X-RAY ANALYSIS OF LIQUID 1-METHYLNAPHTHALENE STRUCTURE AT 293 K

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The paper reports results of the X-ray diffraction structural studies of liquid 1-methylnaphthalene, $C_{10}H_7-CH_3$ at 293 K, using Mo K_α radiation of the wavelength $\lambda = 0.71069$ Å. The interpretation of the results was carried out using the reduction method of Blum and Narten. Experimental distribution of X-ray scattered intensity was compared with theoretical results predicted for a proposed model of 1-methylnaphthalene molecule. The electron-density radial-distribution function $4\pi r^2 \sum_{j,k}^{n} \overline{K}_j[\rho_k(r) - \rho_0]$ was calculated and some intra- and intermolecular distances in liquid 1-methylnaphthalene were determined. The structural data obtainable by X-ray analysis for liquid 1-methylnaphthalene are discussed. X-ray structural analysis was applied to determine the packing coefficient of 1-methylnaphthalene molecules.

PACS numbers: 61.25.Em

1. Introduction

The aim of this paper is to report the results of the X-ray diffraction investigation of the liquid 1-methylnaphthalene $C_{10}H_7-CH_3$ (melting point $-22°C$, boiling point $243°C$), at room temperature. These results can be interpreted in terms of a simple model of local arrangement of molecules, which probably can be valid for a larger class of molecular liquids, i.e. weakly polar monosubstituted derivatives of naphthalene. Results of the study concerning molecular correlations in liquid phase may be important for explanation of mechanisms of certain physical and chemical processes taking place in such systems, e.g. diffusion or thermal conductivity [1]. The knowledge of the structure of naphthalene [2], 1-chloronaphthalene [3] and 1-methylnaphthalene allows to conclude about the structure of their weakly polar derivatives. X-ray diffraction is a very efficient method of studying the structure of matter in both solid and liquid phase [4]. However, to the best of our knowledge,

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no reports have been published on the structure of 1-methylnaphthalene in the crystalline state [5].

This work reports first studies of liquid 1-methylnaphthalene performed by the transmission method for the angular range of the intensity measurements extended to the value of $\vartheta = 60°$. The interpretation of the results was carried out using the reduction method [6, 7]. Structural parameters, the mean distances between the neighboring molecules as well as the coordination numbers were found. The most probable value of the packing coefficient of the molecules was found to be $k = 0.64$.

TABLE I

Physical and structural parameters of liquid 1-methylnaphthalene ($\sum_j Z_j$ denotes the sum of the atomic numbers in one molecule).

Mean effective number of electrons per		$\sum_j \overline{K}_j$	$\sum_j Z_j$	Macroscopic density [g/cm^3]	Molecular mass [g/mol]	Mean electron density [el/Å^3]
hydrogen atom $\overline{K}_H$	carbon atom $\overline{K}_C$					
0.650	6.318	76.000	76	1.001	142.20	0.3223

The values of some structural and physical parameters of liquid 1-methylnaphthalene are collected in Table I. Methylnaphthalene samples of 99% purity were purchased from Aldrich-Chemie (Germany).

2. Experimental

Intensity of radiation scattered by a layer of the studied liquid was determined by the transmission method using a typical X-ray diffractometer adapted to measurements of liquids, Fig. 1. The angular distribution of the X-ray scattered intensity was measured by applying Mo K_α ($\lambda = 0.71069$ Å) radiation for the angles $3° \leq \vartheta \leq 60°$ at every $0.2°$, where 2ϑ is the scattering angle. The radiation was monochromatized by reflection from the (002) planes of flat graphite with the angle of monochromatization of $\alpha = 6°00'$ [8]. The scattered X-ray intensities were measured by a VA-G-120 proportional counter (made by Otto Schön, Germany) 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 [9].

