Solubilities of Nonpolar Gases in Dimethyl Carbonate and Diethyl Carbonate

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This paper reports the mole fraction solubilities of nine nonpolar gases (He, Ne, Ar, Kr, Xe, CH_4, C_2H_4, C_2H_6, and SF_6) in dimethyl carbonate and diethyl carbonate at 278.15 K and 288.15 K at a partial pressure of gas equal to 101.33 kPa. The solubility values were fitted as a linear function of the temperature. Changes in the thermodynamic functions of solution standard (Gibbs energy, enthalpy, and entropy) have been calculated from the solubilities. Finally, these results have been compared with those correlated by the scaled particle theory (SPT).

Introduction

Alkyl carbonates attracted attention as very promising fluids in several fields, for example, electrochemistry[1] (cells), organic synthesis[2] (green methylating reagents), and lubricants.[3] Recently, the solubilities of gases in alkyl carbonates have been investigated from an applied point of view, that is, to find new working pairs in refrigeration[4] or to determine the total amount of carbon dioxide generated in rechargeable lithium cells.[1]

As for the solubilities of gases in liquids, several benefits could be expected, such as characterization of the solvent in which gas dissolves, design of industrial processes, study of intermolecular forces, and physicochemical interpretation of the behavior of matter. Accordingly, we would like to remark here on the relevance of these solubility data. The difficulty involved in modeling gas−liquid equilibrium especially at low pressures is well-known. One of the problems is the lack of an adequate set of data which would allow verification of the reliability of the models. On one hand, the quality required for modeling purposes in these conditions is very difficult to obtain; on the other hand, the experimental determination of gas solubilities is problematic. At present, although the number of papers per year on experimental solubility has increased by 4 times, the experimental work in this field seems to be less “fashionable” than the theoretical work. It is paradoxical when gas−liquid equilibrium at low pressure is an acid test of any theoretical or semiempirical model. Moreover, the experimental data are also relevant for building some models and obtaining parameters that, unfortunately, are sometimes not as accessible (for nonlucrative scientific research) as the experimental data themselves.

Here, we present the mole fraction solubilities of nine nonpolar gases (He, Ne, Ar, Kr, Xe, CH_4, C_2H_4, C_2H_6, and SF_6) in two alkyl carbonates measured at 278.15 K and 288.15 K with the partial pressure of gas being 101.33 kPa. The nonideal behavior of the gas phase has been accounted for. Using these data together with those previously obtained[5] at 298.15 K, the influence of temperature on solubility has been also established. Moreover, all of these data and their variation with temperature have permitted the calculation of the standard changes of Gibbs energy ($\Delta\bar{G}^{\circ}_{m,2}$), enthalpy ($\Delta\bar{H}^{\circ}_{m,2}$), and entropy ($\Delta\bar{S}^{\circ}_{m,2}$) for the solution process.

The scaled particle theory (SPT), which provides expressions for the values of the changes of thermodynamic functions in the solution process, has also been applied in order to characterize the interactions between the solvent particles.

Experimental Section

Materials. All the gases were provided by Air Liquide except neon and krypton that were 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; CH_4, 99.95; C_2H_4, 99.90; C_2H_6, 99.0; and SF_6, 99.5.

Dimethyl carbonate ($(CH_3O)_2CO$) and diethyl carbonate ($(CH_3CH_2O)_2CO$) were Fluka products with purities of at least 99 and 99.5%, respectively. Gases and liquids were used without further purification.

Apparatus and Procedure. The apparatus and procedure used to carry out the solubility measurements have been described in detail elsewhere.[6] The equipment is similar to that of Ben-Naim and Baer.[7] Its main parts are a solution vessel, a mercury manometer, and a buret system. The solution vessel containing a known mass of liquid solvent is immersed in an ethanol 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 a thermostated air bath within ±0.2 K. The temperature of this air bath is higher than that of the ethanol bath to prevent condensation of the vapor solvent out of the solution vessel.

