ALKALI-METAL LIQUID AND VAPOR THERMOPHYSICAL PROPERTIES

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Liquid alkali metals and their vapors are increasingly used in engineering. A monograph by Kirillin [1] classifies the data up to 1967 and gives working equations and thermodynamic tables for the liquid at normal pressure and for the vapor up to 2 MPa; there are also tables of the vapor transport properties, which usually extend up to 1500°K, but which are based mainly on calculations. Thus at the end of the 1960s there were clearly inadequate data for the liquids and vapors at high temperatures.

In the subsequent two decades, researches on these have been conducted at the Institute of High Temperatures, the Baikov Metallurgy Institute, Moscow Physics Research Institute, and Moscow Aviation Institute.

Here we present results on the thermodynamic and transport parameters derived from researches at Moscow Aviation Institute.

These researches have concerned mainly high temperatures (up to 2000°K) and pressures (up to 10 MPa), which have required appropriate methods and equipment composed of materials resistant to alkali metals at these high parameters.

The main results have been published in 1985 in a handbook of thermodynamic and transport properties for alkali metals produced by the International Union of Pure and Applied Chemistry IUPAC [2].

The results have been used in tables of recommended reference data on the viscosities and thermal conductivities of alkali-metal vapors [3], on the thermodynamic parameters for cesium vapor [4], and on the density of liquid cesium [5].

Vapor Thermodynamic Parameters. A constant-volume piezometer [6-8] has been used to measure the pVT dependence for cesium vapor, with the pressure indicator a membrane in the cold zone [9]. This improvement provided exact measurements at high temperatures, the upper limit being set only by the resistance of the piezometer material. The vapor pressure was transmitted to the membrane through a column of liquid metal in a capillary, and the pressure was balanced by argon in a chamber under the membrane. The zero position was monitored by a mechanical displacement sensor. Three studies were made (Table 1), which included 19 measurement series with piezometers having various volumes and contents.

Measurements by Ewing et al. for cesium vapor [15, 16] are familiar but are restricted to p = 3 MPa, where the error was estimated as 0.5%.

Our results and the data of [15, 16] have been fitted [2, 8] to a density-series equation:

$$p = RT \sum_{k=1}^{7} Q_k / v^k, \tag{1}$$

where $Q_1 = 1$, and for $k \geq 2$

$$Q_k = q_0^{(k)} \exp\left(q_{-2}^{(k)} T^{-2} + q_{-1}^{(k)} T^{-1} + q_e^{(k)} \ln T\right). \tag{2}$$

Table 2 gives the $q^{(k)}$. Equation (1) with the coefficients of Table 2 fits the measurements with an error of not more than 0.4%.

This has been used in thermodynamic-parameter tables (specific volume, enthalpy, entropy, isobaric specific heat, speed of sound, and compressibility) between 950 and 2500°K and from 0.1 to 13 MPa [2, 4, 14]. These were the first such tables for alkali-metal vapors at such high parameters derived from measurements.

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TABLE 1. Measurements on Cesium pVT for the Vapor at Moscow Aviation Institute

Source	Ranges		Error,%	No. of	
	T, K	P, MPa		series	exptl. points
[6, 10, 11, 12]	1365—1940	1,5—5,2	0,65	4	29
[7, 13]	1365—2117	1,5—8,9	0,9	6	21
[2, 8, 12, 14]	1365—2515	1,5—13,8	0,49	9	102

TABLE 2. Coefficients $q^{(k)}$ in the Equation of State for Cesium Vapor

$q_i^{(k)}$	k					
	2 m^3/kg	3 $(m^3/kg)^2$	4 $(m^3/kg)^3$	5 $(m^3/kg)^4$	6 $(m^3/kg)^5$	7 $(m^3/kg)^6$
$q_0^{(k)}$	—1,7778	0,016095	—0,057464	0,36063	0,053726	—0,085874
$q_{-2}^{(k)}$	3,0587	7,1707	6,4402	0,92610	—0,40390	2,0843
$q_{-1}^{(k)}$	—2,2400	0,30014	0,16314	2,7083	—0,29621	3,0199
$q_e^{(k)}$	—1,3453	2,4635	1,4803	—2,2034	0,42875	—3,7076

