Some Thermodynamic Properties of C$_{76}$ and C$_{84}$

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The vapor pressures of C$_{76}$ were measured over the temperature range 834−1069 K by the torsion−effusion method. The results are well represented by the following linear equation: $\log(p/\text{kPa}) = (8.23 \pm 0.20) - (10150 \pm 150)/(T/\text{K})$. The standard sublimation enthalpy of this compound, $\Delta_{sub}H°(298) = 206 \pm 4$ kJ mol^{-1}, as derived by second-law elaboration of the experimental data, is proposed. Some structural aspects of C$_{76}$ and C$_{84}$ crystals were investigated by X-ray analysis. Considerations on the thermodynamic properties of these compounds are also reported.

A. Introduction

Apparently at the present no data are reported for the vapor pressure of C$_{76}$. In an ongoing research program concerning the study of the sublimation thermodynamics of fullerenes,[1−3] this work has been undertaken in order (i) to measure the vapor pressure and the sublimation enthalpy of C$_{76}$ and to compare the obtained results with those of the previously studied fullerenes, (ii) to determine, from X-ray analysis, the structures of C$_{84}$ and C$_{76}$, and (iii) to evaluate the thermodynamic properties of both fullerenes in solid and gaseous phases.

B. Experimental Section and Results

Except for the presence of a small amount of toluene employed as solvent for its cystallization, the C$_{76}$ specimen used for this investigation obatined from Technocarbo (France) was 99.9% pure with respect to other fullerenes as checked chromatographically by the supplier.

a. X-ray Measurements. X-ray diffraction photographs of powdered smallest crystals of the C$_{76}$ and C$_{84}$ fullerenes grown from toluene were recorded at room temperature on flat films by means of a Buerger precession camera, using Cu Kα radiation ($\lambda = 1.5418$ Å). The unit cell parameters were $a = 17.7$ Å, $b = 11.1$ Å, $c = 11.0$ Å, $\alpha = 90°$, $\beta = 108°$, $\gamma = 90°$ for C$_{76}$, and $a = 18.3$ Å, $b = 11.4$ Å, $c = 11.4$ Å, $\alpha = 90°$, $\beta = 108°$, $\gamma = 90°$ for C$_{84}$. The crystal data of C$_{76}$ were in agreement with those of Kawada et al.,[4] who proposed a $P2_1$ space group for C$_{76}$ and C$_{82}$ mainly on the basis of X-ray data collected for a C$_{82}$ single crystal. The observed and calculated spacings of the C$_{76}$ and C$_{84}$ reflections are reported in Table 1 together with some possible Miller indices, obtained by varying the h, k, and l values within the ranges 0−5, 0−4, and −4−4, respectively. The spacings of C$_{76}$ are calculated by means of the unit cell parameters given by Kawada et al.[4] ($a = 17.68$ Å, $b = 11.08$ Å, $c = 11.02$ Å, $\alpha = 90°$, $\beta = 108.1°$, $\gamma = 90°$). To determine the density of C$_{76}$ and C$_{84}$, powdered crystals were suspended in a mixture of methanol and CsCl aqueous solution, the density of which was determined by a picnometer. The measured density values of 1.62 g cm^{-3} for both C$_{76}$ and C$_{84}$ coincided with those calculated assuming two fullerene and two toluene molecules in the above-mentioned unit cells. The presence of toluene was supported by a weight loss during the heating of the crystals which changed their X-ray diffraction patterns. The reflections could be assigned to a major face-centered cubic (fcc) or to a minor hexagonal close-packed (hcp)

