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X-RAY DIFFRACTION INVESTIGATION OF INTERMOLECULAR ORDERING IN LIQUID BENZENE, CYCLOHEXANE AND n-HEXANE

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The paper reports results of an analysis of a correlation between the kind of compound (benzene, cyclohexane, n-hexane) and the type of close range molecular configurations (parallel, perpendicular, coplanar) in the liquid phase. The presence of such a correlation was indicated by X-ray diffraction measurements performed at 293 K using $CuK_{\overline{\alpha}}$ radiation $\lambda = 1.5418A$. Angular distributions of X-ray scattered intensity were measured, and electron density radial distribution functions were calculated. The mean distances between the neighbouring molecules and the mean coordination numbers were found. The differential functions of radial distribution of electron density $\sum_m \overline{K}_m 4\pi r^2(\rho(r)-\rho_0)$ were calculated and some intra- and intermolecular distances in liquid benzene, cyclohexane, and n-hexane were determined. The most probable models of local ordering of these molecules were suggested.

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1 Introduction

Benzene C_6H_6 (melting point 5°C, boiling point 80°C) has been studied in the gas [1], liquid [2] and crystalline [3] phase. It has been established that its atoms are arranged in plane and the admissible deviation of carbon atoms from the plane is 0.0013 A [3]. The mean distance between carbon atoms in the ring is C(1) - C(2) = 1.39 A, while the C - H distance is 1.09 A [3]. In order to determine the most probable mutual arrangement of molecules in liquid benzene it was assumed that the benzene ring has a D_{6h} symmetry. An elementary cell of a crystalline benzene belonging to the rhombic system ($a = 7.460$, $b = 9.666$, $c = 7.034$A, space group Pbca) contains 4 molecules.

An elementary cell crystalline cyclohexane C_6H_{12} (melting point 7°C, boiling point 81°C) belonging to the monoclinic cell, at 115 K, $a = 11.23(3)$, $b = 6.44(2)$, $c = 8.20(2)$A, $\beta = 108.83(17)^o$, $V = 561(5)A^3$ for $Z = 4$ [4]. Carbon atoms in cyclohexane undergo sp^3 hybridisation [5]. A cyclohexane molecule has both σ_3 and δ_3 symmetry. The length of C(1) - C(2) bond is 1.55A while that of C - H bond 1.10A. Cyclohexane has also been studied in liquid phase [6].

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Hexane CH_3-$(CH_2)_4$-CH_3 (melting point -95^{o}C, boiling point 69^{o}C) has been studied in solid [7] and liquid [8] phase, its characteristic feature is the zero dipole moment $\mu = 0$. An elementary cell of hexane ($P\overline{1}$) belonging to the triclinic system ($a = 4.19$, $b = 4.75$, $c = 8.62$A; $\alpha = 97^o$, $\beta = 85.6^o$, $\gamma = 105.0^o$) comprises a single molecule [7].The mean distance between carbon atoms in the chain was C(1) - C(2) = 1.54A and the distance C(1) - H(11) = 1.12 A [8]. Owing to the sp^3 type hybridisation of carbon atoms, the angles between the bond directions are of about 109^{o}28'.

The method of X-ray scattering and Fourier analysis [9] enabled determination of the mean structural parameters (inter- and intramolecular distances, radii coordination spheres, coordination numbers and molecule packing coefficients) of the liquids studied. The results were used to propose models of local ordering in these liquids. No comparative analysis of ordering in these liquids has been made. In our studies we used benzene and cyclohexane produced by Aldrich-Chemie (Germany) and n-hexane made by Janssen-Chimica (Belgium), all reagents were 99 % pure.

2 Experimental

Structural studies were conducted using an X-ray diffractometer equipped with a horizontal counter goniometer HZG3. The source of $CuK_{\overline{\alpha}}$ radiation was a lamp working at the optimum voltage $U = 40$ kV and current $I = 30$ mA. The radiation was monochromatised by reflection from the (002) planes of flat graphite [10,11] with the angle of monochromatisation of $\alpha = 13^o$ 09' [12].

Measurements were performed for the angles in the range $3^o \leq \theta \leq 60^o$at every $(0.2 \pm 0.01)^o$, where 2θ is the angle of scattering, by the transmission method [13]. Intensity of the scattered radiation was measured by a proportional counter VA-G-120, coupled with the calculating unit, and the results were recorded by a computer. Deviations in intensity due to instability of the diffractometer work were of about 1% in the whole range considered.