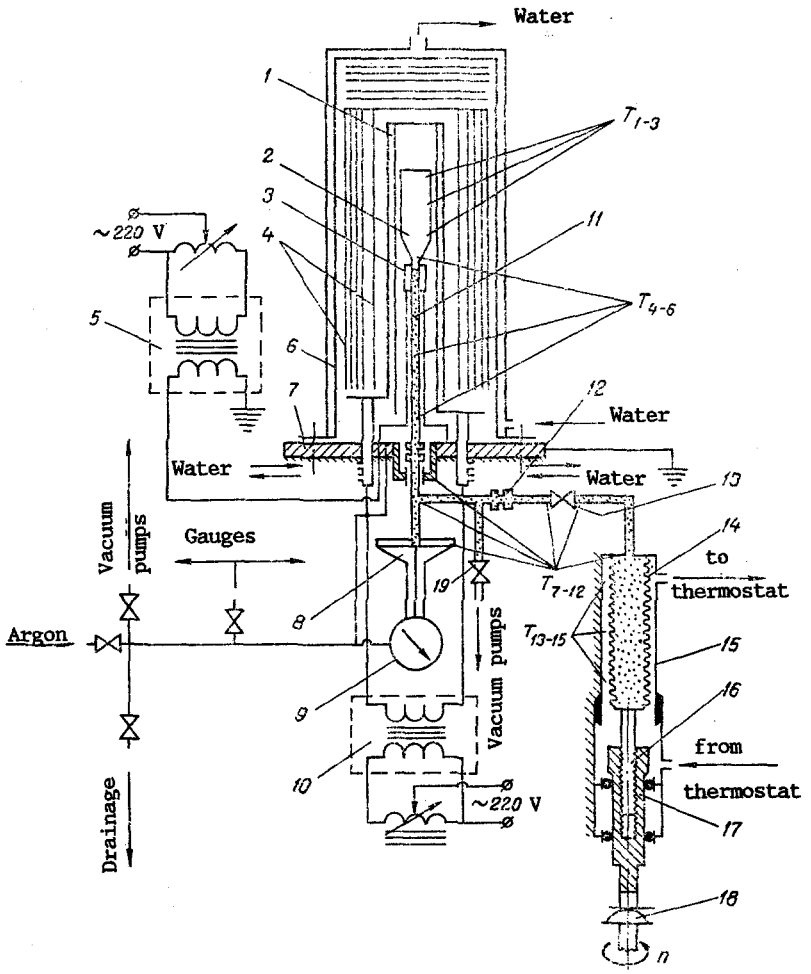

Fig. 1. The apparatus: 1) main heater; 2) piezometer chamber; 3) guard heater; 4) screens; 5, 10) transformers; 6, 7) autoclave; 8) membrane cell; 9) displacement indicator; 11) capillary; 12) coupler; 13, 19) valves; 14) bellows unit; 15) dispenser body; 16, 17) micrometer transmission; 18) spherical hinge; T_{1-3} and T_{4-6} tungsten—rhenium thermocouples; T_{7-12} and T_{13-15} Chromel—Alumel thermocouples.

TABLE 3. Coefficients b_{ij} in (3) for Rubidium Vapor

j	i				
	1	2	3	4	5
0	—0,74463	—0,09077	—0,01095	—0,001246	—0,0001374
1	—0,57586	—0,08393	—0,01117	—0,001314	—0,0001474
2	—0,38530	—0,07386	—0,01129	—0,001395	—0,0001593
3	—0,18818	—0,05981	—0,01122	—0,001477	—0,0001725
4	—0,01266	—0,04112	—0,01087	—0,001559	—0,0001872
5	0,09888	—0,01763	—0,01013	—0,001634	—0,0002034
6	0,09856	0,00989	—0,00887	—0,001696	—0,0002209
7	—0,03322	0,03903	—0,00693	—0,001732	—0,0002396
8	—0,19336	0,06462	—0,00415	—0,001729	—0,0002591
9	0,07298	0,07743	—0,000416	—0,001666	—0,0002785

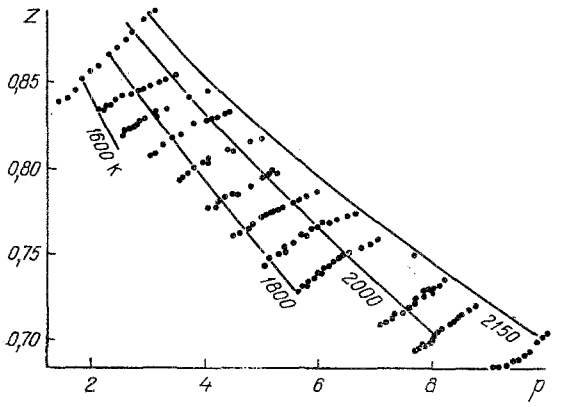

Fig. 2. Measurements on the compressibility for potassium vapor, with p in MPa.