The experimental technique is based on the determination of the volume of wet gas (gas saturated with vapor of solvent) which dissolves in a known mass of the degassed solvent at constant pressure (~101.33 kPa partial pressure of gas) and temperature. To keep the pressure constant during the measuring process, the wet gas dissolved into the liquid phase is replaced by mercury introduced in the graduated burets. The pressure is controlled and measured using a cathetometer (Wild KM 338) with an uncertainty

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Table 1. Densities of Pure Compounds at (278.15, 288.15, and 298.15) K and Comparison with Literature Data

	ρ/kg·m^{-3}			
	dimethyl carbonate		diethyl carbonate	
T/K	this work	literature	this work	literature
278.15	1089.21	1089.83[a]	991.60	
288.15	1075.54	1076.46[a]	980.09	980.27[a]
		1076.30[b]		980.35[a]
		1076.56[c]		
298.15	1062.29	1063.38[a]	968.89	969.06[c]
		1063.11[b]		969.21[d]
		1063.47[d]		969.23[h,i]
		1063.49[e]		969.0[j,k]
		1063.50[f]		969.16[a,l]
		1063.059[g]		

[a] Reference 32. [b] Reference 34. [c] Reference 35. [d] Reference 36. [e] Reference 37. [f] Reference 38. [g] Reference 39. [h] Reference 40. [i] Reference 41. [j] Reference 42. [k] Reference 43. [l] Reference 44.

of ±3 Pa. The volume of mercury introduced, that is, the volume of wet gas dissolved, is measured with an uncertainty of ±0.01 cm^3. The relative uncertainty in the mole fraction of dissolved gas (x_2) is estimated to be <1%.

The densities of pure liquids were determined using an Anton-Paar DMA-58 vibrating tube density meter with an uncertainty of ±0.01 kg·m^{-3}. The values determined are gathered in Table 1 and compared with the literature values.

Results and Discussion

Solubilities and Thermodynamic Properties for the Solution Process. The solubility values were calculated with a reduction method that is a modification[8,9] of that proposed by Wilhelm and Battino.[10,11] The modification takes into account the features of our apparatus, in particular that the temperature of the gas phase in the burets is different from the temperature in the solution vessel and that the gas phase dissolved is a saturated mixture; namely, it includes solvent vapor that must be discounted.

The method determines the actual amount of gas dissolved in the liquid and the actual partial pressure over the solution. Then, the solubility is corrected to a partial pressure of 101.33 kPa. Given the complexity of the equilibrium equations, some simplifications are assumed with respect to the liquid-phase activity coefficients, the fugacity coefficients, and the partial molar volumes in the Poynting correction. The behavior of the gas phase is well described by means of the second virial coefficients. For the gases, second virial coefficients were calculated from the characteristic parameters founded in the literature,[12,13] whereas, for the pure solvents and their mixtures with the gases, second virial coefficients were estimated using the method of Maris and Stiel.[14] The calculation method has been described earlier.[5]

Table 2 shows the solubilities of gases in dimethyl carbonate and diethyl carbonate at 278.15 K and 288.15 K. The mole fractions of gas in liquid (x_2) for T = (278.15 and 288.15) K along with those at 298.15 K[5] have been fitted as a function of the temperature using the following polynomial, which is convenient[15] for this rather narrow temperature range.

$$\ln x_2 = A_0 + \frac{A_1}{T} \tag{1}$$

The coefficients A_0 and A_1, obtained by means of the least-squares method, are listed in Table 3, together with the corresponding standard deviation. Figure 1 shows the variation of ln x_2 with 1/T. As usual, the solubility of helium and neon increases with temperature. A similar behavior is observed for argon.

Table 2. Solubilities of Gases in Dimethyl Carbonate and Diethyl Carbonate at 101.33 kPa Partial Pressure of Gas

	$10^4 x_2$			
	dimethyl carbonate		diethyl carbonate	
gas	278.15 K	288.15 K	278.15 K	288.15 K
He	0.507	0.694	1.048	1.263
Ne	0.810	0.998	1.523	1.763
Ar	6.446	7.135	12.60	12.96
Kr	22.35	21.44	39.71	38.08
Xe	85.10	77.64	159.6	142.8
CH_4	16.64	16.30	28.75	27.97
C_2H_4	128.4	113.0	200.2	174.9
C_2H_6	104.7	92.85	200.3	173.6
SF_6	32.22	30.56	56.86	53.99

