Temperature dependence of the free energy of sorption of *n*-alkanes and energy contribution of their methylene groups on columns with fullerene C_{60}

R. V. Golovnya, M. B. Terenina, and A. L. Samusenko*

N. M. Emanuel' Institute of Biochemical Physics, Russian Academy of Sciences,
28 ul. Vavilova, 117813 Moscow, Russian Federation.
Fax: +7 (095) 137 4101

The partial molar free energy, enthalpy, and entropy of sorption of C_{11}—C_{23} *n*-alkanes were calculated on the basis of the GC data obtained on the glass capillary column coated with fullerene C_{60} (Ful-60) as stationary phase. The thermodynamic parameters of *n*-alkane sorption on a column with Ful-60 and a fused silica capillary column with polydimethylsiloxane OV-1 were determined and compared. The enthalpy-entropy compensation effect for the sorption of *n*-alkanes on Ful-60 and OV-1 was found. A linear dependence of the partial molar free energy of *n*-alkane sorption on the temperature of analysis and carbon chain length was found. The free energy contributions of the methylene groups were calculated, and their temperature dependences were studied. The differences in the temperature dependences of the energy contributions of methylene groups of *n*-alkanes on Ful-60 and OV-1 were revealed. The entropy contribution is 68—82% of the enthalpy contribution which indicates a substantial role of the number of contacts with Ful-60 in retention of *n*-alkanes. The ability of Ful-60 for dispersive interactions is similar to those of nonpolar liquid phases and substantially differs from that for carbon adsorbents. Fullerene columns were shown to be convenient for analysis of highly boiling organic substances in aqueous and organic solutions.

Key words: capillary gas chromatography, fullerene C_{60} as stationary phase, *n*-alkanes, partial molar free energy of sorption, enthalpy, entropy, energy contribution of methylene group, temperature dependence.

Fullerene is a promising stationary phase in chromatography due to the spherical form of the fullerene C_{60} molecule, its thermal stability, and ability for donor-acceptor interactions.[1—3] We were first to use fullerene as the stationary phase in gas chromatography.[4,5] Aqueous and organic solutions of aliphatic and aromatic hydrocarbons, cyclic amines, and alcohols were analyzed on glass columns coated with fullerene C_{60} (Ful-60). It was shown that fullerene C_{60} by its character of dispersion interaction is closer to nonpolar liquid phases than to solid carbon sorbents. The partial molar free energies of sorption of the methylene unit of *n*-alkanes on a capillary column with Ful-60 are the same as those on Apiezon L and twofold lower than on carbon sorbents.[4]

The abilities of fullerene C_{60} and graphitized carbon blacks for dispersive interactions under conditions of gas adsorption chromatography differ substantially.[6,7] Study on columns packed with Chromosorb 750 covered with a fullerene layer showed that fullerene differs in adsorption properties from graphitized carbon black. In particular, *n*-alkanes are retained less on fullerene than on Carbopack C HT.[6,7]

The fact that fullerene is not widely used as the stationary phase is associated with difficulties of obtaining uniform sorbent layers in a capillary and uniformly covered supports in packed columns.[4—7] This is due to a low solubility of fullerene in the major organic solvents.[8] Fullerene C_{60} is of interest for chromatography as a stationary phase that differs in its properties from traditional adsorbents and liquid stationary phases.

In this work, we studied the thermodynamic parameters of sorption of *n*-alkanes on capillary columns with Ful-60. The specific features of sorption of *n*-alkanes on columns with Ful-60 and with the stationary nonpolar polydimethylsiloxane liquid phase were compared.

Experimental

Fullerene C_{60} (purity 99.9%) was isolated from a C_{60}—C_{70} mixture by vacuum fractional sublimation. The purity of fullerene was monitored by UV spectroscopy. A glass capillary column coated with fullerene C_{60} (40 m × 0.30 mm) was prepared by the high-pressure static method.[9] Fullerene was supported from the specially selected mixture of solvents (*n*-pentane—*n*-hexane—*n*-heptane—toluene). The thickness of the stationary phase layer (d_f) was 0.1 μm. A fused silica capillary column coated with OV-1 (25 m × 0.32 mm, d_f = 0.1 μm) (Nordion Instruments, Finland) was used as the

Translated from *Izvestiya Akademii Nauk. Seriya Khimicheskaya*, No. 8, pp. 1490—1495, August, 1999.