The method has since been improved [17, 18]. A loading device has been built for altering the mass in the piezometer very accurately during the experiment, and this is a very important quantity, as the accuracy in measuring it largely determines the overall accuracy. The device also reduces the effort while maintaining the same accuracy.

Guard heaters have been mounted at the end of the piezometer, which improve the temperature uniformity considerably and reduce the data error while providing for approach to the saturation line. Other improvements have also been made [17-19].

The upgraded equipment (Fig. 1) has been used to measure rubidium PVT values; four series were performed with different initial masses, and 40 points were obtained between 1285 and 2090°K and between 0.7 and 5.18 MPa [18, 19]. The error in the compressibility coefficients was 0.7-1.5% for 0.95 probability.

The rubidium vapor-state equation is

$$Z = 1 + \sum_{i=1}^{r} \sum_{j=0}^{s_i} b_{ij} \frac{\omega^i}{\tau^j}, \qquad (3)$$

where $Z = pV/RT$ is the compressibility coefficient, $\tau = T/T_{cr}$ the reduced temperature, $\omega = \rho/\rho_{cr}$ the reduced density, T_{cr} and ρ_{cr} the temperature and density at the critical point, and b_{ij} are coefficients derived by least-squares fitting.

Table 3 gives b_{ij}. From (3) with the Table 3 coefficients, we derived tables for the thermodynamic parameters (specific volume, enthalpy, entropy, isobaric and isochoric specific heats, speed of sound, adiabatic parameter, and compressibility) [18], which cover 975-2150°K and 0.1-5 MPa.

Subsequently, pVT measurements were made for potassium vapor with the same apparatus. In 13 series with various loads, about 200 points were recorded between 1470 and 2130°K and 1.5 to 9.65 MPa [18, 19] (Fig. 2). The error range is 0.6-0.8% for probability 0.95. The measurements were made with the temperatures rising and falling when the points fitted a single curve very closely.

Before our measurements, there were ones in the literature on pVT: for cesium only up to p = 3.5 MPa [15] (and a few points at 7.0-12.5 MPa [16]), for rubidium up to 1.5 MPa [20], and for potassium up to 2.8 MPa [15].

TABLE 4. Coefficients b_{ij} in (3) for Potassium Vapor

j	i				
	1	2	3	4	5
0	—0,67220	1,85940	—0,60309	—0,23054	—0,052217
1	—0,89153	1,83340	—0,77899	—0,27837	—0,061728
2	—0,26319	1,78050	—0,98964	—0,33459	—0,072810
3	0,08680	1,68850	—1,2422	—0,40079	—0,085726
4	0,08753	1,54090	—1,5461	—0,47882	—0,10079
5	—0,19345	1,32130	—1,9130	—0,57096	—0,11839
6	—0,46735	1,02000	—2,3582	—0,67999	—0,13895
7	—0,26365	0,65103	—2,9009	—0,80938	—0,16303
8	0,55070	0,28305	—3,5653	—0,96342	—0,19126
9	—0,17398	0,09825	—4,3819	—1,14750	—0,22444

TABLE 5. Thermal-Conductivity Measurements for Alkali-Metal Vapors Made at Moscow Aviation Institute

Source	Metal	Ranges		Error,%
		T, K	p, MPa	
[2, 38, 39]	Na	946—1167	0,8—83	6
[2, 38, 40]	Na	1095	2,3—48,3	2
[2, 38, 39]	K	805—1088	0,5—132,5	7
[2, 38, 41]	Rb	792—1080	0,5—145,1	2
[2, 38, 42]	Cs	688—1087	0,2—126	6
[2, 38, 43]	Cs	1122—1169	101—150	6

TABLE 6. Measurements on Alkali-Metal Vapor Viscosities Made at Moscow Aviation Institute Before 1976 (T < 1200°K)

Source	Metal	Ranges		Error,%
		T, K	p, MPa	
[2, 38, 44, 45]	Cs	898—1189	50,6—256	2,5
[2, 38, 45]	Rb	898—1183	38,1—155	2,5
[2, 38, 46]	K	1020—1184	50,4—153	2,5
[2, 38, 47]	Na	1100—1186	21,7—80,5	3

The vapor equation of state for potassium was based on our data and the data given by Ewing et al. [15].

the (3) form applies, with the b_{ij} for potassium given in Table 4 [18]; this has given parameter tables for potassium vapor analogous to those for rubidium. The tables [18] cover 1075-2150°K and 0.1-10 MPa.

