Solubilities of Nonpolar Gases in Triethylene Glycol Dimethyl Ether, Tetraethylene Glycol Dimethyl Ether, Dimethyl Carbonate, and Diethyl Carbonate at 298.15 K and 101.33 kPa Partial Pressure of Gas

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The solubilities of 12 nonpolar gases (He, Ne, Ar, Kr, Xe, H_2, N_2, CH_4, C_2H_4, C_2H_6, CF_4, and SF_6) in the liquids triethylene glycol dimethyl ether, tetraethylene glycol dimethyl ether, dimethyl carbonate, and diethyl carbonate have been measured at the temperature 298.15 K and 101.33 kPa partial pressure of gas using a saturation method. The solubilities have been expressed as mole fractions of gas dissolved and also as Henry's constants. The scaled particle theory has been applied to the results in order to obtain the distance and energy parameters of the Lennard-Jones potential for the liquids. The ability of this theory to correlate the solubilities is discussed.

Introduction

The substitution of different compounds, mainly hydrofluorocarbons (HFCs), for the environmentally harmful chlorofluorocarbons (CFCs) in refrigeration systems involves a series of changes in the devices to adapt them to the properties of the new refrigerants. One of these changes is that of the lubricants in the compression systems. Many liquids have been tested for this purpose, among them several poly(ethylene glycol) dialkyl ethers[1] and carbonates,[2] one of the aspects studied being the solubility of the refrigerant HFC134a in these compounds.[3−6]

In this paper, the solubilities of a wide set of nonpolar gases (He, Ne, Ar, Kr, Xe, H_2, N_2, CH_4, C_2H_4, C_2H_6, CF_4, and SF_6) in four of those liquids at the temperature 298.15 K and 101.33 kPa partial pressure of gas are reported. Specifically, the liquids are two complex ethers, namely, triethylene glycol dimethyl ether (TriEGDME) and tetraethylene glycol dimethyl ether (TEGDME), and two carbonates, dimethyl carbonate and diethyl carbonate (DMC and DEC, respectively).

In addition, the aim of the work is to obtain information about the intermolecular forces acting in these liquids, including the Lennard-Jones parameters for the liquid solvents. For this purpose, the correlation of the solubility data by means of the scaled particle theory[7,8] has been used. The solubilities given by the theory using these parameters have been calculated and compared with the experimental ones.

Experimental Section

Materials. All the gases were provided by Air Liquide except neon that was obtained from J.T. Baker and krypton that was purchased from Linde. The purities of the gases

in mole percent were as follows: He, 99.995; Ne, 99.9; Ar, 99.999; Kr, 99.95; Xe, 99.995; H_2, 99.99; N_2, 99.998; CH_4, 99.95; C_2H_4, 99.0; C_2H_6, 99.90; CF_4, 99.999; and SF_6, 99.5.

Referring to the liquids, TriEGDME ($CH_3O(CH_2CH_2O)_3CH_3$) and TEGDME ($CH_3O(CH_2CH_2O)_4CH_3$) were obtained from Aldrich with purities of at least 99 mol %, whereas DMC (($CH_3O)_2CO$) and DEC (($CH_3CH_2O)_2CO$) were Fluka products with purities of at least 99 and 99.5 mol %, respectively.

Gases and liquids were used without further purification although the liquids were stored over an activated molecular sieve (Merck) of 3 Å pore diameter in order to avoid the absorption of water.

Apparatus and Procedure. The apparatus and procedure used to measure the solubility of gases have been described in detail elsewhere.[9,10] The equipment is similar to that of Ben-Naim and Baer.[11] Its main parts are a solution vessel, a mercury manometer, and a buret system. The solution vessel containing a known amount of liquid solvent is immersed in a water bath whose temperature is controlled within ±0.05 K. The vessel is connected through a capillary to the burets and the manometer that are both filled with gas. The whole apparatus is located in an air bath thermostated within ±0.2 K. The temperature of this air bath is higher than that of the water bath to prevent condensation of the vapor solvent out of the solution vessel.

The experimental method is a saturation one based on the determination of the volume of gas phase dissolved in a known mass or volume of a degassed liquid. The gas phase is a wet gas, that is, the gas mixed with vapor of the solvent. The gas dissolved into the liquid phase is replaced by mercury introduced in the graduated burets in such a way that the pressure of the gas phase, controlled through the mercury manometer, is kept constant during the measuring process. The volume of mercury introduced, that is, the volume of wet gas dissolved, is measured with an uncertainty of ±0.01 cm³. The pressure is determined with an uncertainty of ±3 Pa by using a cathetometer (Wild KM 338).