Table 1. Capacity factors (k') for *n*-alkanes at different temperatures of the column with Ful-60

n-Alkane	90 °C	100 °C	110 °C	120 °C	130 °C	140 °C	150 °C	160 °C	170 °C	180 °C	190 °C	200 °C
C_{11}	0.12	0.07	0.043	0.03	—	—	—	—	—	—	—	—
C_{12}	0.25	0.15	0.09	0.06	0.04	—	—	—	—	—	—	—
C_{13}	0.55	0.31	0.17	0.11	0.07	0.04	0.03	—	—	—	—	—
C_{14}	1.18	0.63	0.34	0.21	0.13	0.08	0.05	0.04	—	—	—	—
C_{15}	2.53	1.29	0.67	0.39	0.23	0.14	0.09	0.06	0.04	0.03	—	—
C_{16}	5.41	2.63	1.30	0.74	0.43	0.25	0.16	0.10	0.07	0.05	0.03	—
C_{17}	—	5.18	2.46	1.36	0.76	0.44	0.28	0.17	0.11	0.07	0.05	0.03
C_{18}	—	—	4.91	2.62	1.41	0.79	0.48	0.29	0.18	0.12	0.08	0.05
C_{19}	—	—	—	4.88	2.54	1.38	0.82	0.47	0.29	0.19	0.12	0.08
C_{20}	—	—	—	—	—	—	—	—	—	0.30	0.19	0.12
C_{21}	—	—	—	—	—	—	—	—	—	0.47	0.29	0.18
C_{22}	—	—	—	—	—	—	—	—	—	—	0.45	0.27
C_{23}	—	—	—	—	—	—	—	—	—	—	—	0.41

Table 2. Capacity factors (k') for *n*-alkanes at different temperatures of the column with OV-1

n-Alkane	90 °C	100 °C	120 °C	140 °C	160 °C	180 °C	200 °C
C_{11}	3.21	2.10	0.98	0.52	—	—	—
C_{12}	6.38	4.04	1.77	0.93	—	—	—
C_{13}	12.57	7.7	3.16	1.51	—	—	—
C_{14}	24.72	14.95	5.56	2.55	1.29	—	—
C_{15}	48.40	27.59	9.90	4.28	2.04	1.06	—
C_{16}	—	51.88	17.42	7.13	3.24	1.62	—
C_{17}	—	—	30.78	11.89	5.14	2.43	1.29
C_{18}	—	—	53.38	19.60	8.08	3.67	1.86
C_{19}	—	—	—	32.34	12.71	5.52	2.70
C_{20}	—	—	—	—	—	8.31	3.90
C_{21}	—	—	—	—	—	—	5.61
C_{22}	—	—	—	—	—	—	8.08
C_{23}	—	—	—	—	—	—	11.60

reference column. *n*-Alkanes with the number of carbon atoms from 11 to 23 were analyzed.

Gas chromatographic analysis was carried out on a Micromat-412 chromatograph (Finland) with a flame-ionization detector in the isothermic runs at temperatures of 90—200 °C. The temperatures of the injector and detector were 225 and 250 °C, respectively. The splitting of carrier gas (H_2) flows in front of the column was 1 : 50. The inlet pressure was 0.7 atm for the column with fullerene and 1.1 atm for the column with OV-1. Samples of an *n*-pentane solution of *n*-alkanes C_{11}—C_{23} with a volume of 0.1—0.2 μL were analyzed.