The above tables are at present unique ones for alkali-metal thermodynamic parameters over these wide parameter ranges based on measurements.

Several (3)-type equations have been drawn up for potassium and rubidium, which describe the specific-volume data equally well within the experimental errors but which differ somewhat on the isobaric specific heats. Therefore, we used a mathematical-experiment method devised at Moscow Power Institute [21, 22] to calculate the thermodynamic parameters, which has been used in calculating thermal and caloric parameters from measured PVT. Here a thermodynamic function x (entropy, enthalpy, and so on) is calculated from a set of equations of state equally well fitting the PVT data, and the resulting x are averaged $\bar{x} = 1/n \sum_{i=1}^{n} x_i$, where n is the number of equations of state.

The above (3) for rubidium and potassium with the coefficients of Tables 3 and 4 was one of the equations used as giving x closest to the tabulated $\bar{x}$. See [18] for complete information on the equations used in compiling the tables.

Liquid Thermodynamic Parameters. Liquid alkali metals have also been used in thermodynamic measurements at high parameters, particularly the density of liquid sodium. The methods and apparatus have been described in detail in [23] for 400-1000°K and 0.1-60 MPa, where the error in the data in [24, 25] was about 0.2%.

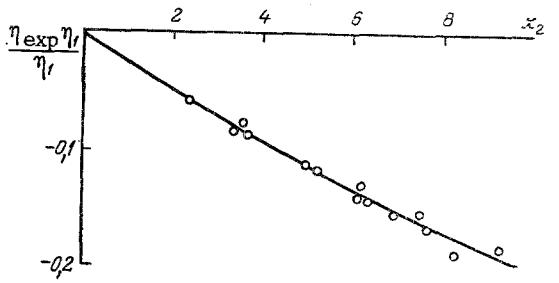

Fig. 3

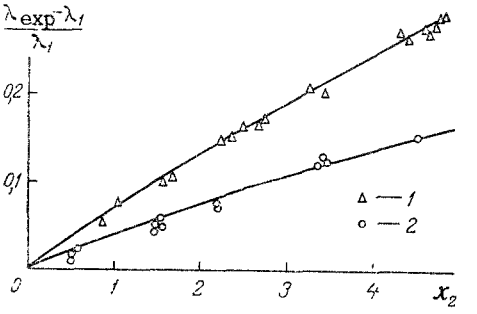

Fig. 4

Fig. 3. Dependence of $\eta_{exp} - \eta_1/\eta_1$ on molecular concentration x_2 in potassium vapor, with x_2 in %.

Fig. 4. Dependence of $\lambda_{exp} - \lambda_1/\lambda_1$ on the molecular concentration x_2 in rubidium vapor at: 1) T = 800; 2) 1075°K.

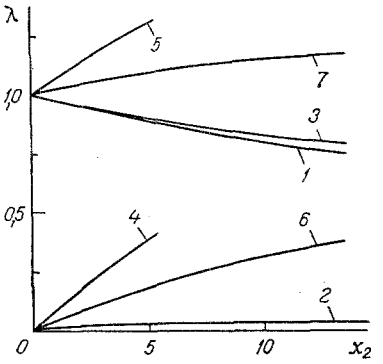

Fig. 5. Relative magnitudes of thermal-conductivity components for rubidium vapor (λ_1 = 1): 1) λ_t; 2) λ_{in}; 3) λ_f; 4) λ_r (T = 800°K); 5) λ (T = 800°K); 6) λ_r (T = 1200°K); 7) λ (T = 1200°K).

Subsequently, the main attention was given to liquid cesium up to temperatures approaching the critical point. A new apparatus has been described in [26]. The density has been measured up to 1900°K and 60 MPa [24, 25, 27-30], while the speed of sound has been measured up to 1500°K and 60 MPa [31, 32].

These measurements and thermodynamic equations have been used to calculate the enthalpy, entropy, and isoberic specific heat [33-35], and recommended reference values have been drawn up on the density of cesium in the liquid state at 350-1950°K and pressures from the saturation line up to 60 MPa [2, 5].

Further measurements have been made on the density of cesium to define the critical parameters accurately and to establish the behavior of the rectilinear diameter, both of which are important in relation to recent studies [36, 37].

