

Solubility of oxygen in liquid perfluorocarbons¹

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Abstract

The solubility of oxygen in several liquid perfluorocarbons was measured in the temperature range between 288 and 313 K and at pressures close to atmospheric. Values were measured with an apparatus based on the saturation method with an accuracy of $\pm 1\%$. Thermodynamic functions such as Gibbs energy, enthalpy and entropy of solution were obtained from the composition and temperature dependence. The Peng–Robinson equation of state was used to model the gas solubility for the studied systems.

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1. Introduction

Fluorinated molecules such as perfluorocarbons (PFCs) and their derivatives represent a very interesting and stimulating class of chemicals in the physical chemistry and polymer science due to their specific and unusual properties. Perfluorochemicals are non-polar highly fluorinated compounds and as a result of the strong intramolecular bonding (C–F bonds are 485 kJ mol^{-1} , that is, 84 kJ mol^{-1} more than a regular C–H bond), they are chemical and biochemical inert. The chemical structure and the weak intermolecular interactions are responsible for the specific properties of PFCs namely the low surface tensions ($< 20 \text{ mN m}^{-1}$), dielectric constants and refractive indices, the high densities, viscosities and gas solubilities that are the largest known for liquids [1].

Due to these properties PFCs can be used on a wide variety of areas such as surfactants in supercritical solvents, environmental probes, anticorrosive and antifriction components, as flame retardants, water repellents, or sliding agents, in paints, coatings, polymer technology, metal working and uranium recovery process. It is, however, in the biomedical field that most of the relevant applications are found, based on the large solubility of respiratory gases: they can be used in tissue oxygenation fluids (blood substitutes, oxygen thera-

peutics), anti-tumoral agents, perfusates for isolated organs, surgical tools for ophthalmology, lubrication and cushioning for articular disorders, cell culture media supplements and drug formulations and delivery.

From the fundamental point of view, solubility measurements of gases in liquids play a special role since they provide information about the organization of the solvent around a solute [2]. The solubility of a gas in a liquid is usually described as a two-step process: the first step consists on the formation of a large enough cavity inside the liquid solvent to accommodate the solute molecule, while the second step is the insertion of the solute molecule in the solvent's cavity. Thus, a large value of gas solubility corresponds to the easiness in forming a cavity within the liquid solvent or/and the establishment of favourable interactions between the solute and the solvent.

The study of the interactions between the molecular oxygen and perfluorinated liquid compounds has recently received a lot of attention due to the promising application of these liquids in the medical field. NMR studies of solutions of oxygen in perfluorocarbons demonstrate that there is a general correlation between the solubility of oxygen and the paramagnetic relaxation brought to the neighbouring carbon nuclei of the solvent: higher values of solubility are associate with low relaxation coefficients, thus showing that the increase in solubility is promoted by the small magnetic dipolar interaction between the oxygen and the solvent nuclei. This fact has usually been rationalized by assuming that the liquid perfluoro-*n*-alkanes form large cavities capable of accommodating easily molecules of solute [3]. This is

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coherent with molecular dynamics results where it is shown that perfluoro-*n*-alkanes have a more rigid backbone than the corresponding *n*-alkanes, for the same temperature, thus tending to adopt linear conformations in the liquid phase [4]. Several ab initio studies have been performed in recent years in order to give some more insight in the interaction between fluorine atoms and small molecules, like water [5] or CO₂ [6].

The main objective of this work is to measure the solubilities of small molecules, oxygen in this case, in perfluorocarbons, in order to gain some insight on the factors governing this process.

2. Experimental

The apparatus and procedure used for the solubility measurements were described in detail in a previous paper [7]. The apparatus is based on the saturation method proposed by Ben Naim and Battino [8], where the solubility is determined by measuring the quantity of gas dissolved in an accurately known volume of solvent, at constant pressure and temperature. It consists mainly of a mercury manometer, with a mercury reservoir, a calibrated dissolution cell and a gas line with a pre-saturator, where the gas phase is pre-saturated with the solvent. The entire apparatus is immersed in an air bath, capable of maintaining the temperature to within 0.1 K. The precision of the experimental apparatus was estimated to be $\pm 1\%$ [7]. The degassing of the solvent and the pre-saturation are very important steps in the measurement of gas solubility in perfluorocarbons, especially perfluoro-*n*-hexane, due to the high volatility and high gas solubility.

