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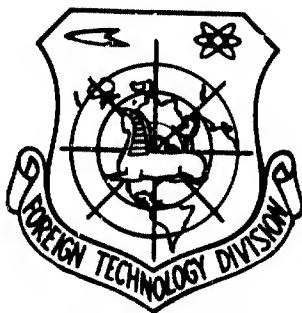


(PART III)

THERMODYNAMIC PROPERTIES OF INDIVIDUAL SUBSTANCES.
VOLUME I: CALCULATION OF THE THERMODYNAMIC PROPERTIES.
(CHAPTERS 28 THRU 31)

by

A. V. Gurvich, G. A. Khachkuruzov, et al.



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UNEDITED ROUGH DRAFT TRANSLATION

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By: A. V. Gurvich, G. A. Khachkuruzov, et al.

English pages: 1790 thru 2409 (Appendix and References included)

Translated under: Contract AF 33(657)-14184, SA2

UR/6260-062-001-000

TM6001137

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CIRC ABSTRACT WORK SHEET

(01) Acc No. TM6001137		(65) SIS Ass No.		(40) Country of Info UR			(61) Translation No. HT6600251		
(42) Author GURVICH, L. V.; KHACHKURUZOV, G. A.; MEDVEDEV, V. A.; VEYTS, I. V.; BERGMAN, G. A.							(41) Priority 2 Distribution STD		
(43) Source TERMODINAMICHESKIYE SVOYSTVA INDIVIDUAL'NYKH VESCHESTV; V DVUKH TOMAKH, TOM I; VYCHISLENIYE TERMODINAMICHESKIKH SVOYSTV									
(02) Ctry UR	(03) Ref 6260	(04) Yr 62	(05) Vol 001	(06) Iss 000	(07) S. Pg 0859	(45) E. Pg 1162	(73) Date NONE	(47) Subject Code 07, 20	
Language RUSS		2ND, REV & ENL		MOSKVA		IZD-VO AN SSSR			
(39) Topic Tags rare gas, thermodynamic property, ideal gas, equation of state, thermodynamic state equation, excited electron state, diatomic molecule, diatomic gas									
(66) Foreign Title (SEE SOURCE)									
(09) English Title THERMODYNAMIC PROPERTIES OF INDIVIDUAL SUBSTANCES; REFERENCE BOOK IN TWO VOLUMES VOL. 1: CALCULATION OF THERMODYNAMIC PROPERTIES (CHAPTERS 28 THRU 31)									
(97) Header Class 0			(63) Class 00		(64) Rel 0		(60) Release Expansion		

ABSTRACT: Thermodynamic properties for the ideal gas state in table form for 335 gases, 44 liquids, and 45 solids compounded from 33 chemical elements and their isotopes, viz.: H, D, T, He, Li, Be, B, C, N, O, F, Ne, Na, Mg, Al, Si, P, S, Cl, Ar, K, Ca, Br, Kr, Re, Sr, Zr, I, Xe, Cs, Ba, Hg, and Pb. Thermodynamic properties are given for the following 22 gases in the range from room temperature to 20,000°K: H, H⁺, H⁻, O, O⁺, H₂, O₂⁺, O₂, OH, OH⁺, H₂O, N, N⁺, N₂, N₂⁺, NO, NO⁺, C, C⁺, CO, CO⁺, and e⁻; for the 14 least stable gases up to 4000°K; and for the remaining 299 gases up to 6000°K. Virial coefficients for 34 gases are also given up to 6000°K.

In Vol. I of the Handbook the methods of calculating the thermodynamic properties of individual substances are described, and critical analysis is given of the literature data until 1960, including the constants necessary for the calculation of the tables of the thermodynamic properties (molecular constants, heats of formation and heats of phase transitions, specific heats, dissociation energies, etc.), the choice of the values of these constants accepted in the Handbook is substantiated and their values are estimated for such cases where experimental data are lacking.

Volume II of the Handbook contains tables of the thermodynamic properties of individual substances. All tables were calculated by the authors of the Handbook on the basis of the data accepted in Vol. I. English Translation: 321 tables, 4392 references; 2409 pages.

Partial Listing of Contents.