The "dead" time of the column was found from the time of elution of the nonsorbed gas, methane. The capacity factors k' (Tables 1 and 2) were used as retention parameters. The partial molar free energies of sorption of *n*-alkanes analyzed were calculated by the formula

$$\Delta G = -2.3RT\log(k'\beta), \qquad (1)$$

where R = 8.3143 J mol^{-1} deg^{-1} is the universal gas constant; T is the temperature of analysis, K; $k' = (t - t_0)/t_0$ is the capacity factor; t is the retention time of the substance; t_0 is the hold-up time of methane in the column; and β is the phase ratio calculated from the formula[10]

$$\beta = (d_c - 2d_f)^2/[4d_f(d_c - d_f)],$$

where d_c is the inner diameter of the column; and d_f is the thickness of the stationary phase layer. The standard deviation of the ΔG values calculated by formula (1) did not exceed ±0.050 kJ mol^{-1}.

The partial molar free energy of sorption per methylene unit ($\Delta G(CH_2)$) was calculated by the formula[11,12]

$$\Delta G(CH_2) = -2.3RT\log(t'_{n+1}/t'_n), \qquad (2)$$

where t'_{n+1} and t'_n are the adjusted retention times of homologs of *n*-alkanes that differ by one CH_2 group.

The enthalpy (ΔH) and entropy (ΔS) components of the partial molar free energy of sorption of *n*-alkanes for each stationary phase were calculated by solution of the system of equations (3) using ΔG values experimentally found at different temperatures corresponding to the region of the $\log k'$—$1/T$ linear function that was confirmed by computer processing:

$$\Delta G_1 = \Delta H - T_1\Delta S,$$

$$\Delta G_2 = \Delta H - T_2\Delta S. \qquad (3)$$

The function of $\log k'$ *vs* inverse temperature for *n*-alkanes C_{13}—C_{19} on Ful-60 is presented in Fig. 1.

Aqueous and benzene solutions of medicines (substances) were analyzed by the known method[13] on a capillary column coated with Ful-60 at linear temperature programming from 150 to 200 °C with a rate of 8 deg min^{-1}.

Results and Discussion

Chromatographic retention is determined by the energy of intermolecular interactions of analyzed substances with the liquid stationary phase or adsorbent. The partial molar free energy of sorption (ΔG) is a quantitative characteristic of these interactions, which are mainly dispersive for *n*-alkanes. The temperature dependence of ΔG for *n*-alkanes C_{11}—C_{23} on Ful-60 and OV-1 can give additional information on the specific features of the behavior of hydrocarbons on these

Table 3. Partial molar free energy of sorption of *n*-alkanes ($-\Delta G$/kJ mol^{-1}) at different temperatures of the column with Ful-60

n-Alkane	90 °C	100 °C	110 °C	120 °C	130 °C	140 °C	150 °C	160 °C	170 °C	180 °C	190 °C	200 °C
C_{11}	13.48	12.31	11.04	9.93	—	—	—	—	—	—	—	—
C_{12}	15.83	14.56	13.25	12.10	11.03	—	—	—	—	—	—	—
C_{13}	18.16	16.83	15.45	14.34	13.15	11.99	11.04	—	—	—	—	—
C_{14}	20.46	19.04	17.63	16.48	15.24	14.04	13.06	11.85	—	—	—	—
C_{15}	22.75	21.28	19.76	18.56	17.29	16.03	14.94	13.74	12.69	11.58	—	—
C_{16}	25.04	23.49	21.89	20.64	19.31	18.00	16.88	15.59	14.46	13.40	11.97	—
C_{17}	—	25.60	23.91	22.60	21.26	19.92	18.74	17.43	16.30	15.16	13.85	12.60
C_{18}	—	—	26.12	24.75	23.31	21.90	20.67	19.30	18.06	16.89	15.59	14.31
C_{19}	—	—	—	26.78	25.26	23.82	22.54	21.12	19.82	18.61	17.30	16.03
C_{20}	—	—	—	—	—	—	—	—	—	20.35	19.00	17.64
C_{21}	—	—	—	—	—	—	—	—	—	22.07	20.69	19.33
C_{22}	—	—	—	—	—	—	—	—	—	—	22.35	20.93
C_{23}	—	—	—	—	—	—	—	—	—	—	—	22.54