Chemicals used for the measurements were perfluoro-*n*-hexane, perfluoro-*n*-heptane and perfluoro-*n*-nonane from Aldrich with a stated purity of 99, 85 and 97%, respectively, and perfluorodecalin from ABCR, with a stated purity of 96%. The oxygen used was from Air Liquide with 99.999% mol/mol minimum stated purity. Solvents and gas were used with no further purification.

3. Results and discussion

3.1. Data reduction

There are many ways to express the solubility of a gas in a liquid. In this work, experimental results are expressed in terms of Ostwald coefficient and solute molar fraction. The Ostwald coefficient for solution volume is defined as [9]:

$$L_{2,1}(T, p) = \left(\frac{V_g}{V_l} \right)_{\text{equil}} \quad (1)$$

where V_g is the volume of the dissolved gas and V_l the total volume of the liquid solution after equilibrium is reached.

Both quantities are obtained directly from the experimental measurement.

The molar fraction of component 2 (the gaseous solute) in the liquid solution can be directly related to the Ostwald coefficient in the following way:

$$x_2 = \frac{L_{2,1}(T, p)p_2V^L(T, p)}{Z_{12}RT} \quad (2)$$

where p_2 is the partial pressure of the solute, $V^L(T, p)$ the molar volume of the liquid solution and Z_{12} the compressibility factor of the solution. In this work, the virial equation of state is used to calculate Z_{12} as follows:

$$Z_{12} = 1 + \frac{P}{RT}B \quad (3)$$

where B is the second virial coefficient for the solvent–solute mixture and is given by

$$B = y_1B_{11} + y_2B_{22} + y_1y_2\delta_{12} \quad (4)$$

B_{11} and B_{22} are the second virial coefficients for the pure solvent and the pure solute, respectively, and $\delta_{12} = 2B_{12} - B_{11} - B_{22}$, being B_{12} the solute–solvent cross second virial coefficient. The mole fractions of the vapor phase in equilibrium with the liquid solution, y_i , are calculated by an iterative process using the vapor liquid equilibrium equation [10]:

$$y_1 = (1 - x_2) \left(\frac{p_1^{\text{sat}}}{p} \right) \left(\frac{\Phi_1^{\text{sat}}}{\Phi_1} \right) \exp \left[\frac{V_1^0(p - p_1^{\text{sat}})}{RT} \right] \quad (5)$$

The molar volume of the liquid solution, $V^L(T, p)$ was taken as the molar volume of the pure solvent, V_1^0 , calculated from density measurements that were performed in our laboratory [11]. The solvents' vapor pressures were taken from the work performed in our laboratory [11] for perfluoro-*n*-hexane and perfluorodecalin and Steele et al. [12] for perfluoro-*n*-heptane. Vapor pressure data for perfluoro-*n*-nonane was not found in the literature. The accuracy of Peng–Robinson equation of state (PR-EOS) [13] in predicting vapor pressures was tested with *n*-hexane, *n*-heptane and *n*-octane, yielding good results. Thus, it was assumed that it can describe as accurately the vapor pressure of perfluoro-*n*-nonane and the vapor pressure data evaluated in this way was used. The second virial coefficient for the solute was obtained from the compilation of Dymond and Smith [14] and both the second virial coefficient for the solvent and the cross virial coefficient were estimated using the correlation proposed by Tsionopoulos [15].

3.2. Experimental results

Experimental values obtained for the solubility of oxygen in liquid perfluorocarbons, between 288 and 318 K are presented in Table 1.