Samples of the 1-methylnaphthalene at 293 K were put into a cuvette with two mica windows limiting the liquid layer to 1 mm. The cuvette was connected with a flow-through ultrathermostate U-10. Fluctuation of temperature did not exceed ±0.2 K [10]. Absorption in the cuvette windows was neglected [11]. The temperature inside the cuvette was measured by a temperature probe made by Testoterm GmbH&Lenzkirch, Germany.
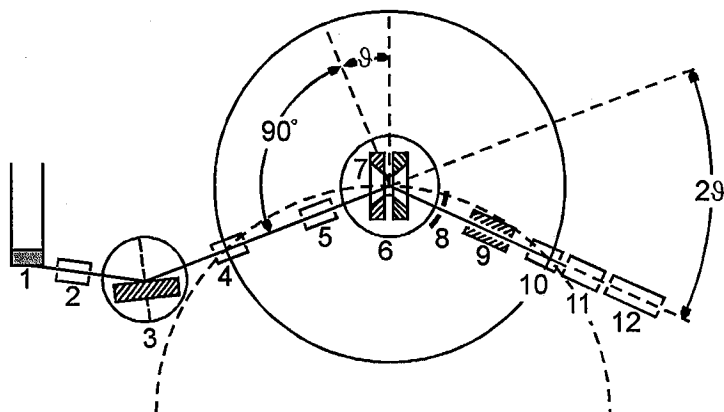
Fig. 1.   A scheme of a measuring system of a goniometer in the transmission method; 1 — X-ray tube anode, 2 — a system of output slits, 3 — monochromator, 4 — a system of input slits of the goniometer, 5 — slits controlling the width and divergence of the beam, 6 — the goniometer table, 7 — cell with the liquid studied, 8 — a shield against the scattered parasite radiation, 9 — Soller slits, 10 — a system of detector slits, 11 — radiation counter, 12 — electronic amplifier.

3. Calculations

The scattered X-radiation was normalized to electron units according to the Norman [12] method. The normalization methods applied are based on the fact that the interference effects have not been observed for large scattering angles. In this range, the experimentally observed intensity distribution of coherent scattering coincides with the distribution of intensity coherently scattered by independent individual atoms. Before normalization of the intensity curve, corrections for absorption [13], polarization [14], background [15] and anomalous dispersion [16] and incoherence of radiation [17] were applied.

The interpretation of the experimental results was performed by the method of the electron-density radial-distribution function (EDRDF) calculated from the equation [18]:

$$\sum_{j=1}^{n}\sum_{k=1}^{n}\overline{K}_j 4\pi r^2 \rho_k(r) = \sum_{j=1}^{n}\overline{K}_j 4\pi r^2 \rho_0$$

$$+\frac{2r}{\pi}\int_0^\infty \mathrm{Si}(S)\exp(-\alpha^2 S^2)\sin(Sr)\mathrm{d}S. \tag{1}$$

In Eq. (1) the function $i(S)$ is defined as

$$i(S) = [I_{\mathrm{eu}}(S)/N - \sum_{\mathrm{uc}} f_j^2(S)]/g^2(S), \tag{2}$$

where $\sum_{\mathrm{uc}}$ stands for summing over unit of composition, $I_{\mathrm{eu}}(S)/N$ is the experimentally observed total intensity of coherently scattered radiation per one structural unit, $\sum_{\mathrm{uc}} f_j^2(S)$ denotes the theoretical independent scattering on atoms of one structural unit, $g(S)$ is a sharpening factor, often written in the form: $\sum_{\mathrm{uc}} f_j$ or

$\sum_{uc} f_j / \sum_{uc} Z_j$ and $\exp(-\alpha^2 S^2)$ is a convergence factor, with coefficient $\alpha = 0.06$. The mean number of electrons in a unit volume (1 Å^3) is given by the formula [19]:

$$\rho_0 = dN_A \times 10^{-24} \sum_j Z_j / M, \tag{3}$$

where d is the macroscopic density of the liquid, N_A — the *Avogadro constant*, Z_j is the number of electrons in the j-th atom and M — *molar mass* (see Table I). The summation in Eq. (1) runs over all atoms of the molecule: $\sum_j \overline{K}_j = n_C \overline{K}_C + n_H \overline{K}_H$, where n_C and n_H are the numbers of carbon and hydrogen atoms, respectively, in one molecule, $\overline{K}_C$ and $\overline{K}_H$ are the average effective numbers of electrons in the atoms of carbon and hydrogen.

