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Solubilities of Gases in Water and Other Associated Solvents

A method for estimating the effect of hydrogen-bonding on gas solubilities in polar solvents is presented. H-bonding factors, which are based on ideal gas solubilities and solubilities in water, appear to be closely related to H-bonding factors in the simple alcohols. Similarly, H-bonding factors in solvents containing a carbonyl group or dihydroxy groups were related to those in acetone. The relation between the various H-bonding factors can be used to estimate solubilities in these and other associated solvents.

WALTER HAYDUK
and
HARRY LAUDIE

Department of Chemical Engineering
University of Ottawa
Ottawa, Ontario, Canada, K1N 6N5

SCOPE

Equilibrium solubilities of gases in polar, associating solvents are frequently required in the design of absorption equipment. Methods which can be used to predict gas solubilities in nonpolar or regular solutions are not successful in polar, associating solvents such as water or alcohols. A systematic approach has been developed to relate the effect of association or H-bonding on gas solubilities.

Solubilities of 20 gases in water and in six other solvents (when available) were selected from the literature. H-bonding factors were defined for each gas based on the ratio of its actual solubility to its ideal solubility. A large reduction in solubility from the ideal solubility was attributed to strong H-bonds in the solvent. H-bonding factors in one solvent, such as water, were examined to determine if they were related to H-bonding factors in other associating solvents.

CONCLUSIONS AND SIGNIFICANCE

H-bonding factors for gases in water are related to those in the primary, normal alcohols but are essentially independent of H-bonding factors in chlorobenzene, a slightly polar solvent. Similarly, H-bonding factors in acetone are related to those in methyl acetate, acetic acid, and ethylene glycol. Except in those cases where the dissolved gas reacts chemically with the solvent, gas solubilities could be approximately predicted from the relation between H-bonding factors in the solvents, provided the solubility was known in at least one of them. It would appear that this simplistic approach may be used to estimate solubilities in other polar, associating solvents for nonreacting gases. It was also found that as for nonpolar solvents, solubilities of all gases in water tended toward a constant molar concentration at the solvent critical temperature.

Solubilities in water of many gases have been measured. They range in solute mole fraction from about 0.000007 for helium to about 0.3 for ammonia at 25°C. Whereas for nonpolar solvents a considerable degree of success has been achieved in explaining the order of gas solubilities and the variation with temperature, such success has largely eluded workers dealing with aqueous and similar solutions.

An ideal gas solubility can be based on the assumption of an ideal gas phase, an ideal solution, and no Poynting correction required for the vapor pressure (or essentially Raoult's law)

$$x^i_2 = p_2/P^0_2 \tag{1}$$

Ideal solubilities are normally of the correct order of magnitude when compared with experimental values for many nonpolar solvents. Solubilities are, however, not the same for any one gas in all nonpolar solvents as predicted by Equation (1), and there appears to be a dependence on the *solvent internal pressure or solubility parameter* δ. On the other hand, solubilities in water and other polar and associating solvents are usually much lower than ideal solubilities except in a few cases. The reduction in solubility has been attributed to hydrogen-bonding or association (Garrett, 1972) which appears to have the effect of excluding solute molecules. Solubilities above ideal solubilities may be considered to involve some degree of chemical association or solvation, for example, when NH_3 dissolves in water.

For nonpolar and even slightly polar solvents it has been found that solubilities of all gases appear to approach a constant molar concentration as the solvent critical temperature is approached (Hayduk and Buckley, 1971; Hayduk and Castaneda, 1973). This supposition is well supported by experimental data for most solvents although solubilities above the solvent normal boiling points were usually not available. It appears desirable to test this hypothesis in the case of water as solvent, since the solubilities of a number of gases are available well above 100°C and at temperature approaching the critical temperature of water. The review by Himmelblau (1960) is a particularly useful compilation of such data.