Vapor Transport Parameters. An alkali-metal vapor is a binary mixture; the atomic and molecular components are linked by dissociation. The viscosity and thermal conductivity are dependent on the composition and thus on pressure, while the reaction energy is transported by the diffusion atomic flux, which increases the thermal conductivity. When these researches began at Moscow Aviation Institute in the 1960s, these aspects of alkali-metal vapor transport had not been examined thoroughly, and there were substantial discrepancies, even at the qualitative level, between the calculated values, mainly semiempirical, and the few measurements. The main ones were: differences in sign for the pressure dependence of the viscosity and considerable discrepancies over the energy transported by the diffusion flux [1, 38].

The Moscow Aviation Institute measurements were begun not only to devise reliable transport-coefficient tables but also to elucidate these transport features.

We first examined the thermal conductivities for the vapors of all the alkali metals (Table 5) by the coaxial-cylinder method. Precautions were taken to reduce the radiation transfer (cylinder material and gap minimization), which was reduced to 20-50%. The apparatus has been described in detail [2, 38, 39]. In [43], periodic heating was used, which reduced the radiation correction considerably (to not more than 4%).

TABLE 7. Coefficients in the Working Equations (4)-(6) for the Viscosity

Metal	$\eta_1(T_0)$, 10^7 Pa·sec	$\frac{d\eta_1}{dT}\cdot 10^7$, Pa·sec/K	A_1	A_2	a_1	a_2
Li	131,4	0,095	—3,652	12,515	4,546	3,989
Na	213,1	0,165	—2,950	8,22	3,789	2,913
K	193,9	0,191	—2,518	6,04	3,321	2,309
Rb	258,7	0,244	—2,133	4,40	2,905	1,817
Cs	283,3	0,232	—1,577	2,51	2,309	1,195

TABLE 8. Coefficients in the Working Equations (7)-(9) for the Thermal Conductivity

Metal	B_p	B_1	B_2	B_3	B_4	B_5	$\lambda_1(T_0)\times 10^4$, W/m·K^2	$\frac{d\lambda_1}{dT}\cdot 10^4$, W/m·K^2	α_0, K	α_1, K^{-1}	$\alpha_2\cdot 10^4$, K^{-2}
Li	0,0950	—3,837	13,65	—47,8	166	—580	591,3	0,428	13582	0,297	0,4
Na	0,1346	—2,310	5,06	—10,9	23	—49	289,4	0,224	9049	0,298	1,8
K	0,1391	—2,216	4,60	—9,3	19	—38	154,9	0,153	6907	0,508	5,3
Rb	0,1511	—1,946	3,52	—6,3	11	—19	94,5	0,089	6286	0,648	6,4
Cs	0,1598	—1,779	2,92	—4,7	7,5	—	66,6	0,055	5788	0,758	7,1

Table 6 gives viscosity data; all the measurements were made with the apparatus described in [2, 38, 44] by the straight-capillary method.

The accuracy in the thermal conductivity and viscosity was sufficient for the proportion due to the molecular component to be established reliably. The viscosity has a negative pressure coefficient (Fig. 3), while dissociation-energy transport plays comparatively small part (Fig. 4).

Studies have been made [48-51] on transport theory for reacting gases, particularly alkali-metal vapors. The above features arise from the large atom—molecule collisional cross sections (relative to atom—atom ones). The relative atom—molecule cross sections were found to be

$$\beta_{12}^2 = \sigma_{12}^2\Omega_{12}^{(2,2)*}/\sigma_1^2\Omega_{11}^{(2,2)*} \approx 2-3.$$

Analytic formulas have been derived for the viscosity and thermal conductivity in a dissociating gas as functions of the molecular-component concentration and the parameter β_{12}^2 [2, 3, 38], which have given working equations for the transport coefficients for all the alkali metals:

$$\eta(x_2,\ T) = \eta_1(T)\Big(1 + \sum_{n=1}^{m} A_n x_2^n\Big), \tag{4}$$

$$10^7\eta_1(T) = \eta_1(T_0) + \frac{d\eta_1}{dT}(T - T_0), \tag{5}$$

$$A_n = -a_1A_{n-1} - a_2A_{n-2} \quad (\text{for } n > 2), \tag{6}$$

$$\lambda(x_2,\ T) = \lambda_1(T)\left[1 + \sum_{n=1}^{m} B_n x_2^n + B_p\left(\frac{T_p}{T}\right)^2\frac{x_2(1-x_2)}{(1+x_2)^2}\right], \tag{7}$$

$$10^4\lambda_1(T) = \lambda_1(T_0) + \frac{d\lambda_1}{dT}(T - T_0), \tag{8}$$

$$T_p(T) = \alpha_0 + \alpha_1(T - T_0) + \alpha_2(T - T_0)^2, \tag{9}$$

where $\eta_1(T)$ and $\lambda_1(T)$ are the viscosity and thermal conductivity due to the atomic component, $T_p = D_0(T)/R$ is the characteristic dissociation temperature $D_0(T)$ is the dissociation energy

