as consisting of a lipid bilayer matrix in which membrane-bound proteins are incorporated, and assumes that the constituent lipids and proteins are mobile by virtue of the fluid nature of the lipid bilayer.

In spite of their considerable functional diversity, the basic structural framework of biological membranes is provided by the lipid bilayer. Lipid bilayers consist of two distinct regions reflecting the structure of lipid amphiphiles [4, 5]:

– The hydrophobic interior, sandwiched between two hydrophilic layers formed by the polar head groups, is fairly homogeneous and consists of hydrocarbon chains. Most biological membrane lipids have two relatively long hydrocarbon chains (usually 16 to 18 carbons per chain), one of which is unsaturated (containing 1 to 3 *cis* double bonds) or branched, thereby ensuring that the membranes are in the fluid state at physiological temperatures.

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– The hydrophilic head-group region may contain charged groups or a wide range of non-ionic polar groups (including zwitterionic compounds), which are in contact with the aqueous surroundings.

The phospholipids (i.e. esters of glycerol phosphate) phosphatidylcholine and phosphatidylethanolamine are the two major lipids encountered in animal membranes, whereas the galactolipids monogalactosyl diglyceride and digalactosyl diglyceride are the major constituents of plant membranes. In some phospholipids, the linkage of the long hydrocarbon chain to the glycerol phosphate group does not involve an ester bond, although the general amphiphile structure is maintained. An important representative of this group is sphingomyelin, which contributes about 18% of the lipid composition of human erythrocyte membranes.

From the above, the continued interest in the solubility of gases in *n*-alkanes (n-C_lH_{2l+2}) and *n*-alkan-1-ols (n-$C_lH_{2l+1}OH$) is hardly surprising [6–12]. In particular, Xe solubilities have attracted considerable attention, because Xe is a prototype inhalational general anaesthetic with a low effective anaesthetic pressure of about 0.8 bar [13] (closely related study areas are decompression sickness and inert-gas narcosis in deep-sea diving).

This communication summarizes briefly our most relevant findings in this field [10, 11]. References [10] and [11] contain the largest set of data on this type of system ever obtained by one group with one instrument: 102 gas/*n*-alkan-1-ol ($1 \le l \le 11$) and 99 gas/*n*-alkane ($6 \le l \le 16$) systems were investigated. Details concerning the experimental method may also be found in Ref. [14]. In the present paper the focus is on a critical discussion of various estimation methods for auxiliary quantities such as second virial coefficients and partial molar volumes at infinite dilution (these quantities are needed to obtain Henry fugacities from measured Ostwald coefficients), and on correlations of solubilities with molecular and/or bulk properties of the solvents and solutes. For a comprehensive discussion of the corresponding thermodynamic formalism, see Wilhelm [15–17].

Experimental

For the solubility measurements, a synthetic method (essentially a flow equilibration method) was used. The instrument is of the Ben-Naim/Baer type recently described in detail [10, 11, 14]. The Mark 2 version incorporates a microprocessor for pressure control and measurement of gas volume. This improved apparatus is for medium-precision work with a minimum design imprecision of about ±0.2%. The actual experimental imprecision does not exceed ±0.5% in most cases. The inaccuracy was shown to be of the same order of magnitude via test measurements of the Ostwald coefficients of selected gases dissolved in water [14]. The instrument's assets are easy handling, relatively fast measurements (<3 h), fully *automated data retrieval* and total *absence of mercury.* The measured Ostwald coefficients may be converted to Henry fugacities by using the thermodynamically rigorous relations given in Refs [10], [11] and [14–18].

The Ostwald coefficient $L_{2,1}$ is a convenient and experimentally directly accessible measure of the solubility of a gas (component 2) in a liquid (component 1).

The thermodynamically exact definition, at experimental temperature T and total pressure P, is

$$L_{2,1}(T,P) = \left(\frac{c_2^{\mathrm{L}}}{c_2^{\mathrm{V}}}\right)_{\mathrm{equil}} \tag{1}$$

where $c_2^{\mathrm{L}}=x_2/V^{\mathrm{L}}$ and $c_2^{\mathrm{V}}=y_2/V^{\mathrm{V}}$ are the concentrations of solute 2 (gas) in the liquid-phase solution (indicated by a superscript L) and in the coexisting vapour-phase solution (indicated by a superscript V), respectively. The mole fractions of the gas in the two phases are x_2 and y_2, and V^{L} and V^{V} are the corresponding molar volumes. The link with experiment is established through

$$L_{2,1}(T,P) = \frac{v^{\mathrm{V}}}{v^{\mathrm{L}}} \tag{2}$$

where v^{V} is the gas volume determined with the gas burettes (and the microprocessor) and v^{L} is the volume of the liquid solution in the equilibration chamber after equilibrium is reached.