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In the case of the carbonates, the amount of liquid in the solution vessel was determined by adjusting the level of liquid to a fixed mark corresponding to a previously calibrated volume. The adjustment is carried out by removing liquid (which is introduced in a slight excess) with the aid of a vacuum pump. For the ethers, a weighing method was used because of their very small vapor pressure.

Results and Discussion

The solubility values were calculated with a reduction method[12] that is a modification of that proposed by Wilhelm and Battino.[13-15] The modification takes into account two features of our apparatus: (i) the difference between the temperatures of the liquid phase in the solution vessel and the gas phase in the burets and (ii) the fact that the gas dissolved is a wet one.

The method considers that the volume of wet gas transferred, V, contains a total number of moles, n_T, which is the sum of n_2 moles of solute (that are consequently dissolved in the solution vessel) and n'_1 moles of vapor. An iteration procedure involving the following equations is used to determine the value of n_2.

$$n_2 = n_T - n'_1 = \frac{P'V}{RT' + B_m(y_2, T')P'} - \frac{P_{s,1}(T)V}{RT' + B_{11}(T')P_{s,1}(T)} \quad (1)$$

$$y_2 = \frac{n_2}{n'_1 + n_2} \quad (2)$$

P' is the total pressure of the gas phase, R is the gas constant, T is the temperature of the liquid in the solution vessel, T' is the temperature of the gas phase in the burets, $P_{s,1}(T)$ is the saturated vapor pressure of the solvent at temperature T, $B_m(y_2, T')$ is the second virial coefficient of the wet gas phase at temperature T', $B_{11}(T')$ is the second virial coefficient of the pure solvent vapor at T', and y_2 is the mole fraction of the gas solute in the gas phase. An ideal behavior of the gas phase is assumed in the first step.

When the mole number of the solute has been determined, the mole fraction of gas dissolved is calculated at the total pressure through

$$x_2(P') = \frac{n_2}{n_1 + n'_1 + n_2} \quad (3)$$

where n_1 is the mole number of the solvent in the solution vessel calculated from either the mass (polyethers) or the volume (carbonates).

To obtain the Henry's constant at the vapor pressure of the solvent and the mole fraction at 101.33 kPa partial pressure of gas, the equilibrium equations

$$y_1 P \phi_1^G(T, P, y_2) = x_1 \gamma_1(T, P, x_2) P_{s,1}(T) \phi_{s,1}^*(T, P_{s,1}) \mathscr{P}_1(T, P) \quad (4)$$

$$y_2 P \phi_2^G(T, P, y_2) = x_2 \gamma'_2(T, P, x_2) H_{2,1}(T, P_{s,1}) \mathscr{P}_2(T, P) \quad (5)$$

are used. In these expressions, y_i denotes the mole fraction of component i in the gas phase, x_i is the mole fraction of component i in the liquid phase, ϕ_i^G is the fugacity coefficient of component i in the gas phase, $\phi_{s,1}^*$ is the fugacity coefficient of the pure solvent at the saturated vapor pressure, γ_1 is the activity coefficient of the solvent respect

Table 1. Densities, ρ, Vapor Pressures, P^{sat}, and Dipole Moments, μ, of the Liquid Solvents

liquid	ρ/kg·m^{-3}	P^{sat}/kPa	μ/D
TriEGDME	980.2[a]		2.16[e]
TEGDME	1006.2[a]		2.44[e]
DMC	1059.1[b]	7.19[c]	0.86[f]
DEC	975.3[b]	1.53[d]	0.88[f]

[a] Reference 17. [b] Reference 18. [c] Reference 21. [d] Reference 22. [e] In hexane; ref 29. [f] In carbon tetrachloride; ref 30.

to the Lewis−Randall rule, γ'_2 is the activity coefficient of the solute with respect to Henry's law, $H_{2,1}$ is the Henry's constant for the solution of 2 in 1, and $\mathscr{P}_i$ is the Poynting correction for component i. Now P is the total pressure of the system but considered as a variable.

Given the complexity of the equilibrium equations, some simplifications are assumed to carry out a mentioned trial and error procedure: (i) the liquid-phase activity coefficients are made equal to unity; (ii) the fugacity coefficients are estimated from a virial equation truncated after the second term;[14,15] (iii) the partial molar volumes in the Poynting corrections[14,15] are substituted by the partial molar volumes at infinite dilution for the dissolved gases, while the molar volumes of the pure compounds were introduced for the solvents.

