PECULIARITIES IN THERMAL CONDUCTIVITY OF ALKALI METALS IN GAS PHASE. RELATIONSHIP BETWEEN THERMAL CONDUCTIVITY AND VISCOSITY

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Results of applying the method of successive approximations to the theory of dissociated gaseous media are offered. Experimental data on thermal conductivity and viscosity of rubidium vapor are considered simultaneously. Peculiarities of thermal conductivity of alkali metals in the gas phase are discussed.

At the present time experimental studies of the thermal conductivity of the vapors of all alkali metals except lithium have been performed. Of special interest are those studies in which thermal-conductivity measurements were performed at various pressures, permitting study of the effect of the molecular component and the thermal effects of dissociation on energy transfer. Such studies have been performed by Timrot, Totskii, Makhrov, et al. [1-3, 28, 29], Vargaftik, Voshchinin, Kerzhentsev, and Studnikov [4-7], and Briggs [8]. The results obtained in these studies show that the contribution of dissociation to the total thermal flux is significantly lower than would be expected from existing theoretical calculations [2].

There is also a disagreement between experimental and theoretical data for viscosity [9, 13, 14]. As is well known, it follows from the experiments of [15-20] that the viscosity of alkali metals in the gas phase decreases with increase in pressure, which contradicts the calculations of [9, 10, 11] in which the effect of pressure on viscosity is positive.

To study the causes of this divergence between theory and experiment and to increase the reliability of available data it is important to consider the experimental results on thermal conductivity and viscosity of the alkali metals in the gas phase together.

§1. Application of the method of successive approximations to the theory of thermal conductivity of dissociated gases. Following the Hirschfelder–Brokaw theory [23, 24], we take

$$\lambda = \lambda_{\text{mix},1} + \lambda_{\text{int}} + \lambda_r = \lambda_f + \lambda_r. \tag{1}$$

Introducing the relative sections

$$\beta_{12}^2 = \frac{\sigma_{12}^2\,\Omega_{12}^{(2,2)*}}{\sigma_1^2\,\Omega_1^{(2,2)*}}; \quad \beta_1^2 = \frac{\sigma_{12}^2\,\Omega_{12}^{(2,2)*}}{\sigma_2^2\,\Omega_2^{(2,2)*}}; \quad \beta_2^2 = \frac{\sigma_2^2\,\Omega_2^{(2,2)*}}{\sigma_1^2\,\Omega_1^{(2,2)*}} \tag{2}$$

and assuming $M_2 = 2M_1$, after several transformations we obtain [22]

$$\lambda_{\text{mix},1} = \lambda_1 \frac{1 + b_1x_2 + b_2x_2^2}{1 + a_1x_2 + a_2x_2^2}, \tag{3}$$

$$a_1 = \frac{4\sqrt{3}}{3}\,\frac{1 - v^{(3)}}{1 + v^{(1)}}\,\beta_{12}^2 + \frac{3\sqrt{6}}{10}\,\frac{A_{12}^*}{1 + v^{(1)}}\,\frac{1}{\beta_1^2} - 2;$$

$$a_2 = -(a_1 + 1) + \sqrt{2}\,\frac{1 + v^{(2)}}{1 + v^{(1)}}\,\beta_2^2;$$

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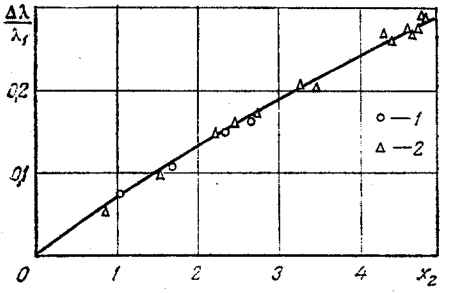

Fig. 1

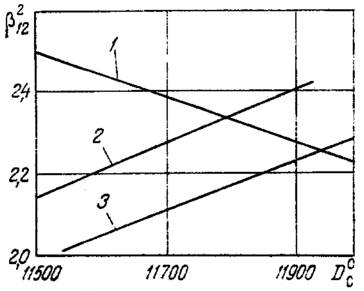

Fig. 2

Fig. 1. Function $\Delta\lambda/\lambda_1 = f(x_2)$ for isotherm III (T = 796°K); 1) points of isotherm IIIA; 2) IIIB; x, %.