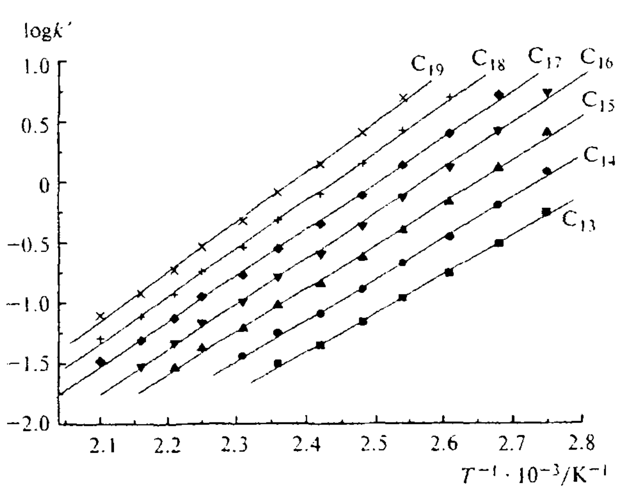

Fig. 1. Dependences of logk' on T^{-1} for *n*-alkanes C_{13}—C_{19} on a capillary column with Ful-60.

phases. The partial molar free energy of sorption of *n*-alkanes C_{11}—C_{23} in the studied temperature range depends linearly on the temperature of analysis for both phases. As the temperature of the column increases, the partial molar free energy of sorption of *n*-alkanes decreases (Tables 3 and 4). The ΔG values for *n*-alkanes on Ful-60 are lower than those on OV-1, which indicates a lower energy of the dispersive interaction of *n*-alkanes with Ful-60. At 120 °C the difference in the values of the partial molar free energy of sorption of *n*-alkanes on columns with OV-1 and Ful-60 is −10.5 kJ mol^{-1}. It increases as the temperature of the column increases and reaches ~13.5 kJ mol^{-1} already at 180 °C.

The partial molar free energy of sorption increases with increasing mass of *n*-alkane; linear dependences of the ΔG values on the number of carbon atoms of the *n*-alkanes are observed for Ful-60 and OV-1 (Fig. 2).

Table 4. Partial molar free energy of sorption of *n*-alkanes ($-\Delta G$/kJ mol^{-1}) at different temperatures of the column with OV-1

n-Alkane	90 °C	100 °C	120 °C	140 °C	160 °C	180 °C	200 °C
C_{11}	23.67	23.00	21.75	20.67	—	—	—
C_{12}	25.74	25.03	23.68	22.67	—	—	—
C_{13}	27.78	27.03	25.57	24.34	—	—	—
C_{14}	29.82	29.01	27.44	26.14	24.94	—	—
C_{15}	31.84	30.98	29.30	27.91	26.60	25.37	—
C_{16}	—	32.94	31.14	29.66	28.26	26.95	—
C_{17}	—	—	33.00	31.42	29.92	28.49	27.24
C_{18}	—	—	34.80	33.13	31.55	30.04	28.70
C_{19}	—	—	—	34.85	33.18	31.57	30.16
C_{20}	—	—	—	—	—	33.11	31.60
C_{21}	—	—	—	—	—	—	33.03
C_{22}	—	—	—	—	—	—	34.46
C_{23}	—	—	—	—	—	—	35.88

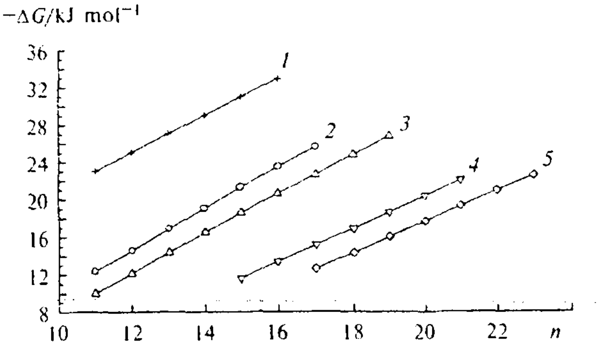

Fig. 2. Change in the partial molar free energy of sorption ($-\Delta G$) with increasing chain length of *n*-alkane (*n*) at 100 (*1*, *2*), 120 (*3*), 180 (*4*), and 200 °C (*5*) on capillary columns with OV-1 (*1*) and Ful-60 (*2*—*5*).