The mean effective number of diffracting electrons as found from the integral average

$$\overline{K}_j = \frac{1}{S_{max} - S_0} \int_{S_0}^{S_{max}} K_j dS, \tag{4}$$

where

$$K_j = \frac{f_j}{f_e} \tag{5}$$

is the effective number of electrons per atom j, and

$$f_e = \frac{\sum_j f_j}{\sum_j Z_j} \tag{6}$$

is the mean value of the atomic factor for a molecule per a single electron, f_j is the atomic factor of the j-th atom and Z_j is the number of electrons in the atom, $S = 4\pi \sin \vartheta / \lambda$, 2ϑ — the angle of scattering and λ — the X-ray scattering wavelength. The calculations were performed for a finite range of S values from $S_0 = 0.430 \text{ Å}^{-1}$ to $S_{max} = 14.311 \text{ Å}^{-1}$.

The least mean intermolecular distances were found from the Voigtlaender-
-Tetzner formula [20]:

$$\overline{R} = \frac{7.73}{4\pi \sin \vartheta / \lambda} - 0.3. \tag{7}$$

The correcting factor "0.3" for molecular liquids has been found empirically [20]. The maximum experimental error in determination of EDRDF was estimated to be $\pm 3\%$. The mean least inter- and intramolecular distances were determined with the following accuracy: for $1 < \overline{r} \leq 2 \text{ Å}$: $\Delta \overline{r} = \pm 0.01 \text{ Å}$, for $2 \leq \overline{r} \leq 3 \text{ Å}$: $\Delta \overline{r} = \pm 0.05 \text{ Å}$, for $\overline{r} > 3 \text{ Å}$: $\Delta \overline{r} = \pm 0.10 \text{ Å}$ [21]. Computer techniques were used to minimise the effects of experimental errors, uncertainties in the scattering factors, and termination errors.

4. Results

The normalized angular-distribution function $I(S)$, in electron units (where $S = 4\pi \sin \vartheta / \lambda$) of 1-methylnaphthalene (Fig. 2) is characterized by four general maxima which correspond to intramolecular interaction. The first maximum, at $S_1 = 1.20 \pm 0.01 \text{ Å}^{-1}$, less intense, is due to the distance of $\overline{R} = 6.14 \pm 0.10 \text{ Å}$,

between molecules and the next one $S_2 = 1.45 \pm 0.01$ Å^{-1} is due to the distance of $\overline{R} = 5.03 \pm 0.10$ Å. The third peak at $S_3 = 1.63 \pm 0.01$ Å^{-1} corresponds to the distance $\overline{R} = 4.44 \pm 0.10$ Å. The fourth maximum at $S_4 = 1.82$ Å corresponds to the distance $\overline{R} = 3.95$ Å. The positions of the S_1, S_2, S_3, and S_4 maxima were found using the Lagrange polynomials method. Small-angle scattering result ($0° < \vartheta < 3°$) was extrapolated to the origin of the coordinate system using a second-order function [22].
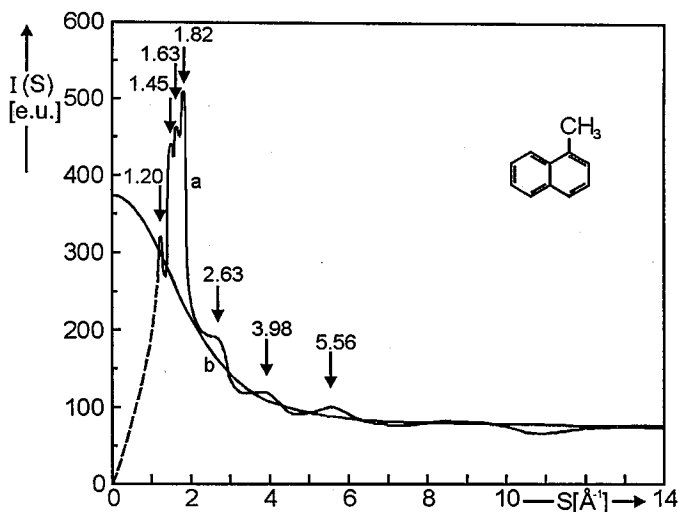