- Part One. Methods of Calculation of Thermodynamic Properties of Individual Substances.
- Chapter 1. Energy States of Atoms and Molecules.
 - Chapter 2. Statistical Methods of Calculating the Thermodynamic Functions of Ideal Gases.
 - Chapter 3. Methods of Calculation of the Thermodynamic Functions of Substances in Solid and Liquid States.
 - Chapter 4. Thermochemical Quantities.
- Part Two. Choice of the Initial Constants and Calculation of the Tables of Thermodynamic Properties.
- Chapter 5. Oxygen.
 - Chapter 6. Hydrogen and Its Isotopes.
 - Chapter 7. Compounds of Hydrogen and Its Isotopes with Oxygen.
 - Chapter 8. Fluorine and Its Oxygen Compounds.
 - Chapter 9. Chlorine and Its Compounds With Oxygen and Fluorine.
 - Chapter 10. Bromine and Its Compounds With Oxygen, Fluorine and Chlorine.
 - Chapter 11. Iodine and Its Compounds With Oxygen, Fluorine, Chlorine and Bromine.
 - Chapter 12. The Compounds of Hydrogen and Its Isotopes With Halogens.
 - Chapter 13. Sulfur and Its Compounds.
 - Chapter 14. Nitrogen and Its Compounds.
 - Chapter 15. Phosphorus and Its Compounds.
 - Chapter 16. Carbon and Its Compounds With Oxygen.
 - Chapter 17. Derivatives of Methane and Halogen Methane.
 - Chapter 18. Ethylene and Its Fluorine-Chlorine Derivates.
 - Chapter 19. Acetylene and the Fluorine Substitution Products of Acetylene.
 - Chapter 20. The Simplest Free Radicals of Hydrocarbons and Their Fluorine and Chlorine Substitution Products.
 - Chapter 21. Compounds of Carbon With Sulfur, Nitrogen and Phosphorus.
 - Chapter 22. Silicon and Its Compounds.
 - Chapter 23. Boron and Its Compounds.
 - Chapter 24. Aluminum and Its Compounds.
 - Chapter 25. Beryllium and Its Compounds.
 - Chapter 26. Magnesium and Its Compounds.
 - Chapter 27. Calcium, Strontium, Barium and Their Oxides.
 - Chapter 28. Lithium and Its Compounds.
 - Chapter 29. Alkali Metals and Compounds of Sodium and Potassium With Fluorine and Chlorine.
 - Chapter 30. Zirconium, Lead, Mercury and Their Compounds.
 - Chapter 31. Inert Gases (He, Ne, Ar, Kr, Xe) and the Electron Gas (e^-).
- Part Three. Appendix.
- Appendix 1. Atomic Weights - Isotope Composition and Nuclear Spins of The Chemical Elements Treated in the Handbook.

- Appendix 2. Fundamental Physical Constants and Transformation Factors For The Units of Energy, Volume and Pressure As Used in The Handbook.
- Appendix 3. The Formulas For the Principal Moments of Inertia of Molecules.
- Appendix 4. Concise Data From the Theory of Molecular Vibrations and Formulas for the Frequencies.
- Appendix 5. Consideration of the Intermolecular Interactions in the Calculation of the Thermodynamic Functions of Gases.
1. The Calculation of the Thermodynamic Functions of Real Gases From an Equation of State in Virial Form.
 2. Methods of Determining the Virial Coefficients.
 3. Calculation of the Thermodynamic Properties of Real Gas Mixtures.
 4. p-V-T-Data and Virial Coefficients of Some Gases.
- Appendix 6. Critical Constants.

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Chapter 28

LITHIUM AND ITS COMPOUNDS

(Li, Li⁺, Li₂, LiO, Li₂O, LiH, LiOH, LiF, LiCl)

Lithium and its compounds with oxygen, hydrogen, fluorine and chlorine will be examined in the present chapter. In addition to the two gaseous compounds of lithium with oxygen, LiO and Li₂O, considered in the Handbook, there are evidently not other sufficiently stable compounds of this class [758]. In the condensed state, lithium peroxide Li₂O₂ is also known in addition to Li₂O. However, this compound is highly unstable and the partial pressure of oxygen over it attains atmospheric at only 185°C. The thermodynamic properties of lithium hydride, the only compound of lithium with hydrogen, and also those of lithium hydroxide (LiOH), the most important compound of lithium in the system lithium-oxygen-hydrogen, are considered in the Handbook.