lattice as previously observed for C$_{76}$ from 372 to 573 K.[4] Using $a = 15.475$ Å for the fcc phase[1] and $a = b = 11.05$ Å, $c = 17.85$ Å, $\alpha = 90°$, $\beta = 90°$, $\gamma = 120°$ for the hcp phase, the observed reflections of a sample heated at about 500 K could be indexed (Table 2). A similar behavior (Table 2) was displayed by C$_{84}$ that was heated at 523 and 823 K ($a = 16.0$ Å for the fcc phase and $a = b = 11.4$ Å, $c = 18.5$ Å, $\alpha = 90°$, $\beta = 90°$, $\gamma = 120°$ for the hcp phase). The hcp phases of C$_{76}$ and C$_{84}$ are strictly related to the monoclinic ones grown by crystallization from toluene. No temperature dependence of the diffraction patterns up to 820 K was observed. Therefore, the phase transition starting at around 370 K is irreversible, owing to the sublimation of toluene, which could form inclusion compounds with C$_{76}$ and C$_{84}$.

b. Vapor Pressure. The vapor pressure of C$_{76}$ was measured by the torsion effusion method by employing the same assembly described in detail elsewhere.[5] Three conventional graphite torsion cells, having different size effusion holes (cell A, B, and C with 0.3, 1.0, and 1.5 mm, respectively nominal diameters), were used. The instrument calibration constants of these cells were determined by vaporizing pure lead and silver, which have well-known vapor pressures.[6] As the sample temperatures were indirectly measured by a calibrated Pt−Pt· 10%Rh thermocouple inserted in a fixed blank cell placed close below the torsion cell containing the sample, the correct temperature was determined following a standard procedure as described in a previous work,[7] employing as standard very pure zinc, having detectable vapor pressure at the melting point. With this correction the uncertainty in the temperature values should not exceed ±2 K. As concerns the error in the absolute vapor pressure, the uncertainties connected to the instrument constant and the torsion angle measurements produced a displacement in the log p values of about ±0.08.

At some temperatures, the molecular weight of the vapor was evaluated through the Knudsen equation[8] by measuring simultaneously the rate of weight loss of the sample and the vapor pressure. To evaluate the rate of weight loss of the sample, the torsion assembly was suspended from an arm of a vacuum balance (Cahn 1000). The average value of the molecular weight so determined (900 ± 50) indicates that C$_{76}$ sublimes congruently as the other studied fullerenes.

As observed during the vaporization of C$_{84}$,[3] when in a preliminary run a C$_{76}$ sample was heated at temperatures around 500 K, a weight loss of about 5−7% of its original mass (probably due to the evaporation of the solvent residue and/or of unidentified impurities, as for example mixtures of hydrocarbons produced during the preparation of the sample) occurred.

TABLE 1: Observed (do) and Calculated (dc) Spacings (Å) and Miller Indices in Parentheses of the Reflections of the C_{76} and C_{84} Samples Grown from Toluene

compound	d_0	d_c	d_c	d_c	d_c	d_c
C_{76}	16.7_8	16.81 (1,0,0)				
	10.5_1	10.47 (0,0,1)	10.47 (1,0,−1)			
	$9.2_0{}^a$	9.25 (1,1,0)				
	8.4_0	8.40 (2,0,0)				
	6.4_2	6.41 (1,1,1)	6.41 (2,1,−1)			
	5.5_1	5.54 (0,2,0)	5.51 (1,0,−2)			
	5.1_1	5.10 (2,1,1)	5.09 (3,1,−1)			
	4.9_1	4.90 (0,2,1)	4.90 (1,2,−1)			
	4.7_4	4.73 (0,1,2)	4.73 (2,1,−2)			
	4.6_4	4.63 (2,2,0)				
	4.4_0	4.40 (3,0,1)	4.40 (4,0,−1)			
	4.2_7	4.26 (1,1,2)	4.25 (3,1,−2)			
	3.9_2	3.91 (1,2,−2)	3.93 (2,0,2)			
C_{84}	17.4_0	17.40 (1,0,0)				
	10.8_4	10.84 (0,0,1)	10.83 (1,0,−1)			
	9.5_3	9.54 (1,1,0)				
	7.8_2	7.85 (1,1,−1)				
	6.6_0	6.61 (2,1,−1)	6.63 (1,1,1)			
	5.7_1	5.70 (0,2,0)	5.70 (1,0,−2)			
	5.2_9	5.27 (2,1,1)	5.26 (3,1,−1)			
	$4.7_0-4.9_9{}^a$	4.77 (2,2,0)	4.89 (2,1,−2)	4.90 (0,1,2)		
	4.0_2	4.03 (1,2,−2)				
	$3.5_5-3.6_3{}^a$	3.56 (3,2,1)	3.56 (4,2,−1)	3.58 (2,1,−3)	3.59 (0,3,−1)	3.59 (1,1,−3)
		3.59 (1,3,−1)	3.61 (0,0,3)	3.61 (3,0,−3)		
	3.4_5	3.44 (1,3,1)	3.44 (2,3,−1)	3.44 (3,1,−3)	3.45 (0,1,3)	3.45 (5,0,−2)
		3.46 (3,0,2)	3.46 (4,2,0)			
	3.3_1	3.31 (2,2,2)	3.31 (3,1,2)	3.31 (4,2,−2)	3.31 (5,1,−2)	
	3.2_0	3.20 (1,1,3)	3.20 (2,3,1)	3.20 (3,3,−1)	3.20 (4,1,−3)	
	3.0_8	3.08 (4,2,1)	3.08 (5,2,−1)			