The approach taken in this work is that the reduction in solubility in associating solvents from the ideal solubility is directly related to H-bonding or association in the solvent. In this regard, solvents with strong H-bonding tendencies would dissolve less of the same gas than those with weaker H-bonding tendencies. Differences in solubilities among solvents as a result of variations in internal pressure, for this zero order view of the solubility picture, are disregarded. While it is difficult to ascribe a priori H-bonding factors to various polar solvents, it seems entirely possible to compare H-bonding tendencies as reflected by experimental gas solubilities in a number of solvents. It would be expected, for example, that the H-bonding tendency for methanol would result in reductions in solubility of all nonreacting gases in that solvent in some way related to its H-bonding tendency. For this purpose water has been chosen as a suitable highly associated solvent in which the reductions in solubility could be utilized to determine the parameters which could be referred to as H-bonding factors for the various gases

$$\sigma_{\text{solvent}} = x_2/x^i_2 \tag{2}$$

Comparisons between H-bonding factors in water and also in other solvents is then possible. Such comparisons were made for solubilities at 25°C for the solvents methanol, ethanol, butanol, acetone, methyl acetate, acetic acid, ethylene glycol, and chlorobenzene. The last-mentioned solvent is, of course, unlikely to be associated, hence the comparison of solubilities in these solvents with the complete range of H-bonding tendencies should be particularly significant.

Typical solubility behavior in a slightly polar solvent is represented in Figure 1 in which the solubilities of a number of gases in chlorobenzene are shown. Most of the data are as originally reported by Horiuti (1931) including that by Saylor and Battino (1958) and Hayduk and Castaneda (1973). Solubilities in slightly polar solvents have been compared (Hayduk and Castaneda, 1973) with those in nonpolar solvents forming regular solutions. Solubilities in slightly polar solvents were considered to have several characteristics. Slopes of the lines on a log *x*—log *T* diagram were not constant but were usually slightly concave upward when compared with the essentially constant slopes obtained for regular solutions. At the solvent critical temperature the solubilities for all gases tended to a common value which could be estimated from

$$\log_{10} (10^4 x_0) = 2.265 - 0.134\ \delta \tag{3}$$

The above equation, empirically obtained for regular solutions, appeared to describe the reference solubility even in slightly polar solvents. Finally, the gas normal boiling points were useful extrapolations for the solubility data, particularly for highly soluble gases even though these boiling points were often below the solvent freezing point.

Solubilities for 20 gases in water were obtained from literature sources as listed in Table 1. While these solubilities do not represent an exhaustive compilation, they are considered to represent all possible types of solute-solvent molecular interactions with water, with a resulting extremely wide range in actual solubilities. Furthermore, solubilities for many of these same gases are also available at least at 25°C, in the other solvents, methanol, ethanol, butanol, acetone, methyl acetate, acetic acid, ethylene glycol and chlorobenzene, as also listed in Table 1. Solubilities at 25°C of vinyl chloride in the solvents acetone, chlorobenzene, methanol, and ethylene glycol were estimated from data given by Sazhinov (1962) at 0°C. Solubilities in water for He, H_2, N_2, CH_4, and O_2 at elevated pressures and temperatures were those as recommended by Himmelblau (1960). Himmelblau also

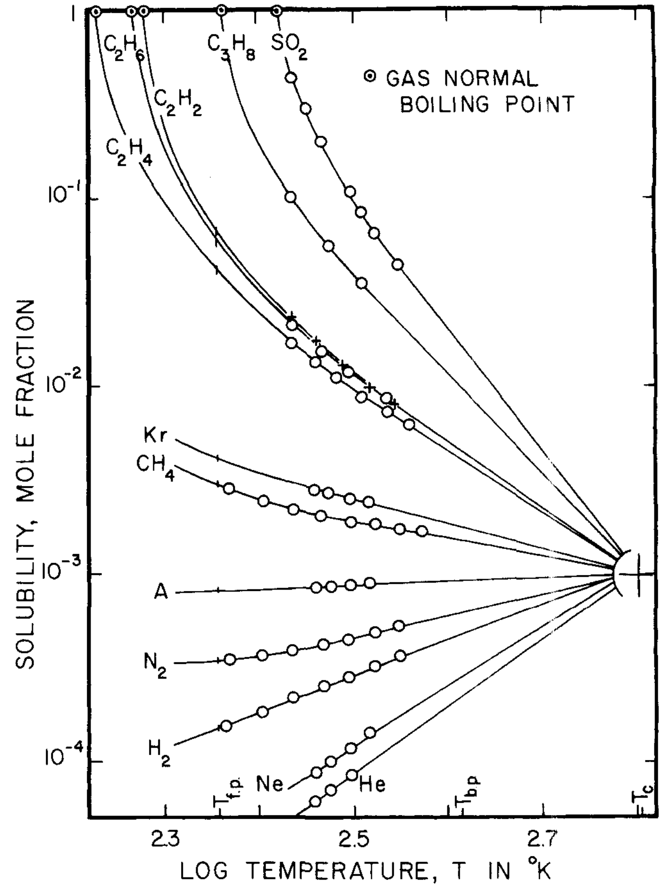

Fig. 1. Solubilities in chlorobenzene.