Fig. 2. β^2_{12} versus D^0_0 and A^*_{12}: 1) from viscosity data of [17]; 2) thermal-conductivity data of [6] with $A^*_{12} = 1.2$; 3) same with $A^*_{12} = 1.1$. D^0_0, cal/mol.

$$b_1 = -\frac{2(v^{(1)}+v^{(4)})}{1+v^{(1)}} + \frac{3\sqrt{3}A^*_{12}}{10(1+v^{(1)})}\frac{1}{\beta^2_{12}} + \frac{3\sqrt{6}}{10(1+v^{(1)})}\frac{A^*_{12}}{\beta^2_1};$$
$$b_2 = -\left(\frac{v^{(1)}-v^{(2)}}{1+v^{(1)}}+b_1\right); \tag{4}$$

$$\lambda_{\text{int}} = \frac{\lambda_{\text{int},2}}{1+\frac{1-x_2}{x_2}\frac{D_{22}}{D_{21}}} = \lambda_1\frac{4\sqrt{3}}{25}\frac{A^*_{12}}{\beta^2_{12}}\left(\frac{C_{p2}}{R}-\frac{5}{2}\right)\frac{x_2}{1-\left(1-\frac{D_{21}}{D_{22}}x_2\right)}, \tag{5}$$

$$\lambda_r = \frac{D_{12}p}{T}\left(\frac{D_0(T)}{RT}\right)^2\frac{x_1x_2}{(1+x_2)^2} = \lambda_1\frac{0.2771\,A^*_{12}}{\beta^2_{12}}\left(\frac{T_r}{T}\right)^2\frac{x_1x_2}{(1+x_2)^2}, \tag{6}$$

$$\lambda_1\cdot 10^4 = 833\frac{\sqrt{T/M_1}}{\sigma_1^2\,\Omega_{11}^{(2,2)*}}\left(\frac{\text{W}}{\text{m}\cdot{}^\circ\text{K}}\right), \tag{7}$$

where x_1 and x_2 are the concentrations of the atomic and molecular components; $\lambda_{\text{int},2} = nD_{22}(C_{p,2} - 5/2\,R)$, thermal conductivity of the molecular component produced by internal degrees of freedom [24]; $T_r = D_0(T)/R$, "temperature" of the dissociation reaction;

$$v^{(1)} = \frac{4}{15}A^*_{12} - \frac{1}{10}B^*_{12} + \frac{5}{24}; \quad v^{(2)} = \frac{4}{15}A^*_{12} - \frac{2}{5}B^*_{12} + \frac{1}{12};$$
$$v^{(3)} = \frac{1}{5}B^*_{12} + \frac{3}{16}\frac{B^*_{12}}{A^*_{12}} + \frac{25}{64}\frac{1}{A^*_{12}} + \frac{1}{2}; \tag{8}$$
$$v^{(4)} = \frac{2}{5}A^*_{12} + \frac{1}{5}B^*_{12} - \frac{1}{6}.$$

Expanding Eqs. (3) and (5) in series in x_2, we obtain

$$\frac{\Delta\lambda}{\lambda_1} = \frac{\lambda-\lambda_1}{\lambda_1} = \sum_{n=1}^{\infty}A_n^{(f)}x_2^n + A_r\left(\frac{T_r}{T}\right)^2\frac{x_1x_2}{(1+x_2)^2}, \tag{9}$$

where

$$A_n^{(f)} = A_n^{(\text{mix},1)} + A_n^{(\text{int})};\quad A_1^{(\text{mix},1)} = -a_1 + b_1;\quad A_2^{(\text{mix},1)} = -a_1A_1^{(\text{mix}\,1)} + b_2 - a_2;$$

TABLE 1. Averaged Experimental Data of [6]

Isotherm	T, °K	Measurement series	No. of exptl. points	$\left(\frac{T_r}{T}\right)^2$	$\lambda_1 \cdot 10^4$, W/m·K	β_{12}^2		
						$A_{12}^*=1,1$ $B_{12}^*=1,1$	$A_{12}^*=1,1$ $B_{12}^*=1,2$	$A_{12}^*=1,2$ $B_{12}^*=1,2$
IA	1071,0	I	11	35,0	101,8	2,02	2,04	2,18
Б	1078,3	II	5	34,6	102,5	2,02	2,04	2,17
IIA	924,2	I	7	45,6	87,7	2,06	2,07	2,21
Б	924,7	II	4	45,6	88,7	2,06	2,07	2,21
В	918,6	III	7	46,1	89,0	2,06	2,07	2,21
IIIA	798,8	I	14	60,0	77,4	2,06	2,07	2,22
Б	792,5	II	4	60,9	77,1	2,11	2,12	2,27
IVA	953,7	III	14	43,1	92,1	2,06	2,07	2,22
Б	963,1	III	18	42,3	93,6	2,07	2,08	2,23
V	871,6	III	7	50,9	84,6	2,06	2,07	2,22
Mean β_{12}^2 value						2,05	2,07	2,21