Data reduction

As indicated above, the Ostwald coefficient depends on both temperature and pressure. However, for conditions well below the solvent critical point, its pressure dependence is usually very small [16, 17]. If the limiting value of the Ostwald coefficient at the vapour pressure $P_{\mathrm{s},1}$ of the pure solvent is denoted by $L_{2,1}^{\infty}$ (i.e. $\{c_2^{\mathrm{L}}, c_2^{\mathrm{V}}\}\rightarrow 0$; $P\rightarrow P_{\mathrm{s},1}$), the variation in the ratio $L_{2,1}(T,P)/L_{2,1}^{\infty}$ over the pressure range covered by our experiments (less than 0.1 MPa) can be detected only by measurement techniques of the highest accuracy, that is to say by methods where the experimental error is less than 0.1% [18–21]. Thus, given the limits of experimental precision attainable with this apparatus, to an excellent approximation [10, 11, 14] the Henry fugacity $H_{2,1}(T,P_{\mathrm{s},1})$ may be obtained from the experimental Ostwald coefficient according to

$$H_{2,1}(T,P_{\mathrm{s},1}) = \frac{RTZ^{\mathrm{V}}\varphi_2^{\mathrm{V}}I_2^{-1}}{V^{\mathrm{L}}L_{2,1}(T,P)} \tag{3}$$

where the Poynting integral is asymptotically approximated by

$$I_2 \cong \exp[(RT)^{-1}V_2^{\mathrm{L}\infty}(P - P_{\mathrm{s},1})] \tag{4}$$

Here, Z^{V} is the vapour-phase compressibility factor, φ_2^{V} is the fugacity coefficient of the solute in the vapour-phase solution, R is the gas constant, and $V_2^{\mathrm{L}\infty}$ is the partial molar volume of the gas at infinite dilution in the liquid phase.

For gas solubility measurements at such low pressures as used by us ($P\approx 0.1$ MPa), the virial equation in pressure truncated after the second virial coefficient B is adequate and yields

$$Z^{V} = 1 + (RT)^{-1}P(y_1B_{11} + y_2B_{22} + y_1y_2\Delta_{12}) \tag{5}$$

$$\varphi_i^{V} = \exp[(RT)^{-1}P(B_{ii} + y_j^2\Delta_{12})], \quad i, j = 1, 2 \tag{6}$$

where $\Delta_{12}=2B_{12}-(B_{11}+B_{22})$. Here, the virial coefficients of the pure components are B_{11} and B_{22}, respectively, and B_{12} is the second virial cross-coefficient. The molar volume of the dilute liquid solution is approximated by the asymptotically valid expression

$$V^{L} \cong x_1V_{s,1}^{L*} + x_2V_2^{L\infty} \tag{7}$$

where $V_{s,1}^{L*}$ denotes the molar volume of pure liquid solvent at saturation conditions. For computational details of the rapidly converging iteration procedure for the calculation of Henry fugacities $H_{2,1}(T,P_{s,1})$ from measured $L_{2,1}(T,P)$ [10], [11], [14] and [18]. The quantities needed are the vapour pressure of the solvent, the second virial coefficients of the pure components, the second virial cross-coefficient, the molar volume of the pure liquid solvent, and the partial molar volume of the gas at infinite dilution in the liquid phase.

The vapour pressures of the liquid *n*-alkanes and *n*-alkan-1-ols are obtained either from appropriate Antoine equations or, for larger temperature ranges, preferably from Wagner-type equations [22]. Reliable sources also exist for the densities, and hence the molar volumes, of the pure solvents under consideration [10, 11]. However, experimental data on $V_2^{L\infty}$ are not plentiful and thus have to be estimated (in most cases) by dependable and convenient correlations. Similar comments apply to the second virial coefficients: while sufficient information is available on $B22$, experimental values for $B11$ and $B12$ are quite scarce and have to be estimated in most cases.