Table 1. Sources of Solubility Data

Solvent	Gases and source
Water	O_2, N_2, H_2, He, CH_4 (Himmelblau, 1960); NH_3, CO_2, CO, H_2, N_2, N_2O, NO, C_2H_6, C_2H_4 (Perry et al., 1963); Ne, Kr, A (Morrison and Johnstone, 1954); C_4H_{10} (Morrison and Billett, 1952); SO_2 (Seidell and Linke, 1965); C_2H_3Cl (Hayduk and Laudie, 1973).
Methanol	O_2, N_2, H_2, He, Ne, A, N_2O, NO, CO_2, SO_2, NH_3, Kr, CH_4, C_2H_4, C_3H_8, C_4H_{10} (Stephen and Stephen, 1963); C_2H_6 (Gjaldbaek and Niemann, 1958).
Ethanol	O_2, N_2, H_2, He, Ne, A, N_2O, CO_2, SO_2, NH_3, CH_4, C_2H_4, C_3H_8, C_4H_{10} (Stephen and Stephen, 1963); NO (Seidell and Linke, 1965); C_2H_6 (Gjaldbaek and Niemann, 1958).
Butanol	O_2, N_2, A (Stephen and Stephen, 1963); CO, C_2H_6 (Seidell and Linke, 1965); CH_4, C_2H_4 (Boyer and Bircher, 1960); C_3H_8, C_4H_{10} (Hayduk and Castaneda, 1973).
Chlorobenzene	O_2, N_2, N_2O, CO, SO_2, CH_4, C_2H_2, C_2H_4, C_2H_6 (Horiuti, 1931); He, Ne, Kr, A (Saylor and Battino, 1958); CO_2 (Gjaldbaek and Andersen, 1954); C_3H_8 (Hayduk and Castaneda, 1973).
Acetone	O_2, N_2, H_2, N_2O, CH_4, C_2H_2, C_2H_4, C_2H_6 (Horiuti, 1931); A, Ne, He (Stephen and Stephen, 1963); CO, CO_2 (Gjaldbaek and Andersen, 1954); C_3H_8 (Hayduk and Castaneda, 1973).
Methyl Acetate	O_2, N_2, H_2, N_2O, CO, SO_2, CH_4, C_2H_2, C_2H_4, C_2H_6 (Horiuti, 1931); CO_2 (Gjaldbaek and Andersen, 1954).
Acetic Acid	N_2, CO, CO_2, N_2O, Kr, NH_3 (Stephen and Stephen, 1963); O_2, A, C_2H_2, C_2H_6, C_3H_8 (Barton and Hsu, 1972).
Ethylene Glycol	N_2, A, C_2H_6 (Gjaldbaek and Niemann, 1958); CO_2 (Hayduk and Malik, 1971).

reported that for these slightly soluble gases Henry's law could be accurately applied up to pressure of at least 100 atm. For comparison on the same basis, all data for low or moderately soluble gases were converted to mole fraction solubilities for an equilibrium gas partial pressure of 1 atm using Henry's law, regardless of the pressure at which the measurements were actually made. Since it is well known that Henry's law does not apply for aqueous solutions of highly soluble gases, NH_3 for example, it would appear that as the solubility increases increased deviations from Henry's law are observed. Solubilities for NH_3 were reported at several pressures for each temperature. These data were plotted as effective Henry's law constants (x_2/p_2) versus gas partial pressure (p_2) on log—log scales and extrapolated to a partial pressure of 1 atm. A marked deviation from Henry's law was observed especially at lower temperatures.