TABLE 2. Effective Atomic Collision Sections for Gaseous Rubidium According to Data of [6] ($\overline{\sigma}_1^2 = \sigma_1^2\Omega_{11}^{(2,2)}$ in (Å)^2)

T, °K	$\overline{\sigma}_1^2$	$\delta\sigma_1^2$ (α=0,95)
700	34,7	0,3
800	32,9	0,2
900	31,3	0,2
1000	29,9	0,2
1100	28,7	0,2
1200	27,6	0,1
1300	26,6	0,1

$$A_{n(n\geqslant 3)}^{(\text{mix},1)} = -a_1 A_{n-1}^{(\text{mix},1)} - a_2 A_{n-2}^{(\text{mix},1)}, \tag{10}$$

$$A_1^{(\text{int})} = \frac{4\sqrt{3}}{25} A_{12}^* \left(\frac{C_{p,2}}{R} - \frac{5}{2}\right) \frac{1}{\beta_{12}^2}; \tag{11}$$

$$A_{n(n\geqslant 2)}^{(\text{int})} = A_1^{(\text{int})} \left(1 - \frac{\sqrt{6}}{2} \frac{A_{12}^*}{A_2^*} \frac{1}{\beta_1^2}\right)^{n-1};$$

$$A_r = \frac{0.2771\, A_{12}^*}{\beta_{12}^2}. \tag{12}$$

Equation (9) defines the thermal conductivity of the dissociated gas as the thermal conductivity of the dominant component λ_1 multiplied by the series in x_2 which characterizes the effect of the second component. By increasing the number of terms in the series, we obtain a system of successive approximations, which converges to any desired degree of accuracy with the results of exact theory. Series (9) is alternating in sign, so that the value of the first term dropped may be used to judge the maximum error of the approximation used. In particular, for λ of alkali metal vapor (β_{12}^2 = 2-2.7) at $x_2 \leq 0.1$ the error of the third approximation $E_3 \leq 0.6\%$, and for the fourth approximation $E_4 < 0.15\%$.

The coefficients of series (9) are defined by the relative sections (2), while A_1 depends solely on β_{12}^2 in accordance with the physical sense of the first approximation. The dependence of the relative excess thermal conductivity $\Delta\lambda/\lambda_1$ on x_2 may be used to determine the relative sections β_{12}^2. However the coefficients of (9) depend on A_{12}^* and B_{12}^*. From the condition $dA_1 = (\partial A_1/\partial\beta_{12}^2)\,\delta\beta_{12}^2 + (\partial A_1/\partial A_{12}^*)\delta A_{12}^* = 0$ the relationship between δA_{12}^* and $\delta\beta_{12}^2$ follows:

$$\delta\beta_{12}^2 = \frac{\beta_{12}^2}{A_{12}^*}\left[1 - \frac{\beta_{12}^4}{(T_r/T)^2}\left(\frac{4.7}{A_{12}^*} - 2.9\right)\right]\delta A_{12}^*, \tag{13}$$

where $A_1 = A_1^{(f)} + A_r(T_r/T)^2$; $\delta\beta_{12}^2$ and δA_{12}^* are the uncertainties of the quantities β_{12}^2 and A_{12}^*.

TABLE 3. Averaging of Experimental Data [29]

T, °K	$\left(\frac{T_r}{T}\right)^2$	$\lambda_1 \cdot 10^4$, W/m · °K	β_{12}^2 $(A_{12}^* = B_{12}^* = 1,2)$
900	47,91	85,1	2,25
1000	39,56	92,5	2,15
1100	33,44	103,3	2,18

In the experiments on thermal conductivity of gaseous alkali metals

$$\beta_{12}^2 = 2 - 2.5; \quad (T_r/T)^2 = 30 - 100$$

and at $\delta A_{12}^* = 0.1$

$$\delta\beta_{12}^2 \approx 0.10 - 0.20.$$

The dependence of β_{12}^2 on B_{12}^* is weaker. At $\delta B_{12}^* = 0.1$, $\delta\beta_{12}^2 = 0.01$-$0.02$, which is an order of magnitude smaller than for the same change in A_{12}^*.