Table 3. Fitting Coefficients of eq 1 for the Variation of the Solubilities of Gases with Temperature and their Corresponding Standard Deviations

	dimethyl carbonate			diethyl carbonate		
gas	A_0	A_1/K	σ[a]	A_0	A_1/K	σ[a]
He	0.226	−2817.0	0.18	−3.246	−1647.7	0.13
Ne	−2.802	−1843.1	0.13	−4.104	−1304.7	0.12
Ar	−4.058	−916.05	0.10	−5.783	−248.78	0.05
Kr	−7.199	304.36	0.05	−6.665	315.82	0.05
Xe	−7.207	678.31	0.08	−7.065	813.45	0.09
CH_4	−6.913	142.34	0.02	−6.629	216.16	0.02
C_2H_4	−8.310	1100.8	0.09	−7.513	1001.0	0.09
C_2H_6	−7.868	919.85	0.07	−7.655	1040.4	0.10
SF_6	−7.181	401.28	0.05	−6.536	379.69	0.06

[a] $\sigma = [(\Sigma \ln(x_2)_{\mathrm{exptl}} - \ln(x_2)_{\mathrm{calcd}})^2)/(N - N_P)]^{1/2}$. N, number of experimental points. N_P, number of adjustable coefficients.

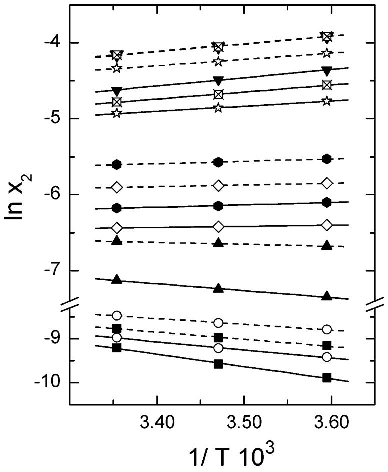

Figure 1. Variation of solubility expressed as ln x_2 with temperature: solid line, dimethyl carbonate; dashed line, diethyl carbonate; ■, He; ○, Ne; ▲, Ar; ⬢, Kr; ☆, Xe; ◇, CH_4; ▼, C_2H_4; ×, C_2H_6; □, SF_6.

The solubilities of gases increase according to the following series for both liquids: He < Ne < Ar < CH_4 < Kr < SF_6 < Xe < C_2H_6 < C_2H_4. There is an exception in the case of diethyl carbonate; specifically, C_2H_4 and C_2H_6 show very close solubilities. As we pointed out previously,[5] given a series of solvents with the same functional group, the greater the size of a molecule, generally the greater the solubility.[16] In fact, the solubilities of the gases are considerably higher in diethyl carbonate compared with dimethyl carbonate.

Table 4. Standard Partial Molar Changes at 298.15 K in the Gibbs Energy, Enthalpy, and Entropy for the Solution Process of the Gases in Dimethyl Carbonate and in Diethyl Carbonate along with the Values Obtained from the SPT

gas	$\Delta\bar{G}^\circ_{m,2}$ / kJ·mol^{-1}	$\Delta\bar{H}^\circ_{m,2}$ / kJ·mol^{-1}	$\Delta\bar{S}^\circ_{m,2}$ / J·K^{-1}·mol^{-1}	$\Delta\bar{G}^\circ_{m,2}$(SPT) / kJ·mol^{-1}	$\Delta\bar{H}^\circ_{m,2}$(SPT) / kJ·mol^{-1}	$\Delta\bar{S}^\circ_{m,2}$(SPT) / J·K^{-1}·mol^{-1}
			Dimethyl Carbonate			
He	22.83	23.40	1.910	22.27	5.643	−55.77
Ne	22.25	15.31	−23.28	19.93	3.342	−55.64
Ar	17.66	7.621	−33.67	16.50	0.114	−54.96
Kr	15.31	−2.532	−59.84	14.83	−1.444	−54.58
Xe	12.22	−5.643	−59.91	12.92	−3.221	−54.14
CH_4	15.95	−1.268	−57.75	15.43	−0.835	−54.55
C_2H_4	11.45	−9.171	−69.16	12.40	−3.676	−53.92
C_2H_6	11.85	−7.646	−65.39	12.19	−3.705	−53.31
SF_6	14.46	−3.339	−59.70	14.79	−0.266	−50.50
			Diethyl Carbonate			
He	21.73	13.71	−26.90	20.84	4.787	−53.84
Ne	21.01	10.85	−34.08	18.40	2.374	−53.75
Ar	16.40	2.069	−48.07	14.82	−1.075	−53.31
Kr	13.89	−2.627	−55.43	13.15	−2.675	−53.08
Xe	10.74	−6.768	−58.72	11.27	−4.450	−52.73
CH_4	14.63	−1.798	−55.10	13.74	−2.077	−53.05
C_2H_4	10.29	−8.328	−62.48	10.76	−4.921	−52.59
C_2H_6	10.31	−8.657	−63.65	10.59	−4.966	−52.18
SF_6	13.04	−2.394	−51.77	13.20	−1.725	−50.06