Of the compounds of lithium with fluorine and chlorine, LiF and LiCl are included in the Handbook. The thermodynamic properties of the polymeric compounds of the type Li_nF_n and Li_nCl_n are not considered in the Handbook. Because these molecules can be the main components of lithium fluoride and chloride vapor under certain conditions, the data presented here are not suitable for computing the pressure and composition of the saturated vapors of these compounds at temperatures of the order of 1000°K and under.

§111. MOLECULAR CONSTANTS

Li. The atom of lithium in the ground state ²S has the electronic configuration 1s²s. The boundary series 1s²nl is situated at 43 487.19

cm^{-1} above the lower state 2S . As in all hydrogen-like atoms, the state $1s^2nl$ of the lithium atom consists of doublet terms with values of $L = l$. The terms connected with the excitation of one of the $1s$ -electrons, have excitation energies of over $500,000 \text{ cm}^{-1}$ and are therefore not considered in the present Handbook.

Table 261 gives the energy levels of the lithium atom, corresponding to the electronic configuration $1s^2nl$ and values of $n \leq 11$. The excitation energies of these levels have been adopted on the basis of the values recommended by Moore [2941]. As in other cases, the states with similar excitation energies have been combined into a single level with the total statistical weight and mean energy.

The summary of Moore [2941] gives data only for a small part of the states of the lithium atom, mainly for the S-, P- and D- terms (with $n \leq 11$), because transitions to the other states of this atom have not yet been experimentally observed. For all the states of Li which have not been investigated, an estimate of the excitation energies has been made in correspondence with the approximate method described in §2 (see page 56) and on the basis of certain regularities, observed in the position of the electronic levels of the lithium atom. Thus, the energies of all the unknown levels with $L \leq 3$ (for a given n) have been assumed to be equal to the excitation energy of the 2D state, corresponding to the same value of the principal quantum number. The sole exception from this rule was made for the state $\dots 5g^2G$, whose energy was assumed to be equal to the excitation energy of the state $\dots 5f^2F$.

The maximum error in the estimated values of the excitation energies of these states can be determined if it is assumed that for the given value of the principal quantum number the difference in the energies of the terms with quantum numbers of the orbital moment L

and $L + 1$ is equal to the difference of the energies of the terms $L - 1$ and L (in reality, the terms converge rapidly with increase in L). The uncertainties of the energy levels with $n = 6$ and $L \geq 3$ thus determined do not exceed 140 cm^{-1} . For the terms with $n = 7, 8$ and 9 , the corresponding values are $120, 100$ and 78 cm^{-1} .

TABLE 261

Energy Levels of the Lithium Atom

Номер уровня A	Состояние B		Стати- стиче- ский вес E	Энергия, см ⁻¹ F
	электронная конфигу- рация C	терм D		
0	1s ² s	¹ S	2	0
1	1s ² p	³ P	6	14903,89
2	1s ² 3s	¹ S	2	27206,12
3	1s ² 3p	³ P	6	30925,4
4	1s ² 3d	³ D	10	31283,1
5	1s ² 4s	¹ S	2	35012,1
6	1s ² 4p, 4d, 4f	³ P, ³ D, ³ F	30	36595,8
7	1s ² 5s	¹ S	2	38299,5
8	1s ² 5p, 5d, 5f, 5g	³ P, ³ D, ³ F, ³ G°	48	39083,5
9	1s ² 6s	¹ S	2	39987,6
10	1s ² 6p, 6d — 6h	³ P, ³ D, ³ F°, ³ G°, ³ H°	70	40433
11	1s ² 7s — 7l	¹ S, ³ P, ³ D, ³ F° — ³ J°	98	41240
12	1s ² 8s — 8k	¹ S, ³ P, ³ D, ³ F° — ³ K°	128	41770
13	1s ² 9s — 9l	¹ S, ³ P, ³ D, ³ F° — ³ L°	162	42130
14	1s ² 10s — 10m	¹ S, ³ P, ³ D, ³ F° — ³ M°	200	42380
15	1s ² 11s — 11n	¹ S, ³ P, ³ D, ³ F° — ³ N°	242	42580

* this state has not been observed experimen-
tally.

A) Number of level; B) state; C) electronic configuration; D) term;
E) statistical weight; F) energy.