Fig. 2. *a* — normalized, experimental curve of angular distribution of X-ray scattered intensity; *b* — total independent scattering curve in liquid 1-methylnaphthalene.

Using the experimental values of $I(S)$, presented in Fig. 2 and Eq. (2), the values of $i(S)$, and the total function of the structure was calculated. By the reduction method [6, 7], the function $i(S)$ which is an important component of the Fourier integral in Eq. (1) can be separated from the total value of intensity $I_{eu}(S)/N$. In order to employ the reduction method, the function $i(S)$ which is now referred to as the total structure function must be redefined and expressed as the sum

$$i(S) = i_m(S) + i_d(S), \tag{8}$$

where $i_m(S)$ is the molecular structure function describing the scattering by a single molecule and $i_d(S)$ is the distinct structure function providing the information about intermolecular correlations from the experimental data.

The molecular structure function $i_m(S)$ is calculated from the equation [23]:

$$i_m(S) = \left[\sum_i f_i(S)\right]^{-2} \left[\sum_{uc}\sum_{i \neq j} f_i f_j \exp\left(-A_{ij}S^2\right) \frac{\sin\left(Sr_{ij}\right)}{Sr_{ij}}\right]. \tag{9}$$

In Eq. (9) the symbols i and j denote summation over stoichiometric unit; $A_{ij} = u_{ij}/2$, $u_{ij} = \left\langle(\Delta r_{ij})^2\right\rangle^{\frac{1}{2}}$ denoting the root-mean-square variation in the distance

r_{ij} between pairs of atoms. The calculated mean amplitudes of vibration (u_{ij}) for 1-methylnaphthalene by Mastryukov–Cyvin method [24–26] are shown in Table II. The numbering of atoms follows common usage in organic chemistry.

TABLE II

The values of parameters of 1-methylnaphthalene molecule model applied in the Debye formula, Eq. (9). Atom notation is the same as in Fig. 2 (u_{ij} denotes the root-mean-square variation in the distance r_{ij} between pairs of atoms [24, 25]).

Type of intramolecular interactions	Intramolecular distances r_{ij} [Å]	Mean amplitude u_{ij} [Å]
C_2-H	1.10	0.077
C_1-C_2	1.36	0.046
C_2-C_3	1.41	0.061
C_1-C_9	1.42	0.047
C_1-CH_3	1.64	0.058
$C_1 \ldots C_3$	2.43	0.062
$C_2 \ldots CH_3$	2.64	0.057
$C_1 \ldots C_4$	2.80	0.056
$C_8 \ldots CH_3$	3.01	0.061
$C_1 \ldots C_5$	3.70	0.058
$C_3 \ldots CH_3$	3.95	0.068
$C_1 \ldots C_6$	4.22	0.063
$C_4 \ldots CH_3$	4.44	0.073
$C_7 \ldots CH_3$	4.45	0.074
$C_2 \ldots C_7$	4.83	0.057
$C_2 \ldots C_6$	5.05	0.061
$C_5 \ldots CH_3$	5.15	0.082
$C_6 \ldots CH_3$	5.30	0.084

Atomic scattering factors f_C, f_H were calculated according to the formula [27]:

$$f(\lambda^{-1} \sin \vartheta) = \sum_{i=1}^{4} a_i \exp(-b_i \lambda^{-2} \sin^2 \vartheta) + C, \tag{10}$$

using the a_i, b_i, and C values determined by Narten [28].