The studies were carried out with a special cuvette with exchangeable plates and closed 0.01 mm thick foil windows, constructed in our laboratory [9]. The material used and the thickness of the windows permitted us to neglect the absorption by the windows, which has been experimentally verified prior to measurements.

The cuvette was connected with a flow-through ultrathermostate U-10. Measurements were carried out at $(20.0 \pm 0.1)^o$C controlled by a copper-constantan thermo- couple and a sensitive mirror galvanometer GL-1. Inside the cuvette, temperature was measured by a temperature probe for the range $(-100$ to $+800 \pm 0.1)^o$C made by Testoterm GmbH&Co Lenzkirch, Germany.

The optimum thickness of the liquid layers to be studied was found from the expression [14]:

$$l \approx \frac{1}{\mu},$$

where μ is a linear absorption coefficient. These optimum thicknesses were: for benzene and cyclohexane 3 mm, while for n-hexane 3.5 mm. For each liquid measurements were performed for different times of counting and a few times repeated so that a final result was an average.

Liquid	$\overline{K}_H$	$\overline{K}_C$	$\sum_m K_m$	$\sum_m Z_m$	d[g/cm^3]	M[g/mol]	ρ_0[elA^{-3}]
benzene	0.652	6.369	42.003	42	0.879	78.12	0.285
cyclohexane	0.674	6.677	48.015	48	0.779	84.16	0.268
n-hexane	0.352	7.536	50.044	50	0.661	86.18	0.232

Table 1. Physical and structural parameters of liquid benzene, cyclohexane and n-hexane. $\sum_m Z_m$ denotes the sum of the atom nunmbers in one molecule.

3 Calculations

The radial distribution function of electron density $\rho(r)$ for a given liquid was determined from the modified Warren-Krutter-Morningstar equation [15] in the form

$$\sum_m \overline{K}_m 4\pi r^2 \rho(r) = \sum_m \overline{K}_m 4\pi r^2 \rho_0 + \frac{2r}{\pi} \int_0^\infty \mathrm{d}S S i(S) \sin(Sr), \tag{1}$$

where $K_m = f_m/f_e$ is the effective number of diffracting electrons in the atom m, and

$$f_e = \frac{\sum_m f_m}{\sum_m Z_m} \tag{2}$$

is the mean value of the atomic factor for a molecule per a single electron, f_m is the atomic factor of the m-th atom and Z_m is the number of electrons in the atom, see Table 1.

The mean effective number of scattering electrons was found from the integral average

$$\overline{K}_m = \frac{1}{S_2 - S_1} \int_{S_1}^{S_2} K_m \mathrm{d}S, \tag{3}$$

where S_1 and S_2 are the lower and upper bound of the range of S values. The result of Fourier transform is affected by the errors arising due to the limited range of intensity measurements. In the computer program for calculation of radial functions from the angular ones [9] we use the value of $S_{\max}$ matching the experimental conditions. In our experiment $S_1 = 0.43$ and $S_2 = 4\pi \sin\theta_{\max}/\lambda = 7.66\mathrm{A}^{-1}$ for the CuK$_{\overline{\alpha}}$ radiation. The value of $i(S)$ appearing in Eq. (1) is obtained from:

$$i(S) = \frac{1}{g^2(S)} \left[\frac{\overline{I}(S)}{N} - \sum_m f_m^2 \right]. \tag{4}$$

This expression represents the structurally sensitive part of the total coherent intensity $\overline{I}(S)/N$ per a single molecule, $\sum_m f_m^2$ denotes the theoretical independent scattering on atoms of one structural unit. According to Warren [16], $g(S) = f_e$, where f_e is defined by Eq. (2). In order to eliminate the erroneous undulations on the functions of density distribution originating from improper truncation of the experimental functions $\overline{I}(S)$, a temperature artificial factor was introduced [16] $\exp(-\alpha^2 S_{\max}^2)$. The radial distribution functions were calculated for $\alpha = 0.05$.