^{a} Broad ring of strong intensity.

TABLE 2: Observed (d_o) and Calculated (d_c) Spacings (Å) and Miller Indices in Parentheses for the Reflections of the C_{76} and C_{84} fcc and hcp Mixed Phase

compound	d_0	d_c(fcc)	d_c(hcp)
C_{76}	9.5_6		9.57 (1,0,0)
	8.9_0	8.93 (1,1,1)	
	8.4_3		8.43 (1,0,1)
	7.7_3	7.74 (2,0,0)	
	6.5_1		6.53 (1,0,2)
	5.9_8		5.95 (0,0,3)
	5.4_5	5.47 (2,2,0)	
	5.0_4		5.05 (1,0,3)
	4.6_7	4.67 (3,1,1)	
	4.4_5	4.47 (2,2,2)	4.46 (0,0,4)
	3.5_6	3.55 (3,3,1)	3.54 (2,1,1); (−3,1,1); (−3,2,1)
			3.57 (0,0,5)
	3.4_5	3.46 (4,2,0)	3.47 (−2,1,4); (1,1,4)
	3.3_1	3.30 (3,3,2)	
	3.1_6	3.16 (4,2,2)	3.14 (3,0,1)
	2.9_7	2.98 (3,3,3)	
C_{84}	9.8_3		9.87 (1,0,0)
	9.2_6	9.24 (1,1,1)	
	8.7_1		8.71 (1,0,1)
	7.9_9	8.00 (2,0,0)	
	6.7_2		6.75 (1,0,2)
	6.1_7		6.17 (0,0,3)
	5.6_5	5.66 (2,2,0)	
	5.2_0		5.23 (1,0,3)
	4.8_1	4.82 (3,1,1)	
	4.6_4	4.62 (2,2,2)	4.63 (0,0,4)
	3.6_6	3.67 (3,3,1)	3.66 (2,1,1); (−3,1,1); (−3,2,1)
	3.5_7	3.58 (4,2,0)	3.59 (−2,1,4); (1,1,4)
	3.4_2	3.41 (3,3,2)	
	3.2_6	3.27 (4,2,2)	3.24 (3,0,1)
	3.0_7	3.08 (3,3,3)	

TABLE 3: Vapor Pressures of C_{76} Fullerene

cell	run	no. of points	$\log(p/kPa) = A - B/(T/K)$		
			ΔT (K)	A^a	B^a
A	1	8	934−1052	8.15 ± 0.19	10075 ± 189
B	2	7	865−952	7.95 ± 0.16	9926 ± 145
A	3^b	5	983−1069	8.27 ± 0.25	10165 ± 256
B	4	7	854−946	8.25 ± 0.07	10216 ± 63
C	5^b	9	851−921	8.58 ± 0.19	10415 ± 164
C	6^b	8	834−954	8.12 ± 0.25	10073 ± 223

^{a} The quoted errors are the standard deviations. ^{b} Run carried out by using recondensed vapor of C_{76} (see text).