Li⁺. The positive ion of the lithium atom Li⁺ has the 1s² con-
figuration of the electron shell, analogous to the configuration of
the electron shell of the helium atom. To this configuration corres-
ponds the state ¹S, which is the ground state of the Li⁺ ion. The ex-
cited electronic states of the ion have energies over $470,000 \text{ cm}^{-1}$
and are not regarded in the present Handbook.

Li₂. Three band systems have been studied to date in the spectrum

of the molecule Li_2 : $A^1\Sigma - X^1\Sigma$, $B^1\Pi - X^1\Sigma$ and $C^1\Sigma - X^1\Sigma$, obtained in absorption upon heating of lithium vapor in a furnace to temperatures of 600-1000°K.

The band system $A^1\Sigma_u^+ \leftarrow X^1\Sigma_g^+$, situated in the red region of the spectrum, was first investigated by Wurm [4348] who obtained 11 bands of this system, corresponding to values of $v'' \leq 3$ and $v' \leq 5$, and derived an equation for their edges and carried out an analysis of the rotational structure of six bands. Later on, this system was again studied by Almy and Irwin [524] who photographed the spectrum of Li_2 on an apparatus with a dispersion of 1.3 Å/mm. The authors of the work [524] carried out an analysis of the rotational structure of 10 bands of the molecule Li_2^7 and 10 bands of the molecule Li^6Li^7 , and determined the rotational constants of both molecules and the vibrational constants, describing the wave numbers of the beginnings of the bands.

The band system $B^1\Pi_u - X^1\Sigma_g^+$, situated in the blue-green region of the spectrum, was investigated first by Wurm who obtained an equation for the band edges [4346] and carried out an analysis of the rotational structure of the bands 0-0 and 1-0 [4347]. Later on, this system was studied on apparatus with great resolving power by Harvey and Jenkins [1970] and McKellar [2712]. The authors of these two works carried out an analysis of the rotational structure of several bands and determined the rotational constants. McKellar obtained the corresponding data for the molecules Li_2^7 and Li^6Li^7 . Harvey and Jenkins [1970] and McKellar [2712] derived equations for the band edges of the system $B^1\Pi - X^1\Sigma$, describing satisfactorily the wave numbers of the band edges with low values of v'' and v' . A more detailed analysis of the vibrational structure of the system $B^1\Pi - X^1\Sigma$ was carried out by Loomis and Nusbaum [2647] who investigated the spectrum of the magnetic rota-

tion and obtained over 80 bands, corresponding to values of $v'' \leq 15$ and $v' \leq 13$. The authors of the work [2647] showed that the equations for the band edges in this system, proposed earlier, do not satisfactorily describe the edges of the bands with larger values of v' . Loomis and Nusbaum proposed a new equation for the band edges, pointing out that the bands with values of $v' \leq 11$ and $v' > 11$ cannot be described by an equation with the same constants.

The ultraviolet band systems of Li_2 were investigated in the work of Sinha [3749] and Vance and Huffman [4054]. An analysis of the vibrational structure of a system, ascribed to the transition $C^1\Sigma - X^1\Sigma$ was carried out in the work [3749]. The analysis of the spectrum of Li_2 in this region was repeated in the work of Barrow, Travis and Wright [662a] published in 1961. Two band systems were investigated in this work and it was shown as a result of an analysis of the rotational structure, that they belong to the systems $C^1\Pi - X^1\Sigma$ and $D^1\Pi - X^1\Sigma$.

The vibrational constants of Li_2 in the state $X^1\Sigma$ have been determined by several authors, but all these, with the exception of Almy and Irwin [524], were obtained for the band edges. The Handbook [649] recommended the values of the constants, found in the work of Almy and Irwin [524] and the monograph of Herzberg [2020] proposed values which differed somewhat from those obtained in the original works and, which were obviously calculated by Herzberg himself on the basis of the above considered data. However, all the constants, known in the literature,* evidently do not correctly describe the energies of the higher vibrational levels of this state, in particular, those which are close to the dissociation limit. Thus, the levels, calculated by means of the constants proposed by Herzberg [2020], converge in the region of $11,880 \text{ cm}^{-1}$, i.e., almost 3000 cm^{-1} above the dissociation limit of the state $X^1\Sigma$ of the molecule Li_2 (see page 1836) and by means

of the constants, found by Loomis and Nusbaum, in the region of 9900 cm^{-1} .