Gas solubilities in water are shown in Figure 2 as log x versus log T with some of the data omitted to avoid confusion. It is readily apparent that the solubilities are usually much lower in water than in most other solvents, and in some cases such as for paraffin gases, the solubilities are reduced by factors of thousands. It is also apparent that even in this highly H-bonding solvent, the solubilities of a number of the gases tend toward a common solubility at the solvent critical temperature. It will be assumed that the above observation is true for all the gases since solubilities are not actually available at high temperatures for a number of them. In this event minimum solubilities for most gases in water can be expected, except for those few having relatively high solubilities. Hence the increase in solubility at elevated temperatures could be related to diminished tendencies for association with an increase in temperature or weakening of the H-bonds. Further it is cautioned that Equation (2) cannot be used to obtain the reference solubility for highly associated solvents. In water, for example, using a solubility parameter at 25°C of 23.4, a reference solubility can be calculated as 1.35 (10^{-5}) mole fraction compared with the best representation of experimental data of about 1.40 (10^{-4}) which is different by a factor of ten.

While it is possible to calculate or estimate an ideal gas solubility, it has been heretofor usually difficult or impossible to estimate the reduction in solubility as a result of H-bonding or association in the solvent. For this purpose it will be assumed that H-bonding in the solvent itself has the effect of excluding solute molecules and hence of reducing the solubility below what it would be without such molecular interaction. The extent of the reduction will be considered to be a measure of the effect of H-bonding or the H-bonding factor, as defined by Equation (2), for the particular gas-solvent system. Since most extreme behavior is observed for gas solubilities in aqueous solutions, H-bonding factors for water will be initially compared with those in the other solvents. The ideal solubility has been used as a basis for calculating the H-bonding factors although solubilities in a simple nonpolar, non-

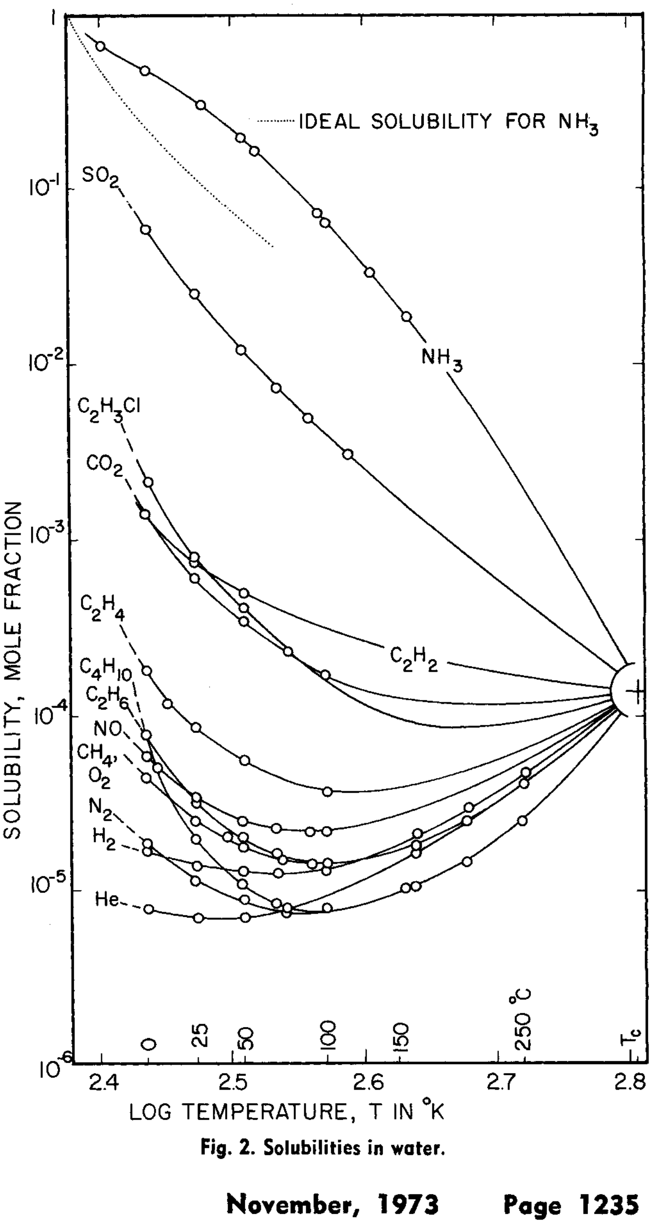

Fig. 2. Solubilities in water.