The accuracy of the method was determined in a previous work and it is believed to be better than 1%. There is very

Table 1

Experimental data for the solubility of oxygen in the perfluorocarbons, between 288 and 313 K, expressed as Ostwald's coefficients, $L_{2,1}(T, p)$, and solute mole fraction, x_2 , at a solute partial pressure of 101,325 Pa

Solvent	T (K)	$L_{2,1}(T, p)$	$10^3 x_2$	δ_i
C ₆ F ₁₄	287.40	0.580 ± 0.004	5.11 ± 0.03	1.37
	291.39	0.530 ± 0.004	4.65 ± 0.02	2.34
	298.65	0.485 ± 0.003	4.24 ± 0.02	1.70
	301.73	0.448 ± 0.004	3.91 ± 0.02	0.34
	306.65	0.395 ± 0.003	3.44 ± 0.02	0.48
C ₇ F ₁₆	287.94	0.530 ± 0.004	5.02 ± 0.03	1.74
	290.94	0.519 ± 0.004	4.89 ± 0.03	-0.44
	293.96	0.511 ± 0.004	4.80 ± 0.03	-0.79
	297.88	0.499 ± 0.004	4.66 ± 0.03	0.12
	303.94	0.468 ± 0.003	4.35 ± 0.03	2.57
	308.35	0.419 ± 0.003	3.87 ± 0.02	0.23
	311.95	0.382 ± 0.003	3.52 ± 0.02	-0.25
C ₉ F ₂₀	288.28	0.503 ± 0.004	5.75 ± 0.04	-0.19
	288.48	0.507 ± 0.004	5.80 ± 0.04	0.71
	292.29	0.492 ± 0.004	5.58 ± 0.04	-1.19
	298.04	0.486 ± 0.004	5.46 ± 0.04	0.32
	302.35	0.472 ± 0.003	5.27 ± 0.03	-0.02
	307.79	0.451 ± 0.003	5.05 ± 0.03	0.41
	311.43	0.439 ± 0.003	4.83 ± 0.03	-0.46
C ₁₀ F ₁₈	288.85	0.414 ± 0.003	4.15 ± 0.03	
	291.08	0.408 ± 0.003	4.07 ± 0.03	
	293.94	0.410 ± 0.003	4.07 ± 0.03	
	299.82	0.407 ± 0.003	3.99 ± 0.03	
	306.31	0.412 ± 0.003	3.99 ± 0.03	
	311.61	0.417 ± 0.003	3.99 ± 0.03	
	313.20	0.408 ± 0.003	3.90 ± 0.03	

little data available in the open literature for the solubility of oxygen in highly fluorinated compounds, and most of this data is widely scattered as confirmed by Wesseler et al. [16]. The results obtained in this work are compared with literature values in Table 2, at 298 K and at a partial pressure of the gas equal to the atmospheric pressure.

Previously published results for perfluoro-*n*-hexane [4] agree with the data measured in this work to within 1.5%. As can be observed, the largest deviation was found for perfluoro-*n*-heptane. Once Gjaldbaek [17] also states an accuracy of 1% it is believed that this deviation is due to differences in the purity of the perfluoro-*n*-heptane used. In this work a mixture of isomers of perfluoro-*n*-heptane with a standard purity of 85% is used, while Gjaldbaek [17] used

Table 2

Comparison of the solubility of oxygen in perfluorocarbons obtained in this work, expressed as oxygen molar fraction, with literature values at 298 K and a solute partial pressure of 101,325 Pa

Solvent	Literature value		Present work		Deviation (%)
	x_2 ($\times 10^{-3}$)	d (g/cm ³)	x_2 ($\times 10^{-3}$)	d (g/cm ³)	
<i>n</i> -C ₆ F ₁₄	4.28 [4]	1.670	4.23	1.678	1.4
<i>n</i> -C ₇ F ₁₆	4.89 [17]	1.721	4.66	1.728	5.0
<i>n</i> -C ₉ F ₂₀	5.46 [16]	1.840	5.45	1.788	0.2
C ₁₀ F ₁₈	3.92 [16]	1.946	3.99	1.930	1.8

a purer compound. Wesseler et al. [16] do not state the accuracy of the method used to measure the solubility of oxygen in perfluoro-*n*-nonane and perfluorodecalin.