Virial coefficients

The second virial coefficients B_{22} of the pure gases were taken from the critical compilation of Dymond and Smith [23], while the virial coefficients B_{11} of the pure solvent vapours were estimated by using the generalized corresponding-states correlation of Tsonopoulos [24]:

$$\frac{B_{ii}P_{c,i}}{RT_{c,i}} = B^{(0)}(T_{r,i}) + \omega_iB^{(1)}(T_{r,i}) + B^{(2)}(T_{r,i}) \tag{8}$$

$B^{(0)}(T_{r,i})$ and $B^{(1)}(\mathrm{T}_{r,i})$ are universal functions of the reduced temperature $T_{r,i}=T/T_{c,i}$, $P_{c,i}$ and $T_{c,i}$ are the critical pressure and the critical temperature of pure *i*, respectively, and ω_i is the acentric factor. The polar contribution $B^{(2)}(T_{r,i})$ is generally given by

$$B^{(2)}(T_{r,i}) = \frac{a}{T_{r,i}^6} - \frac{b}{T_{r,i}^8} \tag{9}$$

It is zero for non-polar or slightly polar substances. For non-hydrogen-bonded polar compounds, $b=0$ and a depends on the compound class. For the hydrogen-bonded n-alkan-1-ols, $a=0.0878$, while b appears to be a function of the reduced dipole moment [25]

$$\mu_{r,i} = \left(\frac{\mu_i^2 N_A}{4\pi\varepsilon_o V_{c,i} k_B T_{c,i}}\right)^{1/2} \tag{10}$$

and hence of the chain length l of the alkanol.

Here, μ_i is the permanent electric dipole moment of substance i, $V_{c,i}$ is its critical molar volume, $\varepsilon_o=8.8542\cdot10^{-12}$ F m^{-1} is the permittivity of vacuum, N_A is the Avogadro constant, and k_B is the Boltzmann constant. Using the most reliable data on the second virial coefficients of methanol, ethanol, 1-propanol and 1-butanol, we obtained optimized b values for these four n-alkan-1-ols, and used them as the basis for a generalized correlation of b with l [10]. Approaches similar in spirit were proposed by Hayden and O'Connell [26], and Tarakad and Danner [27]. The critical quantities and the acentric factors of the n-alkanes and n-alkan-1-ols were taken from the most reliable recent critical surveys [28–30].

For the estimation of second virial cross-coefficients, it is assumed that the same corresponding-states relations Eqs (8) and (9) hold, but with suitably defined characteristic parameters $T_{c,12}$, $P_{c,12}$ and ω_{12} replacing the corresponding pure-substance quantities [24]. Thus, reduced virial cross-coefficients $B_{12}P_{c,12}/(RT_{c,12})$ are obtained at a reduced temperature $T_{r,12}=T/T_{c,12}$, with

$$T_{c,12} = (1 - k_{12})(T_{c,1}T_{c,2})^{1/2} \tag{11}$$

For polar/non-polar binaries, B_{12} is assumed to have no polar term, that is to say $a_{12}=0$ and $b_{12}=0$. The critical quantities of the pure gases and their acentric factors were taken from Reid *et al.* [31]. The binary interaction parameter k_{12} is a characteristic constant for each binary.

The most sensitive combination rule is Eq. (11), and the binary interaction parameter k_{12} may be estimated by various semitheoretical correlations [10, 14, 24, 32, 33]. Because of the quite limited set of experimental data on B_{12}, no generally applicable recommendation can as yet be given. However, for different classes of systems good correlations are obtained, e.g. between k_{12} and $T_{c,2}$ [14] and k_{12} and $V_{c,2}$ [10]. For the gas/n-alkane systems, we adopted a Fender-Halsey-type correlation [34].

Partial molar volumes

Recently, experimental partial molar volumes at infinite dilution have been reported for quite a number of gas/n-alkane and gas/n-alkan-1-ol systems [35–39]. With the exception of CO_2 dissolved in n-alkan-1-ols, they are well represented, that is to say usually to within 10%, by

$$\frac{V_2^{L\infty}P_{c,2}}{RT_{c,2}} = 0.088 + \frac{2.763TP_{c,2}}{T_{c,2}\Pi_1^{L*}} \tag{12}$$

where Π_1^{L*} is the internal pressure of the solvent. This quantity may be calculated according to

$$\Pi_1^{L*} = \frac{T\alpha_{P,1}^{L*}}{\beta_{T,1}^{L*}} - P \tag{13}$$

Here, $\alpha_{P,1}^{L*}$ is the isobaric expansivity and $\beta_{T,1}^{L*}$ is the isothermal compressibility of the solvent. However, each gas retains its individuality, and the partial molar volumes of a given gas in any homologous series of solvents could be correlated separately by individual equations of type Eq. (12) with significantly smaller standard deviations. The partial molar volumes of CO_2 in the *n*-alkan-1-ols are of particular note, since they are virtually independent of l [38, 39].