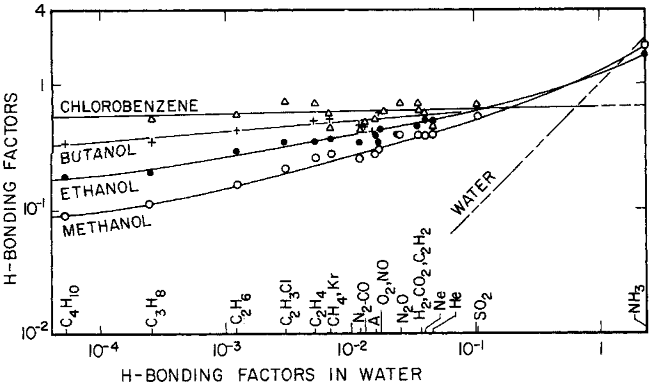

Fig. 3. Comparison of H-bonding factors at 25°C with those in water.

reacting solvent could have been also used. In Table 2 are shown the H-bonding factors as calculated from the actual solubilities, when available, and ideal solubilities at 25°C for all the solvents. If there is a large reduction in solubility of a particular gas in water because of the powerful forces between water molecules, it is expected that a reduction will also occur in other associated solvents. Hence a correlation between H-bonding factors in various associating solvents should exist. Figure 3 shows the relation between H-bonding factors in methanol, ethanol, butanol, and chlorobenzene and those in water. It is apparent that a definite correlation exists between the H-bonding factors in the alcohols and those in water. The effect of H-bonding appears to diminish with an increasing C-content of the alcohols and to essentially disappear for chlorobenzene suggesting, as would be suspected, that no H-bonding occurs in the latter solvent. Further, whereas NH_3 reacts with water yielding a H-bonding factor greater than one, such appears also to be the case in methanol and ethanol solvents. It may be predicted that normal paraffin alcohols containing five carbons and greater would have a diminishing effect on the solubility because of H-bonding in the solvent and that, as in chlorobenzene, the solubilities would roughly approximate simply a constant fraction of the ideal solubility for the various gases. The consistency of the H-bonding factors in the normal paraffin alcohols and water suggests that to better than 25%, gas solubilities can be predicted when experimental data are lacking—for butane in chlorobenzene or vinyl chloride in butanol, for example.

A comparison of H-bonding factors was made with those for water, for the solvents containing the carbonyl group, acetone, methyl acetate and acetic acid, and also for the dihydroxy alcohol, ethylene glycol. This comparison is indicated in Figure 4. While for many of the gases the same simple relation between H-bonding factors as that in the alcohols was observed, for some of the gases significantly higher H-bonding factors were found. Particularly in acetone, the more reactive and oxygen-containing gases, vinyl chloride, acetylene, ethylene, N_2O, CO_2, and SO_2 yielded H-bonding factors nearly equal to or greater than one, suggesting some chemical bonding with this solvent which did not occur with water. Where data were available these same gases gave H-bonding factors greater than those of other gases in the solvents acetic acid and ethylene glycol as well. The suggestion is that in these solvents also, some degree of chemical reaction occurs on solution of the reactive gases. It is noted that the comparable H-bonding factors in methyl acetate very closely parallel those for acetone but are about 10% higher and have been omitted from Figure 4. The conclusion would appear to be that while a correlation between H-bonding factors in water and in ketones, carboxylic acids, and glycols may be possible for nonreactive gases, it is not successful for reactive gases. The two types of solvents, water and monohydric alcohols, and those containing the carbonyl group or dihydroxy groups must be differentiated in solubility behavior. Although it would appear entirely justifiable to simply claim that solubilities in reacting gas-solvent systems are not predictable, some attempt will be made nonetheless. In particular, since acetone, methyl acetate, and acetic acid have a carbonyl group in common, solubility behavior and even the extent of readily reversible chemical reaction may be similar in these solvents. To test this possibility the H-bonding fac-

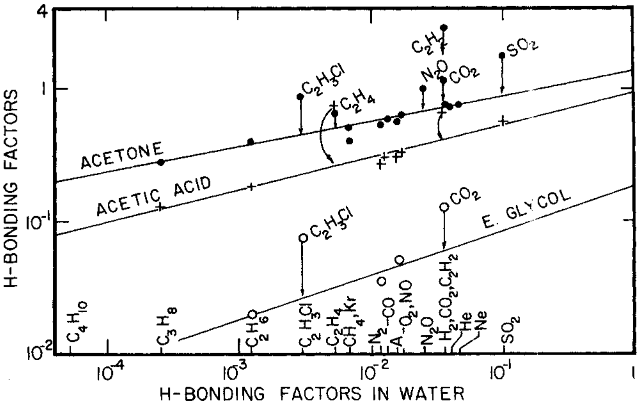

Fig. 4. Comparison of H-bonding factors with those in water when some chemical combination occurs in solution.