An increase in the temperature of the column results in a decrease in the energy contributions of methylene units of *n*-alkanes on capillary columns with Ful-60 and OV-1 (Tables 5 and 6). Comparison of sorption on Ful-60 and OV-1 shows that on fullerene *n*-alkanes are

Table 5. Contributions of the methylene unit ($-\Delta G(CH_2)$/kJ mol^{-1}) to the partial molar free energy of sorption of *n*-alkanes at different temperatures of a column with Ful-60

Number of methylene unit	90 °C	100 °C	110 °C	120 °C	130 °C	140 °C	150 °C	160 °C	170 °C	180 °C	190 °C	200 °C
12	2.35	2.25	2.21	2.17	—	—	—	—	—	—	—	—
13	2.33	2.27	2.20	2.14	2.12	—	—	—	—	—	—	—
14	2.30	2.23	2.18	2.14	2.09	2.05	2.02	—	—	—	—	—
15	2.29	2.22	2.13	2.08	2.05	1.99	1.88	1.89	—	—	—	—
16	2.29	2.21	2.13	2.08	2.02	1.97	1.94	1.86	1.82	1.82	—	—
17	—	2.11	2.02	1.96	1.95	1.92	1.86	1.83	1.81	1.76	1.88	—
18	—	—	—	—	—	—	—	—	1.78	1.75	1.77	1.75
19	—	—	—	—	—	—	-	—	1.76	1.73	1.71	1.72
20	—	—	—	—	—	—	—	—	—	1.72	1.71	1.62
21	—	—	—	—	—	—	—	—	—	1.72	1.69	1.68
22	—	—	—	—	—	—	—	—	—	—	1.66	1.61
23	—	—	—	—	—	—	—	—	—	—	—	1.60

Table 6. Contributions of the methylene unit ($-\Delta G(CH_2)$/kJ mol^{-1}) to the partial molar free energy of sorption of *n*-alkanes at different temperatures of a column with OV-1

Number of methylene unit	90 °C	100 °C	120 °C	140 °C	160 °C	180 °C	200 °C
12	2.07	2.03	1.93	2.00	—	—	—
13	2.05	2.00	1.89	1.67	—	—	—
14	2.04	1.98	1.87	1.79	—	—	—
15	2.03	1.98	1.86	1.77	1.67	—	—
16	—	1.96	1.85	1.76	1.66	1.58	—
17	—	—	1.86	1.76	1.66	1.54	—
18	—	—	1.80	1.72	1.63	1.55	1.46
19	—	—	—	1.72	1.63	1.54	1.46
20	—	—	—	—	—	1.54	1.45
21	—	—	—	—	—	—	1.43
22	—	—	—	—	—	—	1.43
23	—	—	—	—	—	—	1.42

Table 7. Enthalpies (ΔH) and entropies (ΔS) for homologs of *n*-alkanes on capillary columns with Ful-60 and OV-1

n-Alkane	$-\Delta H$/kJ mol^{-1}		$-\Delta S$/kJ mol^{-1} deg^{-1}	
	Ful-60	OV-1	Ful-60	OV-1
C_{11}	56.75	45.41	0.119	0.066
C_{12}	59.54	48.46	0.121	0.062
C_{13}	62.15	52.78	0.122	0.069
C_{14}	64.57	55.06	0.122	0.070
C_{15}	67.03	57.74	0.123	0.072
C_{16}	71.01	60.48	0.128	0.074
C_{17}	72.68	61.30	0.127	0.072
C_{18}	75.96	64.75	0.131	0.076
C_{19}	79.00	67.17	0.133	0.078
C_{20}	81.72	—	0.135	—
C_{21}	84.13	—	0.137	—

adsorbed with lower energies, but the energy contributions of each methylene unit are higher than those for sorption on OV-1. The $\Delta G(CH_2)$ values depend linearly on the temperature of analysis. The correlation coefficients of the linear dependence (R) range from 0.98 to 0.99, and the standard deviation ranges from 0.01 to 0.002 kJ mol^{-1}, which is within the error of determination. The temperature increments of methylene units of *n*-alkanes depend on the number of the homolog and differ for Ful-60 and OV-1.