The molecular function of structure $i_m(S)$ was calculated using the Debye formula (9) for the Bragg angle ϑ varying from $0°$ to $60°$. The courses of the dependencies $i(S)$, $i_m(S)$, and $i_d(S)$ are shown in Fig. 3.
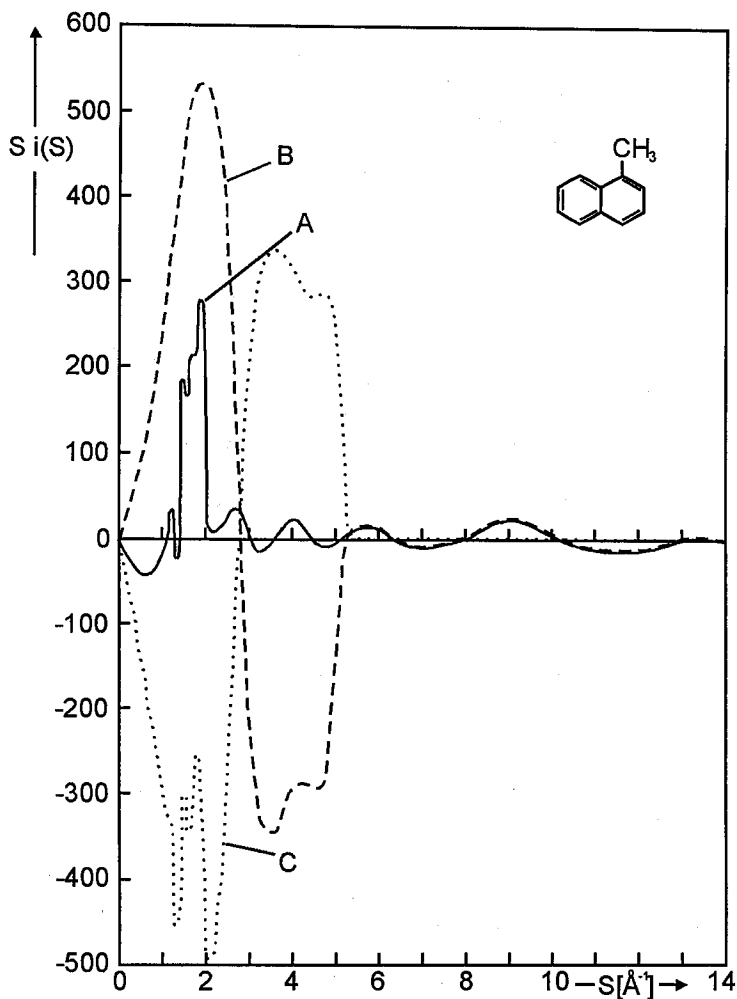
Fig. 3. Curve *A* (continuous line), the experimental structure function $Si(S)\exp(-\alpha^2 S^2)$. Curve *B* (broken line), the molecular structure function $Si_m(S)$ calculated according to Debye. Curve *C* (dotted line), subtraction of the calculated curve *B* from the curve *A*, $S[i(S)\exp(-\alpha^2 S^2) - i_m(S)]$.

The values of $i_m(S)$ from Fig. 3 have been calculated for the proposed model of 1-methylnaphthalene, which is shown in Fig. 4.