The energies of the higher vibrational levels, transitions from which have been observed in the work of Loomis and Nusbaum, are described best by cubic equations with the constants, found by these authors in the work [2647]. In order to make these equations convenient close to the dissociation limit, a fourth term was introduced into it, whose coefficient was found from the equations, given on page . The constants thus obtained are given in Table 262. The rotational constants of Li_2 in the state $X^1\Sigma$ were determined by Harvey and Jenkins [1970], McKellar [2712] and Almy and Irwin [524]. Moreover, approximate values of B_0 and D_0 were found in the works of Wurm [4347, 4348]. The most accurate values of B_0 and α were found by Almy and Irwin. Because the value of D_0 , found by McKellar [2712],* had been used in the work [524] for the analysis of the rotational structure of the bands, the values B_e and α , found by Almy and Irwin and the values, β and H , proposed by McKellar, were recommended in the Handbook [649]. The same constants were adopted in the present Handbook. Herzberg [2020] recommends similar values for the rotational constants.

The constants of Li_2 in the state $A^2\Sigma_u^+$ were determined in the works of Wurm [4348] and Almy and Irwin [524]. Because of the use of an apparatus with large dispersion, the analysis of several bands and the determination of the vibrational constants on the basis of the band heads, the constants found by Almy and Irwin [524] are much more accurate and are recommended in the Handbook [649]. Herzberg [2020], referring to the work of Almy and Irwin and also to another paper of McKellar and Jenkins [2717] which was unknown to us, gives constants which practically coincide with those proposed by Almy and Irwin. The constants, recommended by Herzberg, have been adopted in the present

Handbook.

TABLE 262

Adopted Values of the Molecular Constants of Lithium Compounds

Молекула A	Состояние B	T_e	ω_e	$\omega_e x_e$	B_e	$\alpha \cdot 10^3$	$D_0 \cdot 10^6$	r_e
		cm ⁻¹						
Li ₂	X ¹ Σ _g ⁺	0	351,593	2,586 ^a	0,67293	0,719	9,84 ^b	2,672
	A ¹ Σ _g ⁺	14068,36	255,456	1,574 ^b	0,49749	0,541	7,52 ^c	3,107
	B ¹ Π _g	20439,40 ^a	269,69	2,744 ^d	0,5577	0,888	9,41 ^e	2,936
	C ¹ Π _g	30549	237,9	3,33	0,50756	0,964	—	—
	D ¹ Π _g	34140	205	—	0,465	—	—	—
LiH	X ¹ Σ	0	1405,65	23,20 ^a	7,5131	21,32 ^m	854 ⁿ	1,543
	A ¹ Σ	26516,2	234,413	-28,947 ^a	2,8186	-7,831	—	2,518
	B ¹ Π	34912	215,5	42,4	3,383	98,6 ^m	2,6·10 ⁶	2,378
LiO	X ² Σ	0	800 ⁿ	—	1,075 ⁿ	—	—	1,80 ⁿ
LIF	X ¹ Σ	0	891	4	1,364	1,64	12 ^r	1,555
LiCl	X ¹ Σ	0	662	2,2	0,711 ⁿ	0,35	3,3 ^r	2,02

A) molecule; B) state

a) $\omega_{e y_e} = -0.00939$, $\omega_{e z_e} = 0.000155$ cm⁻¹.

b) $\beta = 3.86 \cdot 10^{-8}$, $H_v = 2.0 \cdot 10^{-10}$ cm⁻¹.

c) $\omega_{e y_e} = 0.0018$ cm⁻¹.

d) calculated by means of the relation (1.36)

e) for $v'' > 11$, the authors of the work [2647] recommend: $T_e = 20440.10$,

$\omega_e = 268.09$, $\omega_e x_e = 2.341$ and $\omega_{e y_e} = -0.0879$ cm⁻¹.

f) $\omega_{e y_e} = -0.0637$ cm⁻¹.

g) $\beta = +7.68 \cdot 10^{-8}$, $H = 2.05 \cdot 10^{-10}$ cm⁻¹.

h) $\omega_{e y_e} = 0.163$ cm⁻¹.

i) $\alpha_2 = 0.00075$ cm⁻¹.

j) $\beta = +0.16 \cdot 10^{-4}$, $H = 0.111 \cdot 10^{-8}$ cm⁻¹.

k) $\omega_{e y_e} = -4.1849$ cm⁻¹.

l) $\alpha_2 = 0.045$ cm⁻¹.

m) estimate.