On further heating of the sample its vapor pressure becomes detectable with our torsion assembly at about 800 K. The pressure values measured in the same heating and cooling cycles are well reproducible fitting a log p vs $1/T$ line. When approximately 60−70% of the sample was vaporized, the pressure values began to be nonreproducible, slowly decreasing below the limit of detectability of the instrument. The residue, which ranges from 2 to 6% of the original sample as evaluated in the successive vaporization runs, did not present detectable vapor pressure up to about 1300 K (upper limit of our furnace), and appeared to be amorphous carbon black. This form of carbon is present in small amounts in the original samples, as observed by SEM analysis, but could as well be produced during the sublimation. To test this hypothesis, a C_{76} sample was sublimated under vacuum in a quartz tube and the condensed vapor was employed as "purified" C_{76} sample in a torsion experiment. The vaporization of this sample showed that (i) no appreciable weight loss occurred at 500 K, (ii) the vapor pressure values fitted well on the line obtained when the original sample was used, (iii) the pressure data were reproducible up to the practically complete evaporation of the sample, and (iv) no detectable residue remained in the cell at the end of the vaporization. These observations lead to the conclusion that apparently all the amorphous carbon residue observed at the end of the vaporization is present in the original sample. In any case, considering the good reproducibility of the data obtained in the previous experiments, the presence of carbon in the condensed phase does not influence the sublimation of the fullerene in the first step. Therefore, taking into account only the vapor pressures measured in the first step of each vaporization run, a log p vs $1/T$ equation obtained by a linear

TABLE 4: Standard Molar Thermodynamic Functions of $C_{76}(g)$ and $C_{84}(g)$

	$C_{76}(g)$			$C_{84}(g)$		
T (K)	$S°$ (T) (J K^{-1} mol^{-1})	$H°(T) - H°(298)$ (kJ mol^{-1})	$[G°(T) - H°(298)]/T$ (J K^{-1} mol^{-1})	$S°(T)$ (J K^{-1} mol^{-1})	$H°(T) - H°(298)$ (kJ mol^{-1})	$[G°(T) - H°(298)]/T$ (J K^{-1} mol^{-1})
298.15	660.622	0.000	660.622	720.875	0.000	720.875
100	328.037	−70.021	1028.247	339.123	−79.731	1136.430
200	467.172	−48.491	709.625	503.799	−54.336	775.477
300	664.573	1.182	660.634	725.229	1.302	720.889
400	885.565	78.617	689.023	966.473	85.807	751.955
500	1109.452	179.317	750.818	1209.092	194.948	819.196
600	1325.871	298.223	828.832	1443.642	323.860	903.876
700	1530.361	430.985	914.668	1665.762	468.125	997.013
800	1721.580	574.237	1003.783	1873.994	624.182	1093.767
900	1899.693	725.479	1093.606	2068.399	789.313	1191.384
1000	2065.522	882.875	1182.648	2249.744	961.486	1288.258
1100	2220.118	1045.072	1270.052	2419.074	1139.188	1383.449
1200	2364.560	1211.066	1355.338	2577.492	1321.282	1476.424
1300	2499.871	1380.105	1438.252	2726.057	1506.910	1566.896
1400	2626.980	1551.614	1518.685	2865.742	1695.415	1654.732
1500	2746.714	1725.151	1596.614	2997.423	1886.288	1739.897
1600	2859.803	1900.370	1672.071	3121.873	2079.129	1822.418
1700	2966.887	2076.999	1745.122	3239.780	2273.623	1902.354
1800	3068.529	2254.819	1815.851	3351.746	2469.517	1979.792
1900	3165.222	2433.655	1884.351	3458.303	2666.604	2054.827
2000	3257.401	2613.362	1950.720	3559.920	2864.719	2127.561