The application of Fourier transform [29] to $i(S)$ gave the function of electron density distribution displayed in Fig. 5. The EDRDF of liquid 1-methylnaphthalene (Fig. 5) has thirteen maxima in the range $1 \leq \bar{r} \leq 20$ Å. Their positions correspond to the most probable interatomic and intermolecular distances in the studied liquid, these are considered in detail below. In liquid 1-methylnaphthalene the presence of the coordination spheres of intermolecular ordering [30] was established. Subsequently, the ranges of the spheres and coordination numbers (Table III) were determined. Coordination shells are delimited by minima of EDRDF [31].
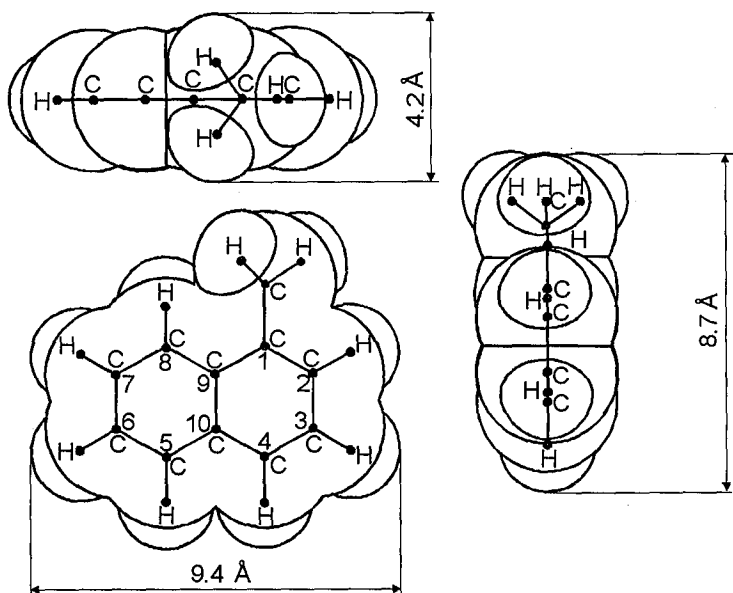
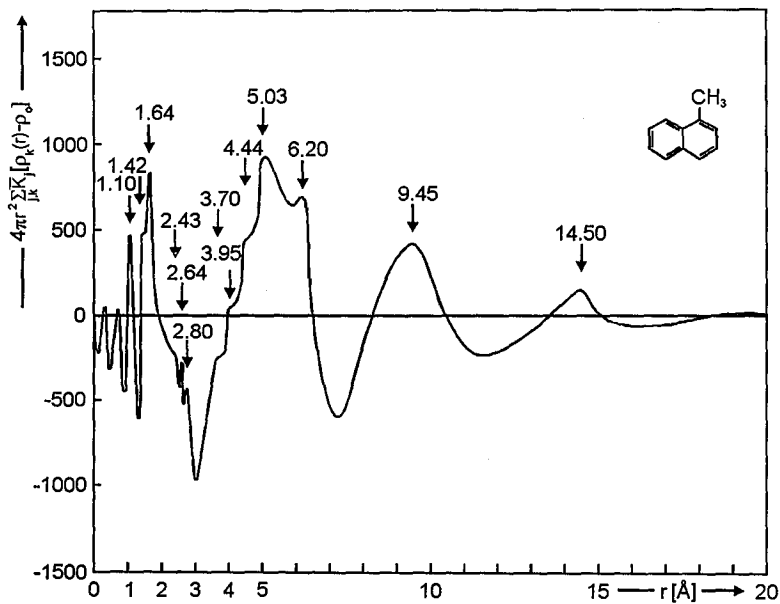Fig. 4.   A model of 1-methylnaphthalene $C_{10}H_7-CH_3$ molecule structure.

Fig. 5.   The electron-density radial-distribution function $4\pi r^2 \sum_{j,k}^{n} \overline{K}_j[\rho_k(r) - \rho_0]$ for liquid 1-methylnaphthalene.

TABLE III

The range of coordination spheres and the number of molecules in each sphere for the studied 1-methylnaphthalene.