The vibrational constants of Li₂ in the state B¹Π were determined by Wurm [4348], Harvey and Jenkins [1970], McKellar [2712] and Loomis and Nusbaum [2647]. All these were obtained for the band edges and are fairly similar. Because several new bands were first observed in the

work of Loomis and Nusbaum [2647], the constants proposed by them describe the energy of the higher vibrational levels best although, as the authors of the work [2647] pointed out, the energies of the levels of the state $B^1\Pi$ with small ($v \leq 11$) and large ($v > 11$) values of the vibrational quantum numbers cannot be described by a single equation. The most accurate values of the rotational constants of Li_2 in the state $B^1\Pi$ were found in the work of McKellar [2712]. These values are recommended in the Handbook [649] and were adopted in the present Handbook. Herzberg [2020] recommended similar values on the basis of the data of Harvey and Jenkins [1970].

The molecular constants of Li_2 in the states $C^1\Pi$ and $D^1\Pi$ are given in Table 262, taken from the work of Barrow and co-workers [662a]. Erroneous values were used for the vibrational constants, obtained by Sinha [3749] in the book of Herzberg [2020] and the Handbook [649].

LiH. The spectra of the molecule LiH have been investigated by Nakamura [3019], Crawford and Jorgensen [1214, 1215, 1216], Klemperer [2439] and Velasco [4076]. The absorption spectrum of lithium hydride and deuteride in the visible region and the near ultraviolet, corresponding to the transition $A^1\Sigma \leftarrow X^1\Sigma$, was obtained in the works [1214, 1215, 1216]. A detailed analysis of 26 bands in this system, recorded on an apparatus with a dispersion of 1 Å/mm during the heating of metallic lithium in a hydrogen atmosphere, was carried out by Crawford and Jorgenson [1215]. The constants of LiH in the states $X^1\Sigma$ and $A^1\Sigma$, found by these authors are presented in Table 262.* These constants were also given in the monograph of Herzberg [2020] and in the Handbook [649].

Because the spectrum of lithium hydride had been obtained in absorption, the observed bands corresponded to transitions from several lower vibrational levels of the ground state of LiH ($v'' = 0, 1, 2$ and

3). Hence, the vibrational constants found by Crawford and Jorgenson [1215] cannot describe the energy of the vibrational levels of this state with sufficient accuracy. It should be pointed out that although Crawford and Jorgenson determined three constants in the equation for $G_0''(v)$, the energy levels, calculated by means of the equation with the three constants, do not converge to any limit. At the same time, the energy levels, calculated without allowing for the constant $\omega_e y_e$, converge in the region of the dissociation limit of the state $X^1\Sigma$.

Velasko [4076] recently investigated the absorption spectrum of LiH and LiD in the region 2000-3200 Å and found a new band system, corresponding to the transition $B^1\Pi \leftarrow X^1\Sigma$. This spectrum was obtained on an apparatus with a dispersion of 0.85 Å/mm and heating of metallic lithium in an atmosphere of hydrogen and deuterium. In order to increase the thickness of the absorbing layer, a multi-pass cell was used, which enabled the optical path to be increased to 16 m. Six bands were observed in the spectra of the two isotopic molecules, corresponding to transitions to levels with $v' = 0, 1$ and 2 . A distinctive feature of all these bands was a discontinuity of their rotational structure at low values of the quantum number J ($v' = 0, J_{\max} = 8$; $v' = 1, J_{\max} = 5$ and $v' = 2, J_{\max} = 2$). Velasko explained the discontinuity of the rotational structure of the bands by a predissociation during the rotation and on the basis of the data thus obtained, determined the dissociation energy of LiH with great accuracy (see p. 1842). Using the molecular constants of the ground state of LiH, found in the work [1215], Velasko calculated the constants in the state $B^1\Pi$ which are given in Table 262.

In addition to the electronic spectra of LiH, Klemperer [2439] recently studied the rotational-vibrational spectrum of this molecule. The spectrum was investigated in the region $970-1500 \text{ cm}^{-1}$, where the

bands 1-0 and 2-1 were obtained. The authors of the work [2439] pointed out that the measured frequencies of the lines of LiH in the infrared spectrum agree within the limits of experimental error with the calculated constants, found in the work of Crawford and Jorgenson [1215].