TABLE 9. Measurements on Lithium and Sodium Vapor Transport Coefficients Performed at Moscow Aviation Institute in 1980-1986 (T > 1200°K)

Source	Property and metal	Ranges		Error,%
		T, K	p, kPa	
[58, 59]	λ Li	1230—1420	0,4—8	8
[64]	λ Li	1400—1800	10—80	6
[57, 58]	η Li	1596—1840	33—75	8
[61, 63]	η Li	1595—1983	15—107	1,7—4,2
[63]	η Na	1115—1524	12—96,2	1,5—4,4

[52], x_2 is the molar fraction of the molecular component, and T_0 = 1000°K.

Tables 7 and 8 give the coefficients in (4)-(9).

The coefficients for sodium, potassium, rubidium, and cesium were determined by fitting to all the measurements made at the institute and collected from the literature, in conjunction with analysis and averaging for the thermal conductivity and viscosity data.

The effective atom—atom collision cross sections for lithium were calculated from the atomic interaction curves defined from spectroscopic data (singlet state) and derived by variational methods in quantum mechanics. The curves were fitted to exponentials, and a correction was made for the potential wall for the triplet state by Smirnov's method [2, 3, 53, 54].

The calculated cross sections confirmed the measurements on the thermal conductivity [55, 56, 58, 59] and the viscosity [57, 58] for lithium vapor, which were correspondingly lower by 7 and 5% in the appropriate temperature ranges.

The molecular-component effects were calculated with β_{12} = 3.5 as the mean of the data from [55, 56, 59] and [57], which agrees with the theoretical estimate [2, 3, 54].

These measurements and theoretical results provide a detailed picture of the transport and in particular enable one to calculate the components of the effective thermal conductivity corresponding to translational molecular motion λ_t, internal-energy transport λ_{in}, frozen thermal conductivity λ_f, and the reaction energy transported by the diffusion flux λ_r (Fig. 5) [2, 38].

Detailed tables have been given for the viscosity and thermal conductivity derived from (4)-(9) [2, 3, 38]; these are the only published tables for the viscosity and thermal conductivity for all the alkali-metal vapors forming a consistent system and based on the entire measurement set (for Na, K, Rb, and Cs) or on the exact interaction-potential curves (for Li).

The cross sections have been calculated from the potential curves via a special program [60] to refine the cross sections for lithium atoms, particularly at low temperatures. The values for 1500°K exceed semewhat those used in compiling the tables (within 2% limits), and the excess increases to about 5% at 1000°K, which is due to the approximate incorporation of effects from the potential well in the triplet state [54].

Recently, measurements have been made on the viscosity and thermal conductivity for lithium and sodium vapors at T > 1200°K; Table 9 gives the data.

The lithium results [63, 64] are in good agreement and also agree completely with quantum-mechanical calculations on $\eta_1(T)$ and $\lambda_1(T)$, while they refine the value of β_{12}^2 given above (from [63] and [64], $\beta_{12}^2 = 3.3 \pm 0.3$).

The sodium viscosity data [63] have been processed to give results in agreement with earlier measurements and calculations on $\eta_1(T)$ [62] from quantum-mechanical potential curves.

Future researches are of some interest:

1) pVT parameters for sodium vapor and the vapors of the other metals near the saturation line and around the critical point;

2) the caloric parameters for all the metals in the vapor state;

3) the thermodynamic parameters for the lqiuid at high temperatures; and

4) the viscosity and thermal conductivity up to 2000°K and at elevated pressures.

NOTATION

p, pressure; T, temperature; v, specific volume; R, universal gas constant; Q_i and q_i, coefficients in the equation of state; η, viscosity; λ, thermal conductivity; α_i, A_i, B_i, α_i, coefficients in the working equations for the viscosity and thermal conductivity; λ_t, λ_{in}, λ_r, λ_f, components of the thermal conductivity corresponding to the transport of translational energy, internal energy, reaction energy, and to frozen thermal conductivity.

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