Results and discussion

Altogether, we measured 201 Ostwald coefficients $L_{2,1}(T,P)$ of He, Ne, Ar, Kr, Xe, N_2, O_2, CH_4, CF_4 and SF_6 dissolved either in *n*-alkanes *n*-C_lH_{2l+2}, $6 \le l \le 16$ [11], or *n*-alkan-1-ols *n*-$C_lH_{2l+1}OH$, $1 \le l \le 11$ [10], at T=298.15 K and P= 101325 Pa. A few representative series are contained in Figs 1 and 2: the former depicts the experimental Ostwald coefficients of Xe, O_2, CH_4 and SF_6 dissolved in *n*-alkanes as a function of chain length l, and the latter a similar plot for the same gases dissolved in *n*-alkan-1-ols. In addition to our results, a few supplementary literature data on Xe solubilities [7–9] have been included. Of particular note is the

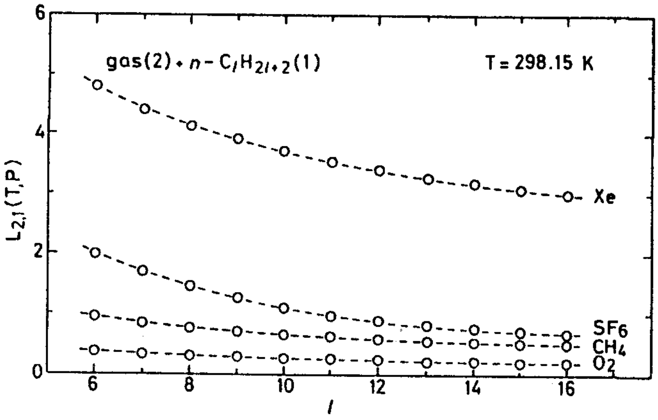

Fig. 1 Ostwald coefficients $L_{2,1}(T,P)$ of Xe, O_2, CH_4 and SF_6 dissolved in *n*-alkanes, *n*-C_lH_{2l+2}, as a function of chain length l for T=298.15 K, P=101 325 Pa, and $6 \le l \le 16$: o, experimental results of Hesse *et al.* [11] and Pollack *et al.* (Xe solubilities) [8]. The broken curves are only meant to indicate the general trends

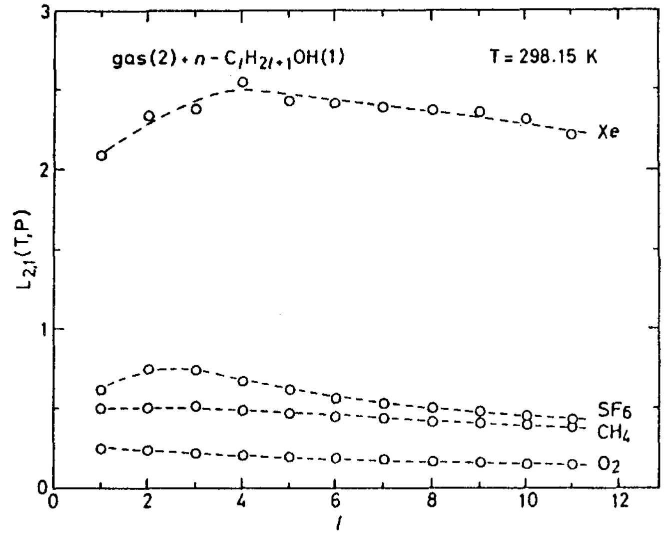

Fig. 2 Ostwald coefficients $L_{2,1}(T,P)$ of Xe, O_2, CH_4 and SF_6 dissolved in *n*-alkan-1-ols, *n*-$C_lH_{2l+1}OH$, as a function of chain length *l* for T=298.15 K, P=101 325 Pa, and $1\le l\le 11$: o, experimental results of Bo *et al.* [10] and Pollack *et al.* (several Xe solubilities) [9]. The broken curves are only meant to indicate the general trends

existence of maxima at small values of *l* for some gas/*n*-alkan-1-ol series ($l\le 4$), in contradistinction to what is observed in the gas/*n*-alkane series, where in all cases $L_{2,1}$ decreases monotonously with increasing *l*. These maxima become more prominent, the larger the solute and/or the solubility of the solute.