Solubilities in mole fraction $x_2(T, p)$, as a function of absolute temperature were fitted to the following equation, as suggested by Benson and Krause [18]:

$$\ln x_2 = \sum_{i=0}^n A_i T^{-i} \quad (6)$$

The average absolute deviation of the experimental results from Eq. (6) was calculated as:

$$\text{AAD} = N^{-1} \sum_{i=1}^N |\delta_i| \quad (7)$$

where N is the number of data points, whose individual percentage deviations are calculated as:

$$\delta_i = \frac{100 [x_2(\text{exp}) - x_2(\text{calc})]}{x_2(\text{calc})} \quad (8)$$

The coefficients A_i of Eq. (5) as well as the AAD for all the studied systems are listed in Table 3. The small values of the AADs describe well the precision of the solubility experimental results. The fact that AAD decreases with the chain length is probably due to the fact that the vapor pressure of the solvents decreases, leading to an easier experimental procedure and lower experimental incertitude.

3.3. Thermodynamic functions

The dissolution of a gas into a liquid is associated with changes in thermodynamic functions namely standard Gibbs energy (ΔG_2°), standard enthalpy (ΔH_2°) and standard entropy (ΔS_2°) of solution, which can be calculated from experimental solubility results. These functions represent the changes that occur in the solute neighbourhood during dissolution process due to the transference of one solute particle from the pure perfect gas state to an infinitely dilute state in the solvent, at a given temperature T [19]. The thermodynamic functions were calculated from the temperature dependence of the molar fraction according to the following equation [20]:

$$\Delta G_2^\circ = -RT (\ln x_2)_p \quad (9)$$

$$\Delta H_2^\circ = RT^2 \left(\frac{\partial (\ln x_2)}{\partial T} \right)_p \quad (10)$$

Table 3

Coefficients for Eq. (6) and average percentage deviation (AAD) for the correlation of the experimental data

System	A_0	A_1	A_2	AAD
O ₂ -C ₆ F ₁₄	-3.292 × 10 ⁶	2.392 × 10 ⁴	-4.866 × 10 ¹	1.25
O ₂ -C ₇ F ₁₆	-5.035 × 10 ⁶	3.488 × 10 ⁴	-6.572 × 10 ¹	0.88
O ₂ -C ₉ F ₂₀	-1.123 × 10 ⁶	8.166 × 10 ³	-1.997 × 10 ¹	0.47

Table 4
Thermodynamic properties of solution for oxygen in different perfluorocarbons

Solvent	<i>T</i> (K)	ΔG_2° (kJ mol ⁻¹)	ΔH_2° (kJ mol ⁻¹)	ΔS_2° (J mol ⁻¹ K ⁻¹)
C ₆ F ₁₄	287.40	12.61	-8.40	-73.22
	291.39	13.01	-11.10	-82.23
	298.65	13.57	-15.57	-97.71
	301.73	13.91	-17.44	-103.94
	306.65	14.47	-20.35	-113.51
C ₇ F ₁₆	287.94	12.68	0.76	-41.51
	290.94	12.87	-2.24	-51.87
	293.96	13.05	-5.20	-61.98
	297.88	13.29	-8.94	-74.65
	303.94	13.74	-14.55	-93.27
	308.35	14.24	-18.49	-106.14
C ₉ F ₂₀	311.95	14.65	-21.62	-116.24
	288.28	12.36	-3.12	-53.69
	288.48	12.35	-3.17	-53.85
	292.29	12.61	-4.01	-56.75
	298.04	12.91	-5.24	-60.93
	302.35	13.19	-6.13	-63.91
	307.79	13.53	-7.23	-67.48
311.43	13.81	-7.94	-69.77	

$$\Delta S_2^\circ = R \left(\frac{\partial(\ln x_2)}{\partial(\ln T)} \right)_p \quad (11)$$

where *R* is the gas constant.