It is of interest to compare the energy contributions of individual methylene units for Ful-60 and Ap-L calculated by the published data.[11] For example, at the temperature of analysis of 120 °C, the $\Delta G(CH_2)$ values for the 12th and 13th methylene units on Ap-L are −2.14 and −2.12 kJ mol^{-1}, and they are −2.17 and −2.14 kJ mol^{-1}, respectively, on Ful-60. Probably, the number of contacts of the sorbate with the sorbent has a considerably stronger effect on the dispersive interaction of higher *n*-alkanes with fullerene C_{60}. Thus, it follows from the results of our work and the data[4–7] on changing the ability for dispersive interactions that the sorbents are related by the following correlation: nonpolar liquid phases (OV-1, Ap-L, Squalane) > fullerene C_{60} < solid carbon supports.

We also estimated the contributions of the enthalpy and entropy components to the partial molar free energy of sorption of *n*-alkanes on columns with Ful-60 and OV-1. Since a linear dependence of logk' on $1/T$ (see Fig. 1) is observed on the column with Ful-60 and on traditional stationary phases for all *n*-alkanes, the ΔH and ΔS values for each *n*-alkane should be unchanged in the studied temperature range. The calculated values of the enthalpy and entropy components are presented in Table 7. Analysis of these data showed that the linear dependences of ΔH and ΔS on the homolog number are fulfilled for both columns with Ful-60 (R = 0.99 and 0.98, respectively) and columns with OV-1 (R = 0.99 and 0.92, respectively). As can be seen in Table 7, on

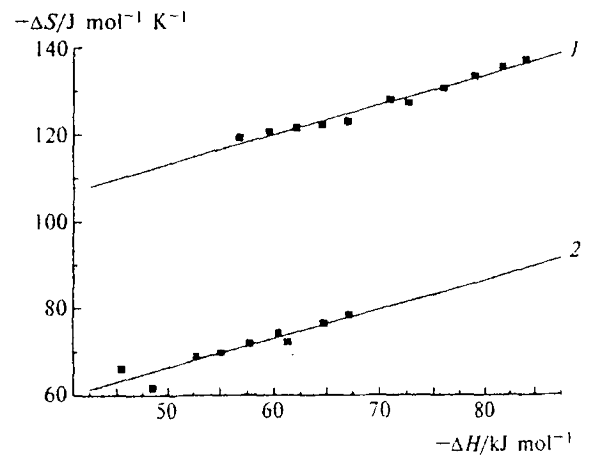

Fig. 3. Correlation between enthalpy ($-\Delta H$) and entropy ($-\Delta S$) of sorption of *n*-alkanes on capillary columns with Ful-60 (*1*) and OV-1 (*2*).

the column with Ful-60, ΔH for *n*-alkanes is higher than on the column with OV-1. The difference in enthalpies of *n*-alkanes on the two columns ranges from −9.29 to −11.83 kJ mol^{-1}, although the resulting free energy of sorption ΔG for Ful-60 is lower than for OV-1. This is related to the difference of the entropy factors on the columns compared. For example, the ΔS values for sorption on fullerene are almost twofold higher than those for sorption on polydimethylsiloxane.

We also compared the contributions of the enthalpy (ΔH) and entropy ($T\Delta S$) components to the partial molar free energy of sorption of *n*-alkanes on columns with Ful-60 and OV-1. At 120 °C the entropy contributions $T\Delta S$ for hydrocarbons C_{11}—C_{19} on the Ful-60 column are 68—82% of ΔH, and those on OV-1 are only 46—57%. Thus, for sorption of *n*-alkanes on fullerene, the contribution of the entropy component to ΔG is much higher than that for sorption on OV-1. Most likely, the structural factors play a more substantial role in sorption on columns with Ful-60, and the sorbate has a higher degree of freedom as compared to that for sorption on OV-1.