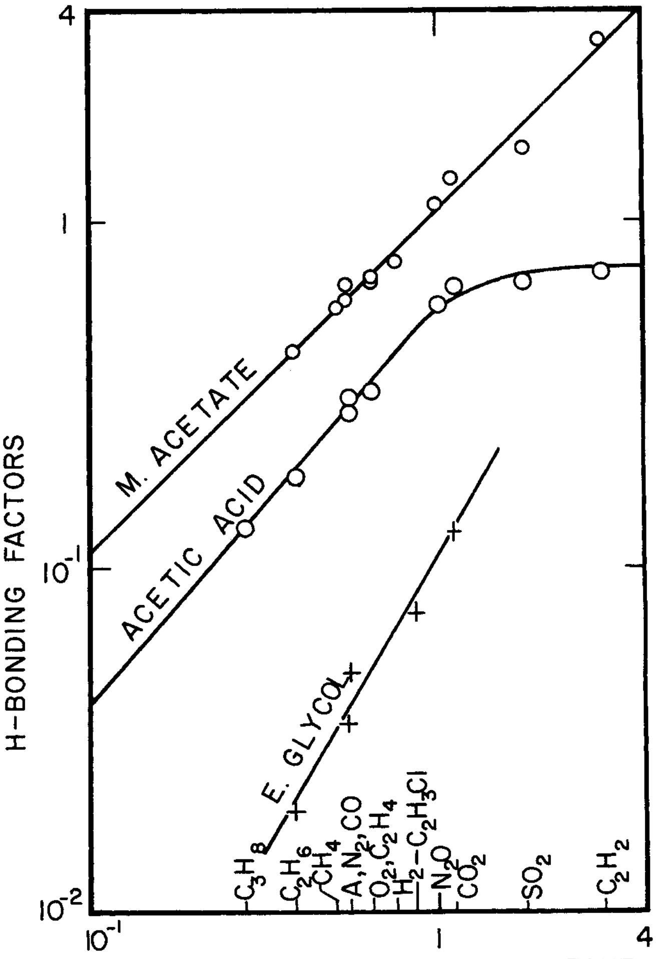

Fig. 5. Comparison of H-bonding factors with those in acetone.

tors in acetone, for which many solubilities are available, were compared with those in methyl acetate, acetic acid, and also ethylene glycol. This comparison is shown in Figure 5. It can be observed that H-bonding factors, including those for the reactive gases, in methyl acetate, acetic acid, and ethylene glycol can be successfully related to those in acetone. In particular, solubilities in methyl acetate and acetone differ only by a small factor. Although H-bonding factors in ethylene glycol are lower than in acetone, suggesting that ethylene glycol is associated to a greater extent than acetone, the effects on solubilities in both solvents appear to be related. That ethylene glycol as well as water was highly associated was observed by Gerrard and Macklen (1959). The H-bonding factors in acetic acid also appear to be related to those in acetone, although not in as simple a manner as those in methyl acetate and ethylene glycol. Whereas SO_2 and acetylene have H-bonding factors greater than one and would seem to react with acetone, using the same criterion these gases do not appear to react with acetic acid. Whether or not other gases which yielded H-bonding factors greater than one in acetone would behave in a manner similar to acetylene in acetic acid would be pure speculation. More data of the type where chemical reactions in the homogeneous liquid solutions are known to occur are required. Nonetheless Figure 5 indicates that a comparison of H-bonding factors in a ketone, ester, carboxylic acid, and a glycol is most useful. For those gases which do not chemically react with the above solvents, solubilities appear to be predictable to within 25% provided that the solubility is known in at least one of them. An extension to other solvents containing a carbonyl group should also be possible.