TABLE 5: Temperature Dependence of Vapor Pressures of C_{60}, C_{70}, C_{76} and C_{84} Fullerenes and Their Sublimation Enthalpies

		$\log(p/kPa) = A - B/(T/K)$		II law		III law
fullerene	ΔT (K)	A	B	$\Delta_{sub}H°$(T) (kJ mol^{-1})	$\Delta_{sub}H°$ (298) (kJ mol^{-1})	$\Delta_{sub}H°$ (298) (kJ mol^{-1})
C_{60}[1]	730−990	8.28 ± 0.20	9154 ± 150	175 ± 3	182 ± 3	171.6
C_{70}[2]	783−904	8.38 ± 0.15	9917 ± 160	190 ± 6	200 ± 6	
C_{76}	851−1069	8.23 ± 0.20	10150 ± 150	194 ± 4	206 ± 4	
C_{84}[3]	920−1190	7.92 ± 0.30	10950 ± 300	210 ± 6	225 ± 6	

least-squares fitting of the data was derived. The equations, referred to six sublimation experiments, are reported in Table 3. No evident dependence of the pressure data on the size of the effusion holes of the cells was observed. From these equations, the following one, representative of the vapor pressure of C_{76} in the range 834−1069 K, was selected:

$$\log(p/kPa) = (8.23 \pm 0.20) - (10150 \pm 150)/(T/K) \quad (1)$$

The associated errors were estimated taking into account the uncertainties in the pressure and temperature values reported above.

c. Thermodynamics Properties. The enthalpy increments and the free energy functions of the gaseous molecule C_{84} were calculated by the methods of statistical thermodynamics in the harmonic oscillator−rigid rotator approximation.[9] No electronic contribution was included in the evaluation. Two nearly isoenergetic structures of the 24 isolated pentagon isomers are predicted by quantum chemical studies to be the most stable ones[11−15] (22 and 23 in the general tabulation[10] with D_2 and D_{2d} symmetry). These two isomers were taken into account in the evaluation of the rotational and vibrational contributions. As far as the rotational one is concerned, its value comes out to be almost identical irrespectively of the isomer considered, the principal moments of the most stable form D_2 being $I_A = 19.106 \times 10^{-44}$ kg m^2, $I_B = 19.751 \times 10^{-44}$ kg m^2, and $I_C = 20.380 \times 10^{-44}$ kg m^2. A number of quantum chemical calculations of the vibrational frequencies of these isomers have been performed in the literature.[11,16,17] Among these we selected the tight-binding potential model evaluations of Zhang et al.[16] because the resulting vibrational contribution appeared to be more in line with those calculated for the C_{60} and C_{70} in our previous works.[1,2] Finally, an average of the vibrational contributions of the D_2 and D_{2d} isomers calculated with selected frequencies provided by Zhang et al.[16] has been

adopted. The resulting entropies, enthalpy increments, and free energy functions are reported in Table 4.

For the C_{76} gaseous molecule, the reported most stable D_2 symmetry structure[18,19] was considered. No electronic contribution was included in the evaluation. The translational and rotational contributions were calculated by using the usual relations[9] ($I_A = 14.494 \times 10^{-44}$ kg m^2, $I_B = 16.407 \times 10^{-44}$ kg m^2, and $I_C = 17.875 \times 10^{-44}$ kg m^2). As for the vibrational one we preferred to estimate this contribution by interpolating those of C_{60}, C_{70}, and C_{84} when plotted as a function of the corresponding number of normal vibrations. Table 4 collects these derived thermodynamic functions.

The thermodynamic functions of the solid phases were not evaluated because of the complete absence in the literature of any relevant experimental data. Nevertheless, as reported later, it has been possible to estimate the difference in the enthalpy increments of the solid and gaseous phases.