It is known[14,15] that hydrocarbons under gas chromatographic conditions exhibit the phenomenon of enthalpy-entropy compensation and a linear dependence

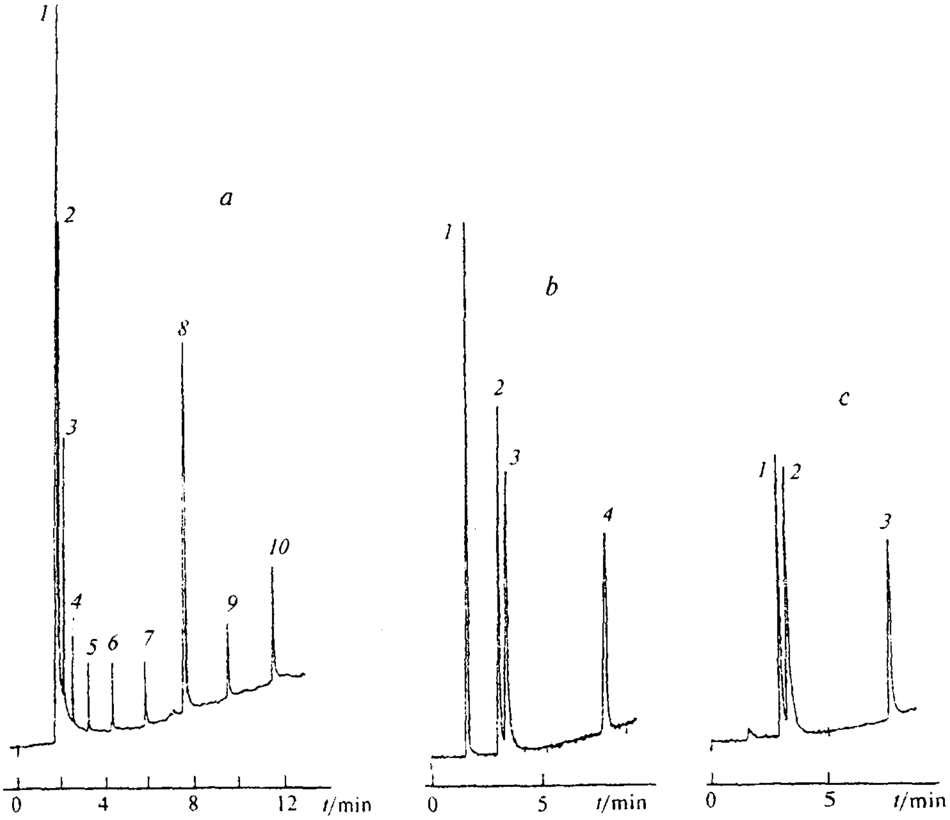

Fig. 4. Chromatograms of compounds on a glass capillary column with Ful-60. *a*: *1*, pentane; *2*—*10*, *n*-alkanes C_{11}—C_{19}, respectively; in pentane, temperature programming from 90 to 150 °C with a rate of 5 deg min^{-1}; *b*: *1*, benzene; *2*—*4*, medicines (substances) in benzene: Dinezin (10-(2-diethylaminoethyl)phenothiazine) (*2*), Dimedrol (β-dimethylaminoethyl ester of benzhydrol) (*3*), amidopyrine (1-phenyl-2,3-dimethyl-4-dimethylaminopyrazolone-5) (*4*), temperature programming from 150 to 200 °C with a rate of 8 deg min^{-1}; *c*: medicines in water: Dinezin (*1*), Dimedrol (*2*), and amidopyrine (*3*), temperature programming from 150 to 200 °C with a rate of 8 deg min^{-1}.

between ΔH and ΔS is fulfilled. For chromatography of *n*-alkanes on columns with Ful-60 and OV-1, we observed linear dependences of the free energy of sorption on the number of carbon atoms, enthalpy and entropy on the number of carbon atoms, and a linear dependence is observed between ΔH and ΔS (Fig. 3). The linear correlation for fullerene is more strictly fulfilled (R = 0.99) than that for polydimethylsiloxane (R = 0.94).