The Henry fugacity $H_{2,1}$ is directly related to the reversible work W required to dissolve 1 mol of solute in an infinitely large amount of solvent:

$$\frac{W}{RT} = \ln\frac{H_{2,1}V_{s,1}^{L*}}{RT} \tag{14}$$

This quantity is therefore well suited to serve as a bona fide measure for the average intermolecular interaction between the solute and the solvent. According to Eq. (3) (see also [14–18] and [40]), for the experimental conditions prevailing in this work to a good approximation we have

$$\ln\frac{H_{2,1}V_{s,1}^{L*}}{RT} \approx -\ln L_{2,1} \tag{15}$$

which justifies the use of $-\ln L_{2,1}$ for discussing thermodynamic solvation quantities in many systems of practical interest [9, 41–43]. In Figs 3 and 4, $\ln[H_{2,1}V_{s,1}^{L*}/(RT)]$ is plotted *vs.* the chain length *l* for the four solutes Xe, O_2, CH_4 and SF_6 dissolved in *n*-alkanes and *n*-alkan-1-ols, respectively, at 298.15 K.

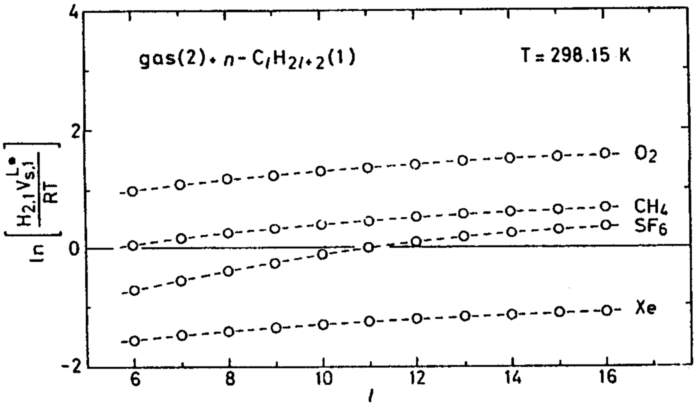

Fig. 3 Correlation of $\ln[H_{2,1}V_{s,1}^{L*}/(RT)]=W/(RT)$ with chain length l for Xe, O_2, CH_4 and SF_6 dissolved in n-alkanes, n-C_lH_{2l+2}, at T=298.15 K, $6 \le l \le 16$. W is the reversible work required to dissolve 1 mol of gas in an infinitely large amount of solvent: o, from experimental results of Hesse *et al.* [11] and Pollack *et al.* (Xe solubilities) [8]. The broken curves are only meant to indicate the general trends

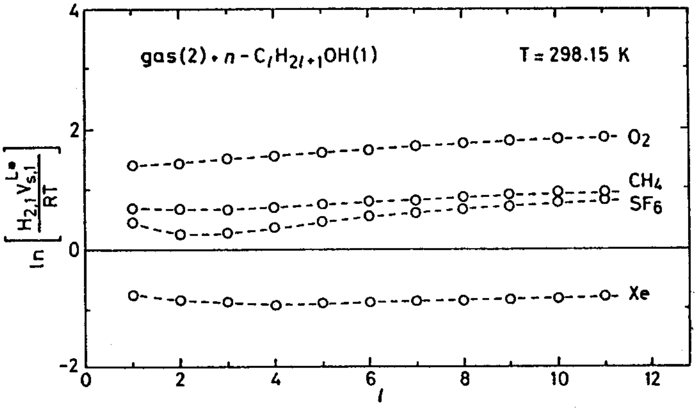

Fig. 4 Correlation of $\ln[H_{2,1}V_{s,1}^{L*}/(RT)]=W/(RT)$ with chain length l for Xe, O_2, CH_4 and SF_6 dissolved in n-alkan-1-ols, n-$C_lH_{2l+1}OH$, at T=298.15 K, $1 \le l \le 11$. W is the reversible work required to dissolve 1 mol of gas in an infinitely large amount of solvent: o, experimental results of Bo *et al.* [10] and Pollack *et al.* (several Xe solubilities) [9]. The broken curves are only meant to indicate the general trends

At this point, some information concerning the magnitude of the various "correction" quantities appearing in Eq. (3) seems indicated. Consider, for instance, the Poynting integral I_2 for gases dissolved in *n*-heptane at 298.15 K: for Xe, I_2=1.0024; for O_2, I_2=1.0018; for CH_4, I_2=1.0021; and for SF_6, I_2=1.0033. Similar values are obtained for the gases in other *n*-alkanes and *n*-alkan-1-ols. The range of the corrections for real-gas behaviour of the vapour phase is indicated by $Z^V\varphi_2^V=1.0013$ for He dissolved in *n*-heptane, and $Z^V\varphi_2^V=0.9748$ for SF_6 dissolved in *n*-heptane. Altogether, the corrections may reach about 3% in the case of SF_6, which far exceeds the minimum design imprecision of our instrument (±0.2%).