The Henry's constant at the vapor pressure of the solvent, $H_{2,1}(T, P_{s,1})$, can be found directly from eq 5. To calculate the mole fraction at 101.33 kPa partial pressure of gas, it is necessary to estimate the corresponding partial pressure of solvent vapor, Py_1. Both quantities are determined simultaneously with an iterative method based on the following equations deriving from eqs 4 and 5:

$$Py_1 = (1 - x_2) \frac{P_{s,1}(T, P) \phi_{s,1}^*(T, P_{s,1})}{\phi_1^G(T, P, y_2)} \mathscr{P}_1(T, P) \quad (6)$$

$$y_2 = \frac{101.33}{P} \quad (7)$$

$$x_2 = \frac{101.33 \phi_2^G(T, P, y_2)}{H_{2,1}(T, P)} \quad (8)$$

where

$$H_{2,1}(T, P) = H_{2,1}(T, P_{s,1}) \mathscr{P}_2(T, P) \quad (9)$$

The iteration starts with

$$Py_1 = P_{s,1}(T, P)(1 - x_2) \quad (10)$$

where x_2 is given by eq 3.

To apply the described method, the densities of the ethers[16] and the carbonates[17] and also the vapor pressures of ethers and carbonates were found in the literature.[18-21] In fact, the vapor pressures of the ethers are so low at 298.15 K that they have been considered negligible. The values of the densities for the four liquids and those of vapor pressures for the carbonates at 298.15 K have been gathered in Table 1. The second virial coefficients for the gases were extracted from the TRC Tables[22,23] except those of helium, neon, and hydrogen that were interpolated from the values provided by Dymond and Smith.[24] The second virial coefficients for the vapor of the liquids and the cross second virial coefficients were estimated through the method proposed by Pitzer and Curl and modified by Tsonopoulos.[25] The dipole moments of the liquids are necessary in this method. They were found in the literature

for both the ethers[26] and the carbonates[27] and have been included in Table 1. The molar volumes at infinite dilution for the gases needed in the Poynting correction $\mathscr{P}_2$ were obtained from the literature[28] except for the gases He, Ne, Xe, CF$_4$, and SF$_6$. For He and Ne, the value of hydrogen provided by Prausnitz and Shair[26] was considered suitable. The partial molar volumes at infinite dilution for Xe, CF$_4$, and SF$_6$ were calculated[29] from their solubilities in benzene by means of a method described by Ng and Walkley.[30] Their respective values are 5.7×10^{-5}, 8.3×10^{-4}, and 9.7×10^{-4} m$^3 \cdot$mol^{-1}. All these values of molar volume are approximated, but the influence of the Poynting correction on the final results is almost negligible.

The uncertainty in both the mole fraction and the Henry's constant so obtained is estimated to be less than $\pm 1\%$ in the most unfavorable case. Recently,[31] the solubilities of argon in water have been determined at temperatures near 298.15 K and then compared with the most reliable values in the literature, which are those provided by Krause and Benson.[32] The Henry's constants obtained were 3659.3 MPa at 292.85 K, 4030.4 MPa at 298.20 K, and 4160.1 MPa at 300.15 K, while the values of Krause and Benson are 3656.3, 4007.3, and 4132.2 MPa, respectively. The deviations are in all cases below the estimated uncertainty.

The values of the solubilities of the gases in the ethers and carbonates are listed in Table 2. The solubilities of the gases increase according to the following series for almost the solvents:

$$He < Ne < H_2 < N_2 < CF_4 < Ar < SF_6 < CH_4 < Kr <$$
$$Xe < C_2H_6 < C_2H_4$$
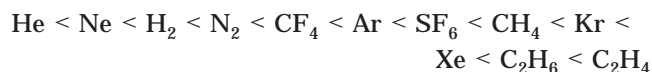
There are exceptions in the cases of fluorinated gases. Specifically, carbon tetrafluoride is less soluble than nitrogen in tetraethylene glycol dimethyl ether and sulfur hexafluoride is more soluble than methane and krypton in both carbonates.

The solubility of fluorinated gases is specially low in the ethers. Moreover, the solubility of these solutes decreases when passing from the triethylene compound to the tetraethylene one, which is an anomalous behavior. Commonly, given a series of compounds with the same functional group, the greater the size of a molecule, the greater the solubility,[33] a trend that is observed for the remaining gases in the ethers and for all of the gases in the carbonates. In fact, the solubility of the gases is considerably higher in diethyl carbonate than in dimethyl carbonate.

The behavior observed for the solubility of CF$_4$ and SF$_6$ in the ethers could be explained considering that there are repulsive interactions between fluorine atoms of the gases and oxygen atoms of the liquids, both of them being very electronegative and then carrying negative partial charges. The additional CH$_2$CH$_2$O group in tetraethylene glycol with respect to triethylene glycol would generate a more repulsive medium for the fluorinated gases, yielding lower solubility values.