Sphere	Range [Å]	Number of molecules
1st	3.05–7.35	8.4
2nd	7.35–11.60	29.1
3rd	11.60–16.35	57.6

The most probable binary radial correlation of molecules in liquid 1-methyl-naphthalene is shown in Fig. 6.
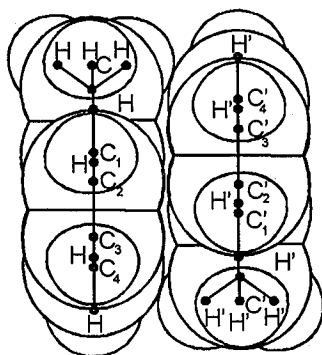

Fig. 6.   Model of probable conformation of molecules in liquid 1-methylnaphthalene. The arrangement of molecules corresponding to the maxima in the electron-density radial-distribution function.

5. Discussion

In this work the experimental distribution of $Si(S) \exp(-\alpha^2 S^2)$ (Fig. 2, curve A) was compared with theoretical data of the curve $i_m(S)$ (Fig. 3, curve B) to the $S_{\max}$ value equal to 14 Å^{-1}. The obtained coincidence of these two curves proves that the proposed model of 1-methylnaphthalene molecule, and in particular the interatomic distances in the molecule, is correct. In this situation the EDRDF (Fig. 5) was calculated. The function of the radial distribution of electron density $4\pi r^2 \sum_{j,k}^{n} \overline{K}_j [\rho_k(r) - \rho_0]$ is, according to Eq. (1), determined by the integral, $\frac{2r}{\pi} \int_0^\infty Si(S) \exp(-\alpha^2 S^2) \sin(Sr) dS$, calculated by the numerical Simpson method for $0 \leq \bar{r} \leq 20$ Å with a step of $\Delta\bar{r} = 0.05$ Å.

In the range of the argument below 1 Å, the values of the radial distribution function making the interpretation difficult or even impossible (for example, the negative ones) are not uncommon. They seem to be due to some approximations assumed in the method, e.g. to the extrapolation of small angle scattering results [32].

   The size of a 1-methylnaphthalene molecule is 9.4 Å (length) × 4.2 Å (width) × 8.7 Å (height). On the average it takes a volume of $V = 235.9$ Å^3, whereas an increment of the molecule volume [33] is $V^{\mathrm{incr}} = 150.8$ Å^3. The packing coefficient of molecules in liquid 1-methylnaphthalene is $k = 0.64$. Due to the size of the 1-methylnaphthalene molecule (Fig. 4) which can be evaluated from the van der Waals radii of carbon and hydrogen atoms, the first nine maxima of the EDRDF (Fig. 5) should be ascribed to the interferences inside a respective single molecule. The maxima fall within the range $1 < \overline{r} \leq 4.5$ Å and their positions correspond to the distances between the following atomic pairs: $C_2-H = 1.10$ Å, $C_1-C_9 = 1.42$ Å, $C_1-CH_3 = 1.64$ Å, $C_1 \ldots C_3 = 2.43$ Å, $C_2 \ldots CH_3 = 2.64$ Å, $C_1 \ldots C_4 = 2.80$ Å, $C_1 \ldots C_5 = 3.70$ Å, $C_3 \ldots CH_3 = 3.95$ Å, $C_4 \ldots CH_3 = 4.44$ Å.

   The subsequent maxima of the EDRDF correspond to the mean intermolecular distances in the liquid studied. The positions of the maxima of this function (Fig. 5), corresponding to the mean least intermolecular distances, are directly related to the main maxima in the intensity distribution. According to the data from Fig. 2 and Table II, the intensity distribution for liquid 1-methylnaphthalene shows three maxima corresponding to intermolecular interactions. The first of them, for $S_1 = 1.20$ Å^{-1}, corresponds to the intermolecular distance of $\overline{R} = 6.14$ Å, calculated [20] from Eq. (7). The second maximum on the intensity distribution curve at $S_2 = 1.45$ Å^{-1} corresponds to 5.03 Å. This maximum should be attributed to intermolecular interactions considering its high intensity and the fact of the occurrence of EDRDF maximum for $\overline{R} = 5.03$ Å. The third maximum (for $S_3 = 1.63$ Å^{-1}) corresponds to the mean intramolecular distance of $\overline{R} = 4.44$ Å (Fig. 5). In the range $\approx 4.5 \leq \overline{r} < 20$ Å the function can be divided into three regions corresponding to three spheres of intermolecular ordering [30].