The values for the thermodynamic functions of solution of oxygen in perfluorocarbons are listed in Table 4. In the O₂/perfluorodecalin case the solubility results are temperature independent, as can be seen from the analysis of Table 1. Thus, no temperature fitting was performed.

These results were obtained considering the ideal gas state at 101,325 Pa. No data obtained by calorimetric measurements were found for comparison.

4. Modelling

In this work, the PR-EOS was used to model the solubility of oxygen in the perfluorocarbons. The original form of the PR-EOS is given by [13]:

$$P = \frac{RT}{V-b} - \frac{a(T)}{V(V+b) + b(V-b)} \quad (12)$$

The experimental data used for the perfluorocarbon systems is the data obtained in this work together with the oxygen-perfluoro-*n*-octane values taken from a previous work [7].

4.1. Pure compounds

The PR-EOS was first used to predict equilibrium properties of the pure components under study. The critical properties of the PFC's were taken from the work of Vandana et al.

Table 5
Critical properties and acentric factors for the PFC's used in this work

Compound	<i>T_c</i> (K)	<i>P_c</i> (MPa)	<i>w</i>
C ₆ F ₁₄	450.60 ± 1.1	1.88 ± 0.02	0.5140
C ₇ F ₁₆	475.30 ± 0.9	1.65 ± 0.02	0.5611
C ₈ F ₁₈	498.20 ± 0.5	1.55 ± 0.02	0.6231
C ₉ F ₂₀	523.95 ± 1.0	1.56 ± 0.04	0.6756 ^a

^a Extrapolated from the other linear perfluorocarbons.

[21] and are reported in Table 5. As it was mentioned before, the Peng–Robinson equation of state describes within 4% the experimental vapor pressure data for both the solute and solvents.

4.2. Mixtures

To predict the properties of a mixture with the PR-EOS, the mixture parameters *a* and *b* are calculated from the parameters of the pure components *a_{ii}* and *b_{ii}* using the one fluid van der Waals mixing rules

$$a = \sum_i \sum_j z_i z_j a_{ij} \quad (13)$$

$$b = \sum_i z_i b_i \quad (14)$$

where *z_i* is the phase mole fraction of component *i*. The combining rule used for *a* is

$$a_{ij} = (1 - k_{ij})(a_{ii}a_{jj})^{1/2} \quad (15)$$

The *k_{ij}* is an adjustable parameter for each binary mixture. The *k_{ij}*'s fitted to the experimental solubility data obtained in this work for O₂/perfluorocarbons, show a strong temperature dependence described by a quadratic equation of the type [22]:

$$k_{ij} = A + \frac{B}{T} + \frac{C}{T^2} \quad (16)$$

The coefficients *A*, *B* and *C* for the systems studied in this work as well as for the oxygen/perfluoro-*n*-octane system, also measured in our laboratory [7] are given in Table 6.

Fig. 1 compares experimental results with the predictions given by the PR-EOS using the average binary interaction

Table 6
Coefficients for Eq. (16) and average binary interaction parameters, for all the temperatures

System	<i>A</i>	<i>B</i>	<i>C</i>	$\overline{k_{ij}}$	AAD (%)
O ₂ -C ₆ F ₁₄	3.66 × 10 ¹	-2.08 × 10 ⁴	2.94 × 10 ⁶	0.122	12.35
O ₂ -C ₇ F ₁₆	2.20 × 10 ¹	-1.26 × 10 ⁴	1.80 × 10 ⁶	0.091	10.07
O ₂ -C ₈ F ₁₈	3.49 × 10 ¹	-2.05 × 10 ⁴	3.00 × 10 ⁶	0.017	7.54
O ₂ -C ₉ F ₂₀	7.48 × 10 ⁰	-4.35 × 10 ³	6.25 × 10 ⁵	-0.073	2.95

AAD is the corresponding average absolute errors for binary oxygen-fluorocarbon mixtures when $\overline{k_{ij}}$ is used.