The mean electron density was found from the formula [14] :

$$\rho_0 = \frac{1}{M} \times 10^{-24} N_A d \sum_m Z_m \tag{5}$$

Fig. 1. The normalized functions of angular distribution of intensity obtained for (1) benzene, (2) cyclohexane and (3) n-hexane: (a) independent coherent scattering, (b) incoherent scattering, (c) total independent scattering.

where d is the macroscopic density of the liquid studied, N_A - the Avogadro constant, M - molar mass (see Table 1) and $\sum_m Z_m$ denotes the sum of the atom numbers in one molecule. The integral in Eq. (1) was calculated numerically by the Simpson method for $0 \leq \overline{r} \leq 20\mathrm{A}$ with a step of $\Delta\overline{r} = 0.05\mathrm{A}$.

The corrections for background [17], polarisation [18] and absorption [19] were taken into account in finding the angular distribution of the scattered radiation inten- sity. Experimental data were normalized according to the method of Krogh-Moe [20], Norman [21] and Wajnstejn [22]. The experimental values of scattered radiation intensity were corrected by the computer program [9,23] according to the scheme:

$$I = (I^{\mathrm{exp}} - I^{\mathrm{INC}} - I^{\mathrm{MULT}} - T)PA \tag{6}$$

where I^{exp} is the experimentally obtained intensity of scattered radiation, I^{INC} - the intensity of incoherent radiation, I^{MULT} - intensity of multiple scattering, T - the apparatus background and noise of the analysing system, P - the polarising factor, and A - the absorption factor.

The mean coordination numbers for the liquids studied were determined by the Pings method of symmetrisation of the radial functions [24] and the Hosemann method [25]. The latter one permits calculation of the coordination numbers free from the error due to truncation of experimental data. The maximum experimental error in determination of the radial distribution functions was estimated to be $\pm 3\%$ [26].

Fig. 2. Electron density radial distribution functions of liquid (1) benzene, (2) cyclohexane and (3) n-hexane.

Liquid	$\sin\theta/\lambda$ [A^{-1}]	$\overline{R}$ [A]	$\overline{r}$ [A]
benzene	0.106	5.51	5.15
cyclohexane	0.099	5.88	6.00
n-hexane	0.108	5.38	5.35

Table 2. The least intermolecular distances in the liquids studied; $\sin\theta/\lambda$ - the maxima of the angular intensity function, $\overline{R}$ - the mean least intermolecular distances, $\overline{r}$ - the radial distribution function maxima.

4 Results and discussion

The normalized functions $\overline{I}(S)$ of scattered radiation intensity obtained for the liquids studied are presented in Fig. 1. The positions of the main maxima on these functions were read out with an accuracy $\Delta S = \pm 0.01\mathrm{A}^{-1}$ and they were used to determine the mean least intermolecular distances $\overline{R}$ by the Voigtlaender-Tetzner method [27], see Table 2.

The functions of radial distributions of electron density found for benzene, cyclohexane and n-hexane at 20°C are presented in Fig. 2. They oscillate about the function of the mean electron distribution function and in the range from 0 to 10A reveal eight (benzene, cyclohexane) and seven (n-hexane) maxima. The first two of them on all functions correspond to intramolecular distances. The maxima $3 < \overline{r} < 6\mathrm{A}$ are ascribed to intra- and intermolecular diffraction.

The mean intermolecular distance for benzene $\overline{r}_3 = 4.10\mathrm{A}$, Fig. 2, corresponds to the case of plane - parallel molecular arrangement. Evans and Watts [28] in their Monte Carlo calculations of liquid benzene structure obtained the planar stacked arrangement of benzene molecules at a distance of $\overline{r} = 4\mathrm{A}$. The appearance of such a configuration they explained by the presence of a local minimum of the Lennard - Jones potential. Existence of this configuration at a distance $\overline{r} \approx 4.10\mathrm{A}$ was also postulated by Narten [2] and Bochyński [29], however only for a small number of molecular pairs.The mean number of molecules at this distance was established as 1.75. In the Narten model of liquid benzene "*The molecules pairs (...) fit together like six-toothed bevel gear wheels whose axes are almost at right angles*" [2]. Such an, arrangement of neighbouring molecules, resembling a letter L, helped interpretation of the maximum of the radial distribution at $\overline{r}_5 = 6.15\mathrm{A}$, Fig. 2 and Table 2. This is the only perpendicular configuration in which the centres of neighbouring molecules are at a distance of about 6 A, in the perpendicular configuration resembling a letter T this distance is $\overline{r}_4 = 5.10\mathrm{A}$. The mean molecular coordination numbers determined from the area under the radial distribution maxima are $Z_4 = 2.57$ and $Z_5 = 3.45$ [30].