Measurements of H-bonding tendencies or H-bonding parameters were made by Gordy and Stanford (1941). Shifts in the spectral peaks in deuterated methanol solutions were measured by infrared spectroscopy. The shift from that obtained with benzene was greatest for water followed by glycerol, ethylene glycol, acetic acid, and the simple alcohols. The arbitrary numerical values for the Gordy-Stanford H-bonding parameters (H) are listed in Table 2 for comparison (where available) with the H-bonding factors as determined in this work. The solubilities of N_2 and ethane are available in all nine solvents so that H-bonding factors and the Gordy-Stanford H-bonding parameters can be compared. It is noted that while acetic acid and ethylene glycol have very similar H-bonding indices, solubilities in the two solvents vary by a factor greater than ten. Only poor correlation is observed between the H-bonding parameters and H-bonding factors used in this work.

EXAMPLES

The effectiveness of the method can be illustrated by estimating the solubilities of propane and ethylene in methanol at 25°C and gas partial pressures of 3 atm. From Figure 3 the H-bonding factors in relation to those in water can be estimated from the line for methanol as 0.105, and 0.23, for propane, and ethylene, respectively. The solubilities for a gas partial pressure of 1 atm can be calculated by means of Equation (2) and using ideal solubilities as listed in Table 2. The solubilities are $0.105\ (0.107) = 0.0112$ for propane and $0.23\ (0.017) = 0.0039$ mole fraction for ethylene. These solubilities can be compared with experimentally determined values as calculated from data given in Stephens and Stephens (1963) of 0.0115 (a difference of 3%), and 0.00439 (a difference of 12%) for propane, and ethylene, respectively. Using Henry's law the mole fraction solubilities at 3 atm partial pressure would be three times as great. It is cautioned, however, that the use of Henry's law is liable to significant error when the mole fraction solubility exceeds about 0.05.

Solubilities in associated solvents can be extrapolated to other temperatures if data for a number of other gases in the same solvent, and over a temperature range, are available. A plot showing the relation between log x_2 and log T can be made, as shown for methanol in Figure 6. The fact that a common reference solubility is observed at the critical temperature makes it possible to estimate the temperature coefficient of solubility for any gas. While in methanol nearly linear extrapolations of log x_2-log T appear possible, this is not necessarily true for all other

TABLE 2. HYDROGEN-BONDING FACTORS AT 25°C

Gas	x_2^i	Water	Methan.	Ethan.	Butan.	Chl. benz.	Acetone	Acetic a.	Glycol	M. acet.
C_4H_{10}	0.415(a)	0.0452	0.0915	0.191	0.339	—	—	—	—	—
C_2H_3Cl	0.257(a)	0.0031	0.218	0.350	—	0.740	0.87	—	0.074	—
SO_2	0.250(a)	0.103	0.552	0.615	—	0.720	1.79	—	—	1.63
NH_3	0.130(a)	2.46	2.08	1.77	—	—	—	—	—	—
C_3H_8	0.107(a)	0.0326	0.107	0.198	0.343	0.530	0.282	0.131	—	—
C_2H_6	0.025(b)	0.0012	0.162	0.302	0.435	0.588	0.391	0.184	0.0196	0.424
C_2H_2	0.21(b)	0.0359	—	—	—	0.707	3.02	0.711	—	3.40
N_2O	0.018(a)	0.0247	0.294	0.374	—	0.732	0.99	0.583	—	1.12
C_2H_4	0.017(a)	0.0053	0.264	0.360	0.525	0.723	0.663	—	—	0.691
CO_2	0.017(b)	0.0359	0.376	0.375	—	0.580	1.12	0.660	0.129	1.33
Kr	0.0061(c)	0.0071	0.278	—	—	0.451	—	0.164	—	—
CH_4	0.035(b)	0.0069	0.248	0.365	0.546	0.572	0.506	—	—	0.560
NO	0.0020(a)	0.0175	0.290	0.341	—	—	—	—	—	—
A	0.0016(b)	0.0158	0.278	0.386	0.386	0.533	0.565	0.312	0.0495	—
O_2	0.0013(b)	0.0175	0.292	0.437	0.597	0.599	0.641	0.326	—	0.687
CO	0.0013(b)	0.0134	—	—	0.485	0.492	0.595	0.313	—	0.671
N_2	0.0010(b)	0.0119	0.275	0.356	0.474	0.426	0.550	0.280	0.0349	0.597
H_2	0.0004(c)	0.0355	0.393	0.470	—	0.635	0.760	—	—	0.766
Ne	0.0002(c)	0.0410	0.387	0.520	—	0.494	0.755	—	—	—
He	0.00015(c)	0.0454	0.395	0.509	—	0.465	0.720	—	—	—
H		39.0	18.7	18.7	—	1.5	9.7	20.0	20.6	—