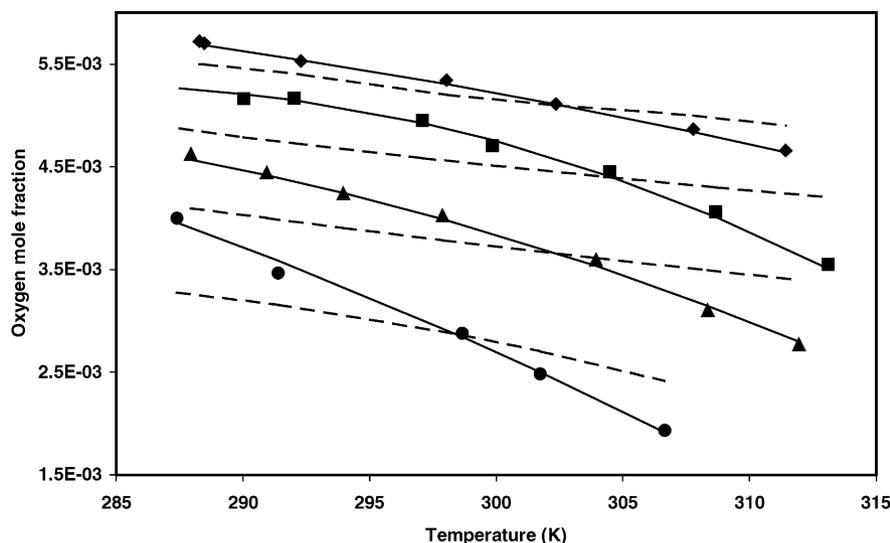


Fig. 1. Solubility of oxygen in liquid perfluorocarbons using the Peng–Robinson equation of state with a constant k_{ij} , for each binary system: perfluoro-*n*-hexane (●), perfluoro-*n*-heptane (▲), perfluoro-*n*-octane (■) and perfluoro-*n*-nonane (◆). Solid lines: prediction given by the Peng–Robinson EOS using temperature dependent k_{ij} 's calculated from Eq. (16). Dashed lines: prediction given by the Peng–Robinson EOS using the average k_{ij} shown in Table 6.

parameter k_{ij} given in Table 6, and using temperature dependent k_{ij} calculated from Eq. (16). It can be observed that a temperature independent k_{ij} cannot describe the oxygen solubility in perfluorocarbons. The average absolute deviation given in Table 6 decreases from the O_2 – C_6F_{14} to the O_2 – C_9F_{20} system because the temperature dependence of the solubility decreases along the perfluorocarbon series.

5. Conclusions

Original data for the solubility of oxygen in linear perfluorocarbons and in perfluorodecalin, in the temperature range between 288 and 313 K and at pressures close to atmospheric, were presented.

Thermodynamic functions, which are difficult to obtain through calorimetric measurements, were calculated from the temperature dependence of the molar fraction.

It was demonstrated that a traditional cubic equation of state like the Peng–Robinson EOS, with one adjusted interaction binary parameter, has limited predictive capability providing only a qualitative description of the temperature dependence of the oxygen solubility in perfluorocarbons.

List of symbols

B	second virial coefficient (m^3/mol)
ΔG°	standard Gibbs energy ($kJ\ mol^{-1}$)
ΔH°	standard enthalpy of solution ($kJ\ mol^{-1}$)
R	universal gas constant ($8.314 \times 10^{-3}\ Pa\ m^3/(mol\ K)$)

ΔS°	standard entropy of solution ($J\ mol^{-1}\ K$)
T	temperature (K)
V	liquid molar volume (cm^3/mol)
x	mole fraction of the solute in the liquid phase
y	mole fraction of the solute in the gaseous phase
Z	compressibility factor

Greek letter

ϕ	fugacity coefficient
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Subscripts

c	critical
calc	calculated
exp	experimental
g	dissolved gas
1	liquid solution in equilibrium
1	solvent
2	solute

Superscripts

0	pure component
i	ideal
L	liquid solution
sat	saturated property

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