Thus the results of calculating β_{12}^2 from experimental data on thermal conductivity depend significantly on the A_{12}^* value chosen, in contrast to the case of viscosity, where the dependence of β_{12}^2 on A_{12}^* is negligibly small [14, 30]. Therefore, simultaneous consideration of viscosity and thermal-conductivity experiments allows us to make certain assumptions relative to the magnitude of A_{12}^*.

Equations (4), (7), (9)-(12) define the dependence of thermal conductivity of dissociated gases on effective particle sections.

Use of the successive approximation method in the theory of viscosity leads to analogous results [14, 30]. Thus, in terms of σ_1^2 and β_{12}^2 we establish a relationship between thermal conductivity and viscosity of dissociated gases, which permits simultaneous consideration of results of experimental studies.

The theoretical results obtained are completely applicable to the case of alkali metal vapors in view of the smallness of x_2 in this case. We will consider the peculiarities of thermal conductivity in the gaseous phase of alkali metals and its relationship to viscosity with the example of rubidium.

§2. Experimental results on thermal conductivity of gaseous rubidium. The isotherm characteristics of [6] and the results of averaging experimental data along the isotherms are presented in Table 1.

The molecular concentrations were determined from [25]. The function $\Delta\lambda/\lambda_1 = f(x_2)$ for one of the isotherms is shown in Fig. 1.

Averaging λ_1 for the isotherm for the series I and II measurements by the method of least squares we obtain

$$\lambda_1(T) \cdot 10^4 = 95.4 + 0.089(T - 1000). \tag{14}$$

The averaging error $\delta\lambda_1 \cdot 10^4 = 0.6$ W/m·°K at a confidence level $\alpha = 0.95$.

To determine $\lambda_1(T)$ the isotherm III λ_1 measurements were not used, since those measurements were performed near the saturation line for large x_2. However the deviation of the series III λ_1 from Eq. (14) comprises 0.5-1.4%, i.e., is significantly less than the experimental uncertainty.

The atomic collision sections calculated from $\lambda_1(T)$ as defined by Eq. (14) are shown in Table 2.

The error in the β_{12}^2 determination is composed of the averaging error $(\Delta\lambda/\lambda_1)_{\text{exp}}$ at T = const (δ_1), the error in averaging β_{12}^2 and λ_1 along the isotherms (δ_2, δ_3), and the inaccuracy in the magnitude of the dissociation energy (δ_4) and the ratio A_{12}^* (δ_5). The dependence of β_{12}^2 on the D_0^0 and A_{12}^* values used in the calculations is shown in Fig. 2.

The results obtained are confirmed completely by the data of [29] (Table 3), according to which $\beta_{12}^2 = 2.2$ (for $A_{12}^* = B_{12}^* = 1.2$), while the thermal conductivity of the atomic component

$$\lambda_1(T) \cdot 10^4 = 94.0 + 0.09(T - 1000) \tag{15}$$

TABLE 4. Errors in Determination of Relative Atom–Molecule Collision Section (confidence level $\alpha = 0.95$)

Experiment	δ_1	δ_2	δ_3	$\delta_{123} = \sqrt{\sum_{i=1}^{3} \delta_i^2}$	δ_4 $\left(\delta D_0^0 = 250 \frac{\text{cal}}{\text{mole}}\right)$	δ_5 $(\delta A_{12}^* = 0{,}05)$
η [17]	0,05	—	0,04	0,07	0,14	—
η [6]	0,03	0,05	0,03	0,07	0,16	0,08

TABLE 5. Coefficients of Expansions of Gaseous Rubidium Thermal Conductivity in Series in x_2

Thermal conductivity component	Coefficient			
	A_1	A_2	A_3	A_4
$\lambda_{\text{mix},1}$	−2,246	3,91	−6,8	11,8
λ_{int}	0,284	−0,33	0,4	−0,4
λ_f	−1,961	3,58	−6,4	11,4
λ_r	0,1505			

has the same temperature dependence, differing from $\lambda_1(T)$ of [6] by 1.8-1.1% (at T = 900-1150°K) (the data were not processed statistically, since in [29] data for only 1/3 of the experimental points were published).

§3. Comparison of experimental results on viscosity and thermal conductivity. The viscosity of gaseous rubidium has been studied by Sidorov, Tarlakov, and Yargin [17], and Bonilla and Lee [20].