A relevant characteristic of the solubilities of gases in liquids is that they can be used to obtain information about the intermolecular forces acting in the solvent through the application of the scaled particle theory (SPT) proposed by Pierotti.[7,8] This theory allows the determination of the parameters of distance, σ_1, and energy, ϵ_1/k, for an additive Lennard-Jones potential which accounts for the dispersive and repulsive forces between the liquid solvent molecules. The SPT divides the solution process in two steps. The first is the creation of a cavity in the solvent great enough to

Table 2. Solubilities Expressed as Mole Fractions, x_2, and Henry's Constants, $H_{2,1}$, Along with the Solubilities, x_2^{SPT}, Obtained from the Scaled Particle Theory

gas	$10^4 x_2$	$H_{2,1}$/bar	$10^4 x_2^{SPT}$	$[(x_2 - x_2^{SPT})/x_2] \times 100$
Triethylene Glycol Dimethyl Ether
He	1.484	6281	1.967	−33
Ne	1.582	6400	5.742	−263
Ar	10.95	922.7	23.81	−118
Kr	31.75	317.6	44.71	−41
Xe	128.4	77.83	87.73	32
H$_2$	3.525	2876	4.044	−15
N$_2$	5.574	1815	8.301	−49
CH$_4$	24.36	414.4	34.29	−41
C$_2$H$_4$	158.5	63.28	104.1	34
C$_2$H$_6$	140.7	71.41	95.59	32
CF$_4$	5.696	1766	6.489	−14
SF$_6$	23.14	431.2	15.80	32
				av deviation[a], 40%
Tetraethylene Glycol Dimethyl Ether
He	1.117	9062	1.738	−56
Ne	1.582	6400	5.692	−260
Ar	11.50	878.5	26.40	−130
Kr	33.71	299.2	51.25	−52
Xe	136.5	73.21	103.2	24
H$_2$	3.726	2717	3.869	−4
N$_2$	5.719	1769	8.130	−42
CH$_4$	25.48	396.2	38.31	−50
C$_2$H$_4$	167.7	59.81	121.1	28
C$_2$H$_6$	144.4	69.57	104.9	27
CF$_4$	5.215	1929	5.461	−5
SF$_6$	21.84	456.9	11.71	46
				av deviation,[a] 42%
Dimethyl Carbonate
He	1.001	10119	1.294	−29
Ne	1.264	8011	3.321	−163
Ar	8.042	1257	13.26	−65
Kr	20.77	485.5	25.97	−25
Xe	72.27	138.2	55.83	23
H$_2$	2.417	4180	2.441	−1
N$_2$	4.960	2040	5.340	−8
CH$_4$	16.04	629.4	20.46	−28
C$_2$H$_4$	98.44	101.9	69.14	30
C$_2$H$_6$	83.88	119.7	75.61	10
CF$_4$	6.871	1464	6.153	11
SF$_6$	29.25	340.9	27.01	8
				av deviation,[a] 22%
Diethyl Carbonate
He	1.560	6489	2.097	−34
Ne	2.087	4851	5.608	−169
Ar	13.38	755.1	23.48	−76
Kr	36.80	274.1	45.84	−25
Xe	131.2	76.15	96.54	26
H$_2$	3.898	2598	4.126	−6
N$_2$	7.657	1321	9.541	−25
CH$_4$	27.29	369.8	36.13	−32
C$_2$H$_4$	157.3	63.78	118.6	25
C$_2$H$_6$	155.9	64.43	126.3	19
CF$_4$	10.72	938.3	10.83	−1
SF$_6$	51.89	192.2	41.43	20
				av deviation,[a] 26%

[a] Neon excluded.

accommodate a solute molecule. The second implies the insertion of the solute molecule and interaction between it and the surrounding solvent. The changes in the partial Gibbs energy for those steps are connected to the Henry's constant through the expression

$$\ln H_{2,1} = \frac{G_c}{RT} + \frac{G_i}{RT} + \ln\left(\frac{RT}{V_{m,1}}\right) \qquad (11)$$

where G_c is the change in the partial Gibbs energy for the creation of the cavity, G_i is the change in the Gibbs energy for the interaction step, and $V_{m,1}$ is the molar volume of the liquid.