Due to low capacity factors (see Table 1), capillary columns with Ful-60 can be convenient for analysis of high-boiling compounds. Aqueous and organic solvents of various classes of organic compounds were analyzed on capillary columns coated with Ful-60.[4,5] The chromatograms of aqueous and benzene solutions of medicinal substances (Dinezin, Dimedrol, and Amidopyrine) and *n*-alkanes are presented in Fig. 4. Only 8 min are needed for analysis of these substances under conditions of temperature programming from 150 to 200 °C with a rate of 8 deg $\min^{-1}$, and *n*-alkanes are eluted as sufficiently symmetrical peaks. For glass capillary columns with Ful-60, no considerable changes in retention parameters and selectivity were observed over 12 months.

Thus, the study of thermodynamic parameters of sorption of *n*-alkanes shows that fullerene C_{60} is close in ability for dispersive interactions to nonpolar liquid phases and differs substantially from carbon sorbents. The thermodynamic sorption parameters indicate that the retention of *n*-alkanes on Ful-60 is mainly determined by the entropy rather than the enthalpy factor. It can be assumed that C_{60} in its adsorption properties occupies an intermediate position between nonpolar liquid phases and solid carbon sorbents.

The authors thank V. L. Karnatsevich for pure samples of fullerene C_{60}.

References

1. W. Kratschmer, L. D. Lamb, K. Fostinopouls, and D. R. Huffman, *Nature*, 1990, **347**, 354.
2. H. W. Kroto, A. W. Allaf, and S. P. Balm, *Chem. Rev.*, 1991, **91**, 1213.
3. G. A. Ozin, *Adv. Mater.*, 1992, **4**, 612.
4. R. V. Golovnya, M. B. Terenina, E. L. Ruchkina, and V. L. Karnatsevich, *Mendeleev Commun.*, 1993, **6**, 231.
5. R. V. Golovnya, M. B. Terenina, E. L. Ruchkina, and V. L. Karnatsevich, *Abstrs. Intern. Symp. Chromatogr. and Mass Spectrometr.*, in *Environmental Analysis*, St. Petersburg, 1994, 212.
6. V. Ya. Davydov, T. M. Roshchina, G. N. Filatova, and N. M. Khrustaleva, *Zh. Fiz. Khim.*, 1996, **70**, 1680 [*Russ. J. Phys. Chem.*, 1996, **70** (Engl. Transl.)].
7. V. Ya. Davydov, T. M. Roshchina, G. N. Filatova, and N. M. Khrustaleva, *Zh. Fiz. Khim.*, 1996, **70**, 1857 [*Russ. J. Phys. Chem.*, 1996, **70** (Engl. Transl.)].
8. R. S. Ruoff, D. S. Tse, R. Malhotra, and D. E. Lorents, *J. Phys. Chem.*, 1993, **97**, 3379.
9. E. A. Mistryukov, R. V. Golovnya, and A. L. Samusenko, *J. Chromatogr.*, 1978, **148**, 490.
10. R. V. Golovnya, *Zh. Anal. Khim.*, 1991, **46**, 12 [*J. Anal. Chem. USSR*, 1991, **46** (Engl. Transl.)].
11. R. V. Golovnya and D. N. Grigor'eva, *Izv. Akad. Nauk SSSR, Ser. Khim.*, 1983, 995 [*Bull. Acad. Sci. USSR, Div. Chem. Sci.*, 1983, **32**, 898 (Engl. Transl.)].
12. B. R. Kersten and C. F. Poole, *J. Chromatogr.*, 1987, **399**, 1.
13. I. L. Zhuravleva, M. B. Terenina, R. V. Golovnya, and M. A. Filimonova, *Khim.-Farm. Zh.* [*Chemical Pharmaceutical J.*], 1993, No. 5, 58 (in Russian).
14. J. Li and P. W. Carr, *J. Chromatogr.*, 1994, **670**, 105.
15. N. A. Katsanos, R. Thede, and F. Roubani-Kalantzopoulon, *J. Chromatogr.*, 1996, **795**, 133.

Received July 6, 1998;
in revised form December 29, 1998