According to the data of [17],

$$\lambda_1(T) \cdot 10^4 = 94.7 + 0.074(T - 1000), \quad \delta\lambda_1 \cdot 10^4 = 0.35, \tag{16}$$

$$\beta_{12}^2 = 2.44 \pm 0.07. \tag{17}$$

The components of the error in β_{12}^2 are presented in Table 4.

From [20] it follows that

$$\lambda_1(T) \cdot 10^4 = 92.6 + 0.110(T - 1000), \quad \delta\lambda_1 \cdot 10^4 = 1.6, \tag{18}$$

$$\beta_{12}^2 = 2.40 \pm 0.18. \tag{19}$$

The atomic sections of [6] at T > 950°K are smaller than those found by Eq. (16). But in the temperature range studied (800-1200°K) the difference between the two does not exceed the uncertainty of the more accurate experiment [17]. The difference between the sections of [6] and [20] is somewhat greater but for the experimental temperatures it does not exceed the uncertainty of [6].

The viscosity studies [17] and [20] produced practically identical values for the section β_{12}^2 (the difference does not exceed the mean square averaging error of [17]). A lower value of β_{12}^2 follows from thermal-conductivity experiments. However for $A_{12}^* = 1.2$ the difference $(\beta_{12}^2)_\eta - (\beta_{12}^2)_\lambda$ proves to be smaller than the tolerance determined by $\delta D_0^0 (\delta D_0^0 = 250$ cal/mol [25]).

§4. Results of averaging experimental data on gaseous rubidium thermal conductivity:

$$\lambda(x_2, T) = \lambda_1(T) \left[1 + \sum_{n=1}^{m} A_n^{(f)} x_2^n + A_r \left(\frac{T_r}{T}\right)^2 \frac{x_2(1 - x_2)}{(1 + x_2)^2} \right., \tag{20}$$

$$\lambda_{\text{mix},1}(x_2, T) = \lambda_1(T) \left[1 + \sum_{n=1}^{m} A_n^{(\text{mix},1)} x_2^n \right], \tag{21}$$

$$\lambda_{\text{int}}(x_2, T) = \lambda_1(T) \sum_{n=1}^{m} A_n^{(\text{int})} x_2^n, \tag{22}$$

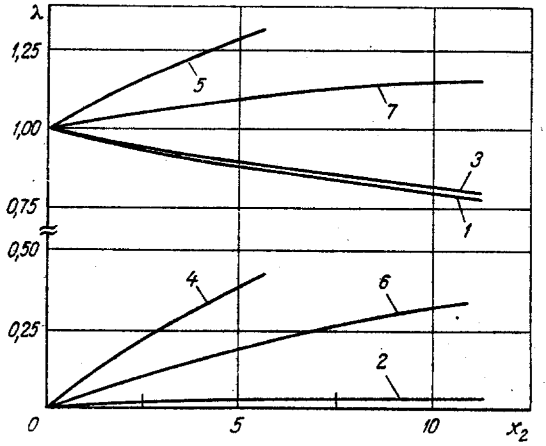

Fig. 3. Relative values of thermal-conductivity components of rubidium vapor (λ_1 = 1): 1) $\lambda_{cm,1}$; 2) λ_{int}; 3) λ_f; 4) λ_r; 5) λ (4, 5 for T = 800°K); 6) λ_r; 7) λ (6, 7 at T = 1200°K).

$$\lambda_r(x_2, T) = \lambda_1(T) A_r \left(\frac{T_r}{T}\right)^2 \frac{x_2(1-x_2)}{(1+x_2)^2}. \qquad (23)$$

The thermal conductivity of the atomic component is given by Eq. (15).

The best agreement of the β_{12}^2 values found from experimental data on thermal conductivity and viscosity indicates that for rubidium

$$A_{12}^* = B_{12}^* = 1.2 \text{ and } \beta_{12}^2 = 2.21. \qquad (24)$$

the coefficients of Eqs. (20)-(23) corresponding to these β_{12}^2 and A_{12}^* values are presented in Table 5.

The contributions of the various components to total thermal conductivity of the gaseous phase of rubidium are shown in Fig. 3. Existing theoretical calculations give a markedly different estimate of the roles of thermal-conductivity components. Thus, according to [10] $\lambda_f > \lambda_1$ (by ≈0.6% at T = 1200°K), while the ratios λ_r/λ_1 and λ_{int}/λ_1 are twice as great as the experimental data.