Table 5. Experimental and Theoretical Values of Gibbs Energy according to the Ben-Naim Standard State, Cavity, and Interaction Terms Using the Scaled Particle Theory at 298.15 for Dimethyl Carbonate and Diethyl Carbonate

	dimethyl carbonate				diethyl carbonate			
gas	$\Delta\bar{G}^*_s$ (exptl) / kJ·mol^{-1}	$\Delta\bar{G}^*_s$(SPT) / kJ·mol^{-1}	$\Delta\bar{G}_c$ / kJ·mol^{-1}	$\Delta\bar{G}_i$ / kJ·mol^{-1}	$\Delta\bar{G}^*_s$(exptl) / kJ·mol^{-1}	$\Delta\bar{G}^*_s$(SPT) / kJ·mol^{-1}	$\Delta\bar{G}_c$ / kJ·mol^{-1}	$\Delta\bar{G}_i$ / kJ·mol^{-1}
He	8.79	8.22	10.22	−1.997	8.59	7.69	9.717	−2.028
Ne	8.21	5.89	11.05	−5.163	7.87	5.25	10.47	−5.217
Ar	3.62	2.46	14.59	−12.13	3.26	1.68	13.70	−12.02
Kr	1.27	0.79	16.23	−15.44	0.75	0.01	15.20	−15.19
Xe	−1.82	−1.13	18.17	−19.30	−2.40	−1.87	16.96	−18.83
CH_4	1.91	1.38	16.42	−15.04	1.49	0.60	15.38	−14.78
C_2H_4	−2.59	−1.64	18.94	−20.58	−2.85	−2.38	17.66	−20.04
C_2H_6	−2.19	−1.85	21.19	−23.04	−2.83	−2.56	19.70	−22.26
SF_6	0.42	0.74	30.51	−29.77	−0.10	0.06	28.17	−28.11

From the solubility values, the changes in Gibbs energy ($\Delta\bar{G}^\circ_{m,2}$), enthalpy ($\Delta\bar{H}^\circ_{m,2}$), and entropy ($\Delta\bar{S}^\circ_{m,2}$) can be determined for the hypothetical solution process

$$M(\text{gas, hyp., 101.33 kPa}) \rightarrow M(\text{solution, hyp., } x_2 = 1) \quad (2)$$

using the relations reported by Clever and Battino[15] and Wilhem et al.[17]

$$\Delta\bar{G}^\circ_{m,2} = -RT(\ln x_2)_P = -RT\left(A_0 + \frac{A_1}{T}\right)_P \quad (3)$$

$$\Delta\bar{H}^\circ_{m,2} = RT\left[\frac{\partial \ln x_2}{\partial \ln T}\right]_P = -RA_1 \quad (4)$$

$$\Delta\bar{S}^\circ_{m,2} = R\left(\left[\frac{\partial \ln x_2}{\partial \ln T}\right]_P + \ln x_2\right) = RA_0 \quad (5)$$

The values of these standard partial molar thermodynamic properties at 298.15 K are reported in Table 4. An analysis about enthalpies and entropies obtained from solubility measurements has been done by Olofsson et al.[18]

Scaled Particle Theory (SPT). *General Features.* A model often used in the treatment of the solubilities of gases in liquids is Pierotti's application[19−23] of the scaled particle theory (SPT) based on the mechanic-statistical model for fluids developed by Reiss et al.[24,25]

Pierotti's application, apart from its simplicity, provides a reasonable mechanism for the solution process, leads to acceptable results for the solubility values, and allows the determination of molecular parameters for the liquid solvent, namely, the effective hard-sphere diameter and the depth of the energy well.