These values are in agreement with the model proposed by Lowden and Chandler [31]. The perpendicular configuration may be favoured in liquid benzene because of the quadrupole moment of its molecule. The sixth maximum of the radial distribution function corresponds to the distances from the range 6.20 to 8.30 A beetwen neighbouring molecules. The mean most probable distance between the molecules in this range is $\overline{r}_6 = 7.20 \pm 0.12\mathrm{A}$. This value may indicate molecular arrangement close to the coplanar one [2]. Such a structure of liquid benzene was indicated by the Cotton-Mouton constant measurements [32], degree of depolarisation of scattered radiation [33] and temperature dependence of the optical Kerr effect [34].

On the radial distribution function of electron density obtained for cyclohexane, the maximum $\overline{r}_4 = 5.10\mathrm{A}$, Fig. 2, comes from the intermolecular interactions assuming the plane parallel arrangement of molecules. The radius of the first coordination sphere of intermolecular ordering $\overline{R} = 5.88 \pm 0.12\mathrm{A}$, determined from the intesity distribution function [27], corresponds to the fifth maximum for $\overline{r}_5 = 6.00\mathrm{A}$, Fig. 2, in the radial distribution function. It can be interpreted as a result of the interaction between mutually perpendicular molecules [6,37]. The sixth maximum on the radial distribution function was interpreted as corresponding to the molecules at a distance from 6.00 to 7.80 A from the reference molecule.The mean intermolecular distance is close to that among the coplanar side by side arranged molecules [5,37]. The mean coordination numbers corresponding to these maxima are $Z_4 = 1.43$, $Z_5 = 3.14$, and $Z_6 = 2.30$. The maximum at $\overline{r}_3 = 4.20\mathrm{A}$ proved difficult to interpret, most probably it comes from the interactions between atoms of neighbouring molecules.

In the radial distribution function of electron density by n-hexane, the maximum at $\overline{r}_3 = 4.25\mathrm{A}$, Fig. 2, corresponds to a configuration of molecules lying one next to the other with their axes parallel. Such a configuration for n-hexane was proposed by Stewart [35], Habenschuss and Narten [36]. The mean intermolecular distance in liquid n-hexane calculated from Voigtlaender-Tetzner equation $\overline{R} = 5.38\mathrm{A}$, (Table 2) coincides with the position of the fourth maximum of the function $\overline{r}_4 = 5.35\mathrm{A}$, Fig. 2. At this distance the molecules are in parallel orientation but are shifted relative to one another by about 2.5 A along the chain axis [9,36]. This shift increases the distance between the centres of molecules to the value corresponding to $\overline{r}_4$. Such anarrangement of n-hexane molecules would give high birefringence and significant anisotropy of thermal expansion [36].

Fig. 3.The differential radial distribution functions for (1) benzene, (2) cyclohexane and (3) n-hexane.

The position of the fifth maximum $\overline{r}_5 = 6.45\mathrm{A}$, corresponds to the perpendicular arrangement of neighbouring molecules, resembling a letter T [9]. The mean coordination number corresponding to these maxima are $Z_3 = 1.32$, $Z_4 = 2.27$, and $Z_5 = 2.55$. The mean coordination numbers determined in the first coordination sphere of intermolecular ordering are in agreement with those reported by other authors [37,38].

The last two maxima of the radial function of electron density for benzene $\overline{r}_7 \approx 8.15\mathrm{A}$ and $\overline{r}_8 \approx 9.15\mathrm{A}$, cyclohexane $\overline{r}_7 \approx 7.80\mathrm{A}$ and $\overline{r}_8 \approx 8.75\mathrm{A}$, and n-hexane $\overline{r}_6 \approx 8.45\mathrm{A}$ and $\overline{r}_7 \approx 9.40\mathrm{A}$, are attributed to the effect of further neighbours belonging to the second coordination sphere of intermolecular ordering [9].