LiF. Until recently, direct experimental measurements of the molecular constants of lithium fluoride were absent in the literature. On the basis of a study of the deviation of the molecular beams of lithium fluoride in an inhomogeneous electrical field, Trischka [4018] determined the product of the moment of inertia of the molecule LiF and its dipole moment and, estimating the latter, found that $1.44 \text{ \AA} \leq r_{\text{LiF}} \leq 1.65 \text{ \AA}$. Later on, Braunstein and Trischka [912] repeated these investigations and determined the bond length Li-F in the states $v = 0$ ($1.51 \pm 0.08 \text{ \AA}$) and $v = 1$ ($1.57 \pm 0.08 \text{ \AA}$) on the basis of the nuclear dipole-dipole interaction, found by them. On the basis of the intensity of the lines, corresponding to the values $v = 0$ and 1, the authors of the work [912] found approximate values for the vibration frequencies of the two isotopic modifications of lithium fluoride, equal to 756 ± 38 and $646 \pm 32 \text{ cm}^{-1}$ for Li^6F and Li^7F , respectively. However, these values of the vibration frequency of the two molecules are only rough approximations because of the inaccuracy of the measurement of the line intensity and temperature of the gas in the molecular beam, as pointed out by the authors of the work [912]. Even the ratio of the vibrational frequencies of the molecules Li^6F and Li^7F (1.17 ± 0.02), found in the work [912] differs greatly from the calculated on the basis of their masses, which is 1.06. Moreover, the values found by this method for other molecules (CsF and RbF), are much lower than those obtained in the investigations on the absorption spectra.*

Of the experimental studies, carried out in order to determine

the molecular constants of LiF, we must also mention the attempts to measure the interatomic distance in the molecule LiF by the method of electron diffraction [9, 65]. Electron diffraction patterns were obtained in these works from lithium fluoride vapor, but the analysis of the electron diffraction patterns, carried out with allowance being made for the data on the composition of the vapor of this compounds, enabled only the bond length Li-F in the dimeric molecule Li_2F_2 , but not the diatomic molecule LiF, to be determined. The values thus found was 1.69 ± 0.04 A.

Because of the absence of reliable experimental data on the constants of the molecule LiF, approximate estimates of these values were made in several works. The interatomic distance in the molecule LiF was estimated by Rittner [3444], Honig et al [2116] and Krasnov [260]. In the two firstmentioned works, $r_{\text{LiF}} = 1.53$ A, was found, in the last one $r_{\text{LiF}} = 1.584$ A. Thus, the results of the approximate estimates of r_{LiF} are in satisfactory agreement with each other and with the results of the experimental investigations [912, 4018]. The comparison of these values with the length of the bond Li-F in the molecule Li_2F_2 , found by an electron diffraction method in the work [65] also confirms the accuracy of the estimate because the length of the bond M-X in other halogen compounds of the alkali metals differ in the dimer and monomer by approximately 0.15-0.17 A.*

The frequency of the normal vibration of LiF was estimated by Khachkuruzov [436] on the basis of a method of estimating constants, proposed by him, based on their dependence in several analogous compounds of the elements (for example, of the diatomic fluorides) of a given group of the Periodic System on the number of s- and p- electrons of the elements, and was found to be 940 cm^{-1} . Benson and van der Hoff [750] carried out a quantum-mechanical calculation of the molecule Li^6F

and found $\omega_e = 1036 \text{ cm}^{-1}$. Rittner [3444] performed a theoretical calculation of the molecular constants of halogen compounds of alkali metals on the basis of the ionic model of their molecules, making allowance for the polarizability of the ions M^+ and X^- (the values found by Pauling [3200] for ions in crystal lattices were used for the polarizability of the ions). The vibration frequency of LiF thus found is 773 cm^{-1} , but this value must contain an error due to the utilization in the work [3444] of inaccurate constants for the molecules of the other halogen compounds of the alkali metals, in particular, their interatomic distances, taken from the inaccurate data of Maxwell et al [2818]. Huff, Gordon and Morrell [2142], based on a comparison of the vibrational frequencies of molecules which are isoelectric with LiF (BeO, BN, and C_2), adopted the value $\omega_e = 1343 \text{ cm}^{-1}$. This last value was known to be erroneous because it is similar to the vibration frequency of LiH, while the vibration frequencies of the diatomic fluorides of all elements are considerably lower than the vibration frequencies of the hydrides of these elements.