According to SPT,[34,35] G_c at pressures near 101 kPa is given by the equation

$$G_c = RT\left\{\frac{65y}{1-y}\left[2\left(\frac{a_{12}}{a_1}\right)^2 - \frac{a_{12}}{a_1}\right] + 18\left(\frac{y}{1-y}\right)\left[\left(\frac{a_{12}}{a_1}\right)^2 - \frac{a_{12}}{a_1} + \frac{1}{4}\right] - \ln(1-y)\right\} \quad (12)$$

where a_1 is the hard-sphere diameter of the solvent molecule, a_{12} is the arithmetic mean of the hard-sphere diameters of the solvent and solute molecules, and y is the compactness factor defined by

$$y = \frac{\pi a_1^3 N_A}{6 V_{m,1}} \quad (13)$$

where N_A is Avogadro's number.

On the other hand, G_i for a system formed by a nonpolar solute and a polar solvent can be calculated[7,8] through

$$G_i = -3.555\pi\rho_1 R\sigma_{12}^3\left(\frac{\epsilon_{12}}{k}\right) - \frac{1.33\pi\rho N_A\mu_1^2\alpha_2}{\sigma_{12}^3} \quad (14)$$

where ρ_1 is the density of the solvent; σ_{12} is the arithmetic mean of the distance parameters of solvent and solute, σ_1 and σ_2, for the Lennard-Jones potential; ϵ_{12} is the geometric mean of the energy parameters of solvent and solute, ϵ_1 and ϵ_2, for the Lennard-Jones potential; μ_1 is the dipole moment of the solvent; and α_2 is the polarizability of the gas solute. In the SPT, the distance parameters, σ_i, are equated to the hard-sphere diameters, a_i. The first term on the right-hand side accounts for the dispersive and repulsive forces, whereas the second one considers the dipole–induced dipole interactions.

First of all, the distance parameter, σ_1, is calculated, taking into account that G_i becomes zero when the polarizability of the solute approaches zero. Then

$$\lim_{\alpha_2 \to 0} (\ln H_{2,1}) = \frac{G_c}{RT} + \ln\left(\frac{RT}{V_{m,1}}\right) \quad (15)$$

The experimental $\ln H_{2,1}$ data for the noble gases are fitted to a series of powers of α_2 and extrapolated to polarizability zero. Thus, σ_1 ($=a_1$) is found substituting G_c in eq 15 by the expression in eq 12.

Once the distance parameter is known, the energy parameter can be calculated by means of the following method. Substituting eq 14 in eq 11, operating, and rearranging, one obtains this expression:

$$RT\ln H_{2,1} - G_c - RT\ln\left(\frac{RT}{V_{m,1}}\right) + \frac{1.33\pi\rho N_A\mu_1^2\alpha_2}{\sigma_{12}^3} = -3.555\pi\rho R\sigma_{12}^3\left(\frac{\epsilon_1}{k}\right)^{1/2}\left(\frac{\epsilon_2}{k}\right)^{1/2} = X\left(\frac{\epsilon_1}{k}\right)^{1/2} \quad (16)$$

There is a linear relationship between the left-hand side and X, its slope being the square root of the energy parameter of the solvent, ϵ_1/k. This slope can be obtained by correlating the left-hand side values with the X values for all the gases. The parameters of the Lennard-Jones potential and the polarizabilities for the gases required for these calculations can be found in an earlier paper.[10]

The values of the parameters for the Lennard-Jones potential are gathered in Table 3. As can be expected, the position and depth of the potential well (determined by σ_1

Table 3. Parameters of the Lennard-Jones Potential Estimated from the Solubility Data Using the Scaled Particle Theory

parameter	TriEGDME	TEGDME	DMC	DEC
σ_1/Å	7.00	7.62	5.11	5.86
(ϵ_1/k)/K	721	897	452	553

and ϵ_1/k, respectively) increase with the size of the molecules. For the ethers, the parameter of distance is especially high, but this is not a strange result if the great size of the molecules is borne in mind.

Once the parameters have been calculated, the equations of SPT can be reversed to estimate the solubilities and in the last term to evaluate the performance of the model. In the last columns of Table 2, the solubilities expressed as mole fractions obtained from this calculation, x_2^{SPT}, are listed along with the percentage deviations for each gas. The greatest deviations correspond as usual to the gas neon.[36] Excluding this gas, the average deviations are about 40% for the ethers and 20−25% for the carbonates. These last values are within the common results of the model,[37] but those of ethers are clearly higher. Probably this is due to the fact that the SPT reduces the real molecule of liquid to an equivalent sphere of radius σ. But the ether molecules are far from being spherical, and the model is quite stretched when applied to these liquids. Nevertheless, the SPT, despite its limitations in correlating the solubility values, is able to reproduce the lesser solubility of fluorinated gases in TEGDME than in TriEGDME, showing in this way the usefulness of the parameters deduced, whatever the caution with which they must be employed.

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