The maxima on the differential functions of radial distribution, Fig. 3, have been ascribed to the following pairs of atoms: for benzene $C(1) - C(2) = 1.39\mathrm{A}$, $C(1) - C(3) = 2.40\mathrm{A}$, for cyclohexane $C(1) - C(2) = 1.55\mathrm{A}$, $C(1) - H(5) = 2.80\mathrm{A}$, and for n-hexane $C(1) - C(2) = 1.54\mathrm{A}$, $C(1) - H(3) = 2.75\mathrm{A}$. The mean least inter- and intramolecular distances were determined with the following accuracy : for $1 < \overline{r} \leq 2\mathrm{A}$: $\Delta\overline{r} = \pm 0.05\mathrm{A}$, for $2 < \overline{r} \leq 3\mathrm{A}$: $\Delta\overline{r} = \pm 0.10\mathrm{A}$, for $\overline{r} > 3\mathrm{A}$: $\Delta\overline{r} = \pm 0.12\mathrm{A}$ [39].

The differential functions of electron radial distribution, Fig. 3, reveal three broad maxima in the range $3.5 < \overline{r} < 20\mathrm{A}$, which have been ascribed to the three subsequent spheres of intermolecular ordering [40]. The spread of each sphere and the coordination numbers of molecules have been determined, see Table 3. Analysis of the shape and courses of the angular distributions of scattered radiation intensity (Fig. 1), radial distribution functions (Fig. 2) and differential radial distribution functions (Fig. 3), for all three compounds studied, leads to a conclusion that

Fig. 4. Mean coordination numbers for liquid benzene, cyclohexane and n-hexane, corresponding to the parallel Z (=), perpendicular Z (- I), and coplanar Z (–) arrangement of neighbouring molecules.

Liquid	Range [A] / Number of molecules		
	Sphere I	Sphere II	Sphere III
benzene	3.20 - 8.00 (10.6)	8.00 - 12.75 (36.2)	12.75 - 17.60 (95.3)
cyclohexane	3.50 - 8.50 (10.2)	8.50 - 13.60 (27.3)	13.60 - 18.50 (76.80)
n-hexane	3.15 - 7.70 (6.1)	7.70 - 11.60 (21.4)	11.60 - 16.20 (52.3)

Table 3. The range of coordination spheres and the number of molecules in each sphere for the studied liquids.

local orderings in these liquids are similar. The mean intermolecular distances (Table 2) and mean coordination numbers (Table 3) are close. In liquid benzene and cyclohexane the perpendicular configuration is dominant and makes about 56% and 46% of all possible arrangements, respectively. In liquid n-hexane the parallel configuration prevails and makes about 57% of all possible arrangements. A comparison of the data for the three compounds is shown in Fig. 4.

5 Conclusions

Results of X-ray diffraction studies and Fourier analysis proved that there is a correlation between the internal structure of liquids studied and the type of their local ordering . The appearance of clear maxima on the functions of angular distribution of X-ray radiation and radial distributions of electron density indicates the presence of short range ordering in these liquids. The molecules are arranged in parallel, perpendicular and coplanar configurations. In benzene the dominant is the perpendicular indent conformation, in cyclohexane the perpendicular one and in n-hexane the parallel one. The local structures of benzene and cyclohexane are similar.

In the range to about 18 A, three spheres of intermolecular ordering were distinguished. The maxima on the radial distribution functions in the range $1 < \overline{r}_0 < 3\mathrm{A}$ are attributed to intramolecular interactions. The maxima in the range $3 < \overline{r} < 5\mathrm{A}$, are interpreted as due to intra- and intermolecular diffraction, whereas those for $\overline{r} > 5\mathrm{A}$ are due to intermolecular diffraction.

The intermolecular distances determined by the method of Voigtlaender-Tetzner are in a good agreement with the maxima on the radial distribution functions.

The radial distribution functions of electron density are charged with a certain error following from the application of the integral Fourier formula derived for infinite limits of integration to an experimental situation in which measurements are conducted in a finite range of argument values. Replacement of the Fourier transform from Eq. (1) by the proper integral in Eq. (3) implies the appearance of additional oscillations for $0 \leq \overline{r} \leq 1\mathrm{A}$. These maxima have no physical meaning and are only a mathematical effect, a consequence of the applied approximations.

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