C. Discussion and Conclusion

The selected eq 1 referred to the temperature dependence of the vapor pressure of C_{76} is reported in Table 5 and in Figure 2 together with those of the other studied fullerenes. From the slope of the straight line, the second-law sublimation enthalpy of the C_{76} mixed phase (fcc and hcp), $\Delta_{sub}H°(910) = 194 \pm 4$ kJ mol^{-1}, was derived.

It is interesting to note that by combining the standard sublimation Gibbs energy changes of C_{60}, C_{70}, C_{76}, and C_{84} evaluated at 900 K (a common temperature included in the temperature ranges covered in the pressure measurements[1−3]), with their sublimation enthalpies at this temperature (181 ± 2, 200 ± 6, 194 ± 4, and 210 ± 6 kJ mol^{-1} going from C_{60} to C_{84}), the corresponding sublimation entropies of these fullerenes ($\Delta_{sub}S°(900) = 120, 122, 118$, and 114 kJ mol^{-1} K^{-1} going from C_{60} to C_{84}) were derived. These entropy values are practically similar within their uncertainties, estimated about 5
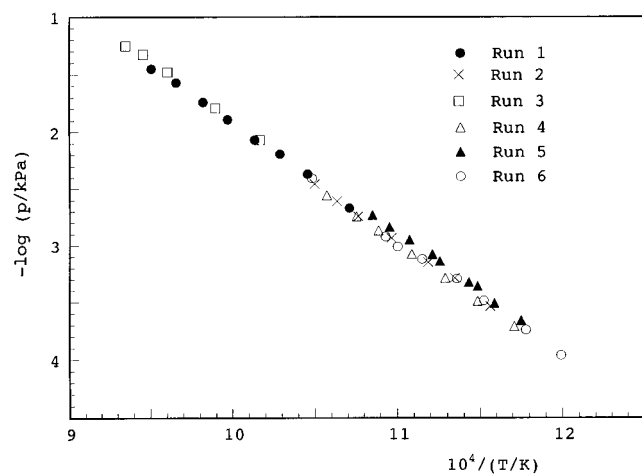
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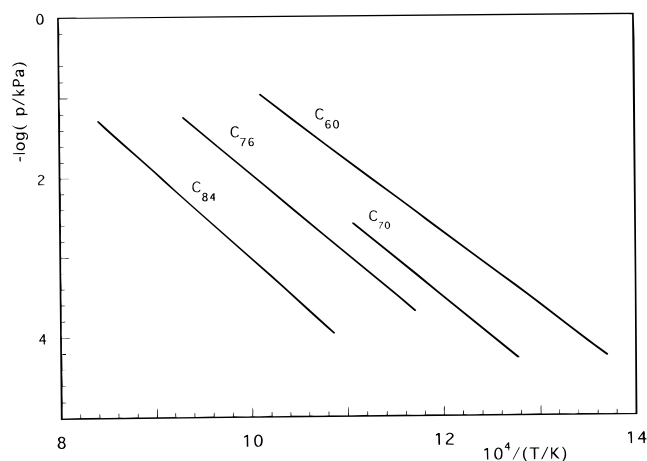**Figure 1.** Vapor pressure of C_{76}.

Figure 2. Comparison of the vapor pressure of C_{60},[1] C_{70},[2] C_{76}, and C_{84}[3] fullerenes.

kJ mol^{-1} K^{-1}. Incidentally, as concerns our previous work on the sublimation of C_{84},[3] regretfully, we missed a previous mass-spectrometric investigation by Moalem et al.[20] These authors report an estimated temperature dependence equation of the vapor pressure for C_{84} yielding vapor pressure values which, within the experimental uncertainties, fairly agree with our absolute values. But, the derived sublimation enthalpy (247 ± 25 kJ mol^{-1}) is higher than ours. We believe that the value proposed by Moalem et al.[20] should definitely be considered an upper limit in view of the large spread of the limited number of experimental points (eight) from which it was derived.