   The most probable simple configurations of neighboring molecules in the liquid in question can be examined by fitting the positions of the maxima of the EDRDF to the distances between the centers of neighbors, resulting from their van der Waals models. Such models can be constructed on the basis of the bond lengths within the molecule and of the van der Waals radii of C and H atoms. Because of the permanent dipole moment of the molecule, $\mu = 1.49$ D [34], the neighboring molecules are arranged so that their dipolar moments are antiparallel [35]. This orientation is confirmed by the peaks on the EDRDF appearing at: $C_3 \ldots C_3' = 5.03$ Å, and $CH_3 \ldots CH_3' = 6.20$ Å. The maxima of this function at $\overline{r} = 9.45$ Å and $\overline{r} = 14.50$ Å bring information about the difference between the observed and the average distribution of electron density. The positions of the maxima on the functions presented in Figs. 2 and 5 made the basis for the most probable model of molecular arrangement in liquid 1-methylnaphthalene, see Fig. 6.

6. Conclusions

   This paper is devoted to determination of a structural model of 1-methylnaphthalene using the reduction method, which permits a more exact interpretation of the X-ray diffraction data. The maxima on the EDRDF in the range $1 < \overline{r} \leq 4.5$ Å have been ascribed to the following pairs of atoms: $C_2-H = 1.10$ Å,

$C_1-C_9 = 1.42$ Å, $C_1-CH_3 = 1.64$ Å, $C_1...C_3 = 2.43$ Å, $C_2...CH_3 = 2.64$ Å, $C_1...C_4 = 2.80$ Å, $C_1...C_5 = 3.70$ Å, $C_3...CH_3 = 3.95$ Å, $C_4...CH_3 = 4.44$ Å.

The X-ray diffraction method permitted determination of mean structural parameters of liquid 1-methylnaphthalene (the intra- and intermolecular distances, the radii of coordination spheres, the coordination numbers) at 293 K. The appearance of clear maxima on the functions of angular-distribution of X-ray radiation and EDRDF indicates the presence of short-range ordering in liquid 1-methylnaphthalene up to the distance of about 20 Å.

The use of a short-wave radiation from an X-ray tube with a molybdenum anode allowed determination of the 3 spheres of intermolecular ordering that is the supermolecular structure in the studied liquid. The values of $\overline{R} = 5.03$ Å and $\overline{R} = 6.20$ Å determined provide the information about intermolecular spatial configurations in the liquid studied. The mean intermolecular distances determined by the method of Voigtlaender-Tetzner are in good agreement with the maxima of the EDRDF. It seems that naphthalene rings of two molecules are situated in parallel planes, which results in the antiparallel setting of the dipole moments of the liquid 1-methylnaphthalene molecules. These results can be interpreted in terms of a simple model of local arrangement of molecules, which probably can be valid for a larger class of molecular liquids, i.e. weakly polar monosubstituted derivatives of naphthalene.

The packing coefficient of molecules in liquid 1-methylnaphthalene is approximately constant in all coordination spheres and equal to 0.64. This value falls within the range of k values acceptable for the liquid phase substances. These results are also consistent with the values presumed in the conformational structure investigation of 1-methylnaphthalene performed by empirical and semi-empirical molecular orbital – linear combination of atomic orbitals (MO-LCAO) calculations [36]. The results have also been confirmed by the conformational analysis based on minimization of the potential energy [37, 38].

Because of the supposed role of the ring in mutual configurations of molecules in liquid 1-methylnaphthalene, it seems very probable that the proposed model of local arrangement can be valid for other weakly polar derivatives of naphthalene in the liquid phase. This approach gives a good description of intermolecular interactions in liquids and shows that the X-ray method is useful for their analysis.

Acknowledgment

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