For all the gases dissolved in *n*-alkanes, essentially the same trend is observed: $W/(RT)$ increases with increasing l, with the curvature suggesting (as intuitively expected) a limiting value as l becomes very large. A qualitatively appealing interpretation of this behaviour has been given by Ben-Naim and Marcus [43] in terms of a simplified segment interaction model, focusing mainly on energetic considerations, i.e. on the weaker interaction of the solute with the CH_2 groups than with the CH_3 groups.

As can be seen from Fig. 4, the curves $\ln[H_{2,1}V_{s,1}^{L*}/(RT)]$ *vs.* l for the gases dissolved in *n*-alkan-1-ols are markedly different: already the O_2 curve shows an inflection point, and for the series with Xe, CH_4 and SF_6 minima are observed at small values of l. For higher values of l, the curves qualitatively resemble those of the *n*-alkane series. However, as compared to their counterparts in the gas/*n*-alkane systems, all the curves are shifted to more positive values of $W/(RT)$, which reflects the substantially smaller solubility of a gas in an *n*-alkan-1-ol than in an *n*-alkane with the same chain length l. This is quite unexpected if only interaction-energy aspects are considered (see below).

Finally, by inspecting the results given in Figs 3 and 4, we note that the increment

$$\Delta W = W(\mathrm{gas}/n - C_lH_{2l+1}OH) - W(\mathrm{gas}/n - C_lH_{2l+2}) \qquad (16)$$

diminishes rapidly with increasing l. In fact, this is expected, since for the gaseous solute (because of the short-range dispersion forces) only the interactions with the nearest neighbours are significant. Any group further away, such as an OH group in a long-chain alcohol, exerts less and less influence with increasing l. However, such behaviour may be significantly modified if some specific interaction with the OH group takes place. This is apparently the case with CO_2 dissolved in alcohols [44, 45]: when the very few relevant experimental data reported in the literature are used, it appears that $\Delta W/(RT)$ for CO_2 dissolved in *n*-alkanes and *n*-alkan-1-ols is (a) much smaller than for the other gases investigated, and (b) decreases much more weakly with increasing l than for the other gases, thereby lending support to our conjecture. Specifically, at 298.15 K we estimate $\Delta W/(RT)=0.28$ for $l=6$ [46], 0.25 for $l=7$ [46], 0.23 for $l=8$ [45] and 0.21 for $l=10$ [45]. Further corroboration comes from $V_2^{L\infty}$ of CO_2 dissolved in *n*-alkan-1-ols being virtually independent of l [38, 39].

The scaled particle theory (SPT) [15–17, 47–50] has been used successfully to predict and correlate Henry fugacities. For the suggested two-step dissolution process:

$$\ln\frac{H_{2,1}V_{s,1}^{L*}}{RT} = \frac{\mu_{CAV}}{RT} + \frac{\mu_{INT}}{RT} \qquad (17)$$

where μ_{CAV} denotes the free energy of cavity formation and μ_{INT} is the free energy of interaction. SPT provides a reasonable approximation for μ_{CAV} in that it yields an asymptotic expansion in the radius of the (spherical) cavity to be created in a solvent of compactness

$$\eta = \frac{N_A \sigma_1^3 \pi}{6V_{s,1}^{L*}} \tag{18}$$

where N_A is Avogadro's constant and σ_1 is the effective hard sphere diameter of the solvent [51, 52]. The interactional contribution μ_{INT} may, as usual, be approximated by an effective Lennard-Jones term augmented by a term representing dipole-induced dipole interactions. Calculations show that on a molecular level the decisive factor causing a smaller solubility of a given gas in an *n*-alkan-1-ol, as compared to its solubility in an *n*-alkane with the same chain length, is $\eta(n\text{-}C_lH_{2l+1}OH)$ being larger than $\eta(n\text{-}C_lH_{2l+2})$: μ_{CAV} for the *n*-alkan-1-ols is thus considerably more positive than for the *n*-alkanes and is only partly compensated by a somewhat more negative μ_{INT} in the alcohols [44, 45], whence the larger values of $H_{2,1}$ (i.e. the smaller solubilities) in the alcohol series may be rationalized.

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