As is evident from Eqs. (4), (10)-(12), the approach of $\lambda_{mix,1}$ to λ_1, and the increase in contributions of dissociation energy and internal energy to effective heat capacity are a result of reduction in the size of the relative molecular section. This same fact explains the positive effect of pressure on viscosity [14]. In [9] and [10] $\beta_{12}^2 \approx 1$. (In [10] $\beta_{12}^2 = 1.02$-1.0 with $A_{12}^* = 1.1$). The equality of the sections atom–atom and atom–molecule shows the very approximate nature of existing methods of calculating the interaction between atoms and molecules in the gaseous phase of alkali metals.

Thus, the lower contribution of dissociation energy to total thermal flux and the negative dependence of viscosity on pressure characteristic of gaseous alkali metals are explained by the significant value of the atom–molecule collision section as compared to the atomic collision section.

Experiments on thermal conductivity and viscosity of rubidium confirm these transfer process peculiarities completely and agree with each other as to values of atom–atom and atom–molecule collision sections.

NOTATION

λ	is the effective thermal conductivity;
$\lambda_{mix,1}$	is the thermal conductivity of a mixture of "monatomic" components, connected with energy transfer by translational motion;
λ_{int}	is the thermal conductivity corresponding to energy transfer by internal degrees of freedom;
λ_f	is the thermal conductivity of "frozen" mixture;
λ_r	is the thermal conductivity connected with reaction energy transfer;
D_{ij}	is the binary mixture diffusion coefficient;
η	is the viscosity;

$D_0(T)$	is the molecular dissociation energy;
D_0^0	is the molecular dissociation energy at $T = 0$;
$\overline{\sigma^2} = \sigma^2\Omega^{(2,2)*}$	are the effective collision sections;
β_{ij}^2	are the relative collision sections;
A_{ij}^*, B_{ij}^*	are the ratios of corrected collision integrals appearing in equations for transfer coefficients of gas mixtures;
A_n	are the coefficients in expansion in series in x_2;
x	is the molar fraction;
T	is the temperature, °K;
$T_r = D_0(T)/R$	is the reaction "temperature";
P	is the pressure;
C_p	is the molar heat capacity at P = const;
M	is the molar weight;
R	is the universal gas content;
n	is the number of particles per unit volume;
α	is the confidence level;
δ, δ_i	are the uncertainties of of quantities.

Indices

1 is the atomic component;
2 is the molecular.

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THERMAL-CONDUCTIVITY COEFFICIENTS OF SIX LIQUID AROMATIC HYDROCARBONS

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Using experimental λ and ρ data, equations are developed for six aromatic hydrocarbons which describe λ within the limits of experimental accuracy over the temperature range 298-573°K at pressures **up to 500 bar.**

On the basis of experimental p, v, T data the authors have established [1] that the equation of state of six aromatic hydrocarbons, benzol, toluol, ethylbenzol, and o-, m-, and n-xylol, can be described with great accuracy over the temperature range 298-673°K at pressures to 500 bar by the formula

$$\frac{pv}{RT} = 1 + B\rho + H\rho^7. \tag{1}$$

In [2-4] it was indicated that the equation of state and transfer properties of liquids can be described by functions having the same form. Therefore, the thermal-conductivity coefficient of the six hydrocarbons referred to above will be described by a function

$$\frac{\lambda}{\lambda'_s} = 1 + B_\lambda\rho + H_\lambda\rho^7, \tag{2}$$

from which it follows that for λ calculations data on λ'_s, the thermal-conductivity coefficient of the saturated liquid, will be required. The latter were found for the liquids studied by extrapolation of isotherms in the coordinates λ and p with use of the experimental λ values presented in [5-10]. Then the λ' values thus found were expressed by the single empirical expression

$$\lambda'_s = \frac{T}{a + bT} + c. \tag{3}$$

The values of the constants *a*, b, and c of Eq. (3) for each hydrocarbon are presented in Table 1.

TABLE 1. Values of Constants of Eq. (3) for Six Liquid Aromatic Hydrocarbons

Coefficient	Benzol	Toluol	Ethylbenzol	o-Xylol	m-Xylol	n-Xylol
a	—0,24795	—0,07389	—0,03264	—0,14310	—0,09312	—0,15576
b	—0,0001118	—0,0002759	—0,0002378	—0,0002976	—0,0003007	—0,0003088
c	2486	3203	4149	2569	2914	2466

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