Although the theory was developed for organic solvents with low dipole moments,[19] it has yielded good results when it has been extended to rather polar liquids[5,26] or associated solvents such as alcohols[11,27,28] and even water.[29−31] Therefore, its application to these systems can lead to reliable information about dimethyl carbonate and diethyl carbonate. The thermodynamic functions are derived using the SPT equations for hard spheres on the basis of an effective Lennard-Jones (6,12) potential.[17,24] The hard-sphere diameter of liquid solvent (σ_1) determined with SPT equations is identified with the distance parameter of the potential.

In this paper, we have considered the changes of Gibbs energy, enthalpy, and entropy at 298.15 K for both solvents.

Gibbs Energy, Enthalpy, and Entropy of Solution. The change of Gibbs energy and enthalpy accompanying the solution process can be expressed by

$$\Delta\bar{G}^\circ_{m,2}(\text{SPT}) = \Delta\bar{G}_c + \Delta\bar{G}_i + RT\ln\left(\frac{RT}{V_{m,1}}\right) \quad (6)$$

$$\Delta\bar{H}^\circ_{m,2}(\text{SPT}) = \left[\frac{\partial\left(\frac{\Delta\bar{G}^\circ_{m,2}(\text{SPT})}{T}\right)}{\partial\left(\frac{1}{T}\right)}\right]_P = \Delta\bar{H}_c + \Delta\bar{H}_i - RT + \alpha_p RT^2 \quad (7)$$

where $\Delta\bar{G}_c$ and $\Delta\bar{H}_c$ and $\Delta\bar{G}_i$ and $\Delta\bar{H}_i$ are the changes of Gibbs energy and enthalpy in the formation of cavity and interaction steps, respectively, $V_{m,1}$ is the molar volume of the pure solvent, and α_p is the thermal expansion coefficient of the solvent.

The entropy can be directly obtained from the following relation

$$\Delta\bar{S}^\circ_{m,2}(\mathrm{SPT}) = \frac{\Delta\bar{H}^\circ_{m,2}(\mathrm{SPT}) - \Delta\bar{G}^\circ_{m,2}(\mathrm{SPT})}{T} \qquad (8)$$

As can be seen from the results shown in Tables 4 and 5, the results of the SPT formalism for the changes of enthalpy are, in general, not as good as they are for the corresponding Gibbs energy changes. On the other hand, taking into account the limits of this model, the changes in entropy are reasonably described except for He, Ne, and Ar.

The detailed expressions for $\Delta\bar{G}_c$, $\Delta\bar{G}_i$, and $\Delta\bar{H}_c$ are widely described in the literature.[5,19,20,24,25] The parameters needed in those equations for gases and liquids were taken from the literature.[8,32] We must note that no dependence of σ_1 on temperature was considered for the $\Delta\bar{H}_c$ calculations ($l_1 = 0$) and that for low pressures $\Delta\bar{H}_i = \Delta\bar{G}_i$.

The values of the changes in Gibbs energy and enthalpy associated with the cavity formation and interaction steps are shown in Table 5 as well as the experimental and calculated thermodynamic quantity for the solvation process defined by Ben-Naim and Marcus.[33]

$$\Delta\bar{G}^*_s = \Delta\bar{G}^\circ_{m,2} - RT\ln\left(\frac{RT}{V_{m,1}}\right) \qquad (9)$$

For He, Ne, Ar, Kr, and CH_4, the solvation process in both solvents is not favored thermodynamically ($\Delta\bar{G}^*_s > 0$) under the Ben-Naim considerations. The SPT well reproduces this behavior, indicating a prevalence of the cavity creation term over the interaction one.

The high solubilities of Xe, C_2H_4, and C_2H_6 in the carbonates suggest a solvation process thermodynamically favored as is pointed out by $\Delta\bar{G}^*_s < 0$. This behavior is also well reproduced by the SPT formalism. For these voluminous molecules of gas, the interactive term is higher because of their greater energy parameters.

We can conclude that Pierotti's approach, despite its limitations, gives a physicochemical explanation for the solution process for gases that are low or moderately soluble in the alkyl carbonates, even if the molecules of solvent are far from being spherical.

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