(a) Estimated from Equation (1).
(b) From Hildebrand and Scott (1964).
(c) Estimated from order of solubilities ($H_2 > Ne > He$; $Kr > A$) in nonpolar solvents.

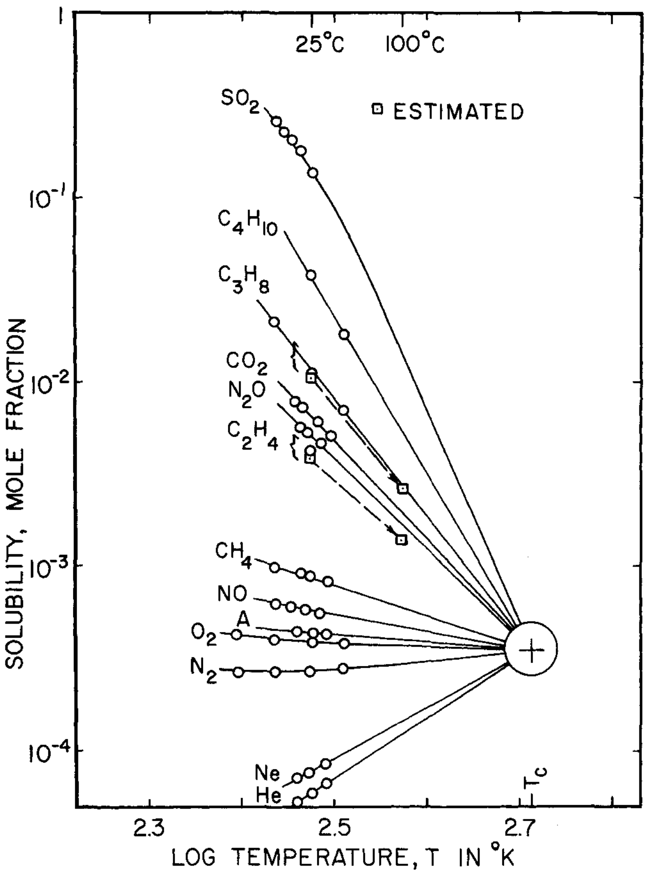

Fig. 6. Solubilities in methanol.

polar or associated solvents. As a further example, propane, and ethylene solubilities at 100°C and corresponding to a gas partial pressure of 1 atm are estimated as 0.0027, and 0.0014 mole fraction, respectively. No comparative experimental values have been found in the literature. Since the solvent is above its normal boiling point, the total pressure for such equilibrium conditions can only exist at pressures above atmospheric.

For application to ketones or glycols, ethylene glycol for example, a comparison of H-bonding factors for several gases with those in acetone would be required. If the solubility at 25°C were known in acetone it would be possible to estimate the solubility at 25°C in ethylene glycol. Extrapolations to other temperatures would again require solubilities of several gases in ethylene glycol over some temperature interval to establish a reference solubility. The accuracy of such a procedure would normally increase with the availability of solubilities for different gases, particularly for ones having solubilities similar to that being predicted.

ACKNOWLEDGMENT

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NOTATION

L = Ostwald coefficient, cm^3 gas/cm^3 solvent
p_2 = partial pressure of gas, atm.
P^0_2 = vapor pressure of pure liquified gas, atm.
V = liquid molar volume, cm^3/gm mole
x_2 = gas solubility, mole fraction in solution
x^i_2 = ideal solubility, mole fraction in solution
σ = solvent H-bonding factor
δ = $\left[\frac{\Delta H_v - RT}{V}\right]^{0.5}$, solubility parameter, (cal/cm^3)$^{0.5}$
ΔH_v = heat of vaporization, cal/gm mole
T = absolute temperature, °K

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