As no thermodynamic data of solid C_{76} and C_{84} were available in the literature, their second-law sublimation enthalpies, determined in the present work for C_{76} and in the previous work[3] for C_{84} ($\Delta_{sub}H°(1050) = 210 \pm 6$ kJ mol^{-1}), were reduced to 298 K employing enthalpy increment values (12 and 15 kJ mol^{-1}, respectively) estimated considering the increments used in fullerenes C_{60} and C_{70} (7 and 10 kJ mol^{-1} respectively). The obtained values, $\Delta_{sub}H°(298) = 206 \pm 4$, and 225 ± 6 kJ mol^{-1} for C_{76} and C_{84}, are reported in Table 5 for comparison with those previously determined for C_{60} and C_{70}. The values present an interesting trend with their molecular weights. As for the enthalpy increments, also for the free energy functions (fef), $[G°(T) - H°(298)]/T$, no data for solid C_{76} and C_{84} are available in the literature. Therefore, using the second-law $\Delta_{sub}H°(298)$ values of both fullerenes and the vapor pressures calculated at five temperatures at 100 K intervals from 800 to 1200 K by eq 1 for C_{76} and by the equation $\log(p/kPa) = (7.92 \pm 0.30) - (10950 \pm 300)/(T/K)$ for C_{84},[3] the Δ(fef) values associated with the sublimation of C_{76} and C_{84} at these temperatures were derived. The results are reported in Table 6. From the fef

TABLE 6: Free Energy Functions (in J·K^{-1}·mol^{-1}) of Solid C_{76} and C_{84} Obtained from the Third-law Treatment of Their Vapor Pressures

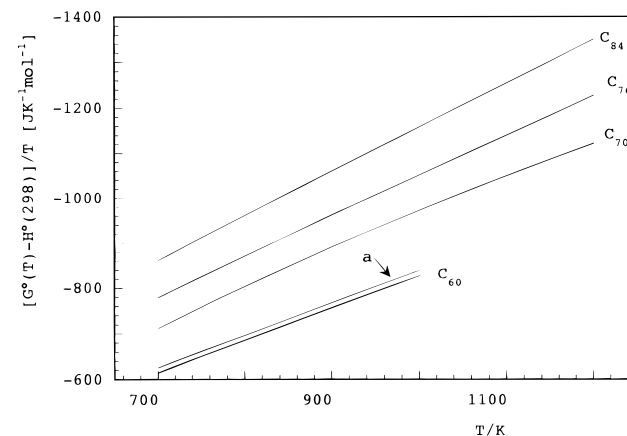|	C_{76} [$\Delta H°(298) = 205$ kJ mol^{-1}]			C_{84} [$\Delta H°(298) = 225$ kJ mol^{-1}]		
T (K)	p/kPa	Δ(fef)	fef (s)	p/kPa	Δ(fef)	fef (s)
800	3.5×10^{-5}	−132.4	−871.2	1.7×10^{-6}	−132.4	−961.4
900	8.9×10^{-4}	−130.9	−962.6	5.7×10^{-5}	−130.2	−1061.1
1000	1.2×10^{-2}	−129.7	−1053.0	9.3×10^{-4}	−128.5	−1159.7
1100	1.0×10^{-1}	−128.8	−1141.5	9.2×10^{-3}	−127.1	−1256.3
1200	5.9×10^{-1}	−127.9	−1227.7	6.2×10^{-3}	−126.0	−1350.7

Figure 3. Free energy function of solid fullerenes evaluated by a third-law procedure (see text); (a) Korobov's[21] data.

values of the gaseous C_{76} and C_{84} calculated in the present work (section c), the corresponding fef values of both solid fullerenes were derived and reported in Table 6 and in Figure 3 together with those of C_{60}(s) and C_{70}(s), determined in the previous works.[1,2]

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