A sound choice between the above-given values is difficult. Hence, the authors of the Handbook carried out another estimate of the vibration frequency of LiF on the basis of a comparison of the force constants of the fluoride bonds of the elements of group 11 of the Periodic system as a function of the number of the valency electrons, and also by the method of Guggenheimer [1880] with $r_{\text{LiF}} = 1.53 \text{ \AA}$. Both estimates gave values close to that proposed by Khachkuruzov and in the work of Benson and van der Hoff. The value 1000 cm^{-1} , which is the average of the four estimates, was adopted in the first edition of the Handbook and also during the preparation of the present edition. However, after the work on the present chapter had already been completed, Koryazhkin, Tatevskiy and Kharitonov [52, 250] carried out a new the-

oretical calculation of the constants of LiF and at the same time Vasilevskiy and Baykov [19b, 118, 118a] completed an experimental investigation on the infrared spectrum of this molecule.

In the work of Koryazhkin, Tatevskiy and Kharitonov [52, 250], the vibrational constants of LiF and all other diatomic compounds of the halogens with alkali metals were calculated theoretically for the ionic model of their molecules, using the relation proposed by Rittner [3444] for the interaction energy of the ions. However, in contrast to the latter, the polarizabilities of the Me^+ and X^- ions were not chosen in this work [52, 250] on the basis of the data obtained for the crystal lattice, but were calculated on the basis of the dipole moments and interatomic distances of six gaseous molecules MeX and the use of a comparative method for the estimation of the missing values.* Introducing additional hypotheses on the relationship between the interaction energies of the ions and the dissociation energies of these molecules, and also on the possibility of approximation of their potential energy in the minimum of the potential curve of the Morse functions, Koryazhkin, Tatevskiy and Kharitonov obtained good agreement between the calculated values of ω_e and $\omega_e x_e$ and the most reliable experimental data for several molecules of alkali-halogen compounds. In the case of LiF, the values of these constants found by them are 885 and 5.41 cm^{-1} .

The infrared emission spectrum of lithium fluoride vapor was obtained for the first time in the work of Vasilevskiy and Baykov [118]. The spectrum was studied in the region $210\text{-}5000 \text{ cm}^{-1}$. It consisted of the edges of four bands in the region $925\text{-}975 \text{ cm}^{-1}$, corresponding to the transitions $\Delta v = 1$ and a partially resolved rotational structure of the band 1-0 in the region $750\text{-}800 \text{ cm}^{-1}$. The short-wave boundary of the spectrum, which at $\omega_e x_e > 0$ and $\alpha_1 > 0$ is the edge of the R-Branch

of the band 1-0, had the wave number $975.2 \pm 2 \text{ cm}^{-1}$ according to the data of Vasilevskiy and Baykov. The authors of this work carried out an approximate analysis of the rotational structure on the basis of approximate values of the rotational constants of the molecule LiF and using the method of consecutive approximations for improving the numbering of the lines. The rotational constants of LiF, obtained by Baykov and Vasilevskiy, are given in Table 262 and were adopted in the present Handbook. According to an estimate by the authors of the works [19a, 118 and 118a], the uncertainty in the value of B_r does not exceed $\pm 0.09 \text{ cm}^{-1}$.

By means of the rotational constants of LiF thus determined, Vasilevskiy and Baykov calculated the value of the quantum number J , corresponding to the edge of the band 1-0, and, determining the difference of the wave numbers of the edge and the beginning of the band from the relation

$$v_{\text{кант}}^{i-0} - v_0^{i-1} = 2(B_e - \alpha_1) \cdot m_{\text{кант}} - \alpha_1 m_{\text{кант}}^2 - 4D_0 m_{\text{кант}}^3$$

where $m_{\text{кант}} = J_{\text{кант}} + 1$, calculated the wave number of the beginning of this band $\Delta G_{1/2} = 881 \text{ cm}^{-1}$. The anharmonicity constant $\omega_e x_e$ was determined by the difference of the wave numbers of the edges of the bands 1-0 and 2-1, taking this difference as being equal to $2\omega_e x_e + 2\alpha_1 m_{\text{кант}}$. Thus, $\omega_e x_e = 7.3$ and $\omega_e = \Delta G_{1/2} + 2\omega_e x_e = 896 \text{ cm}^{-1}$ was found. The authors of the works [118, 118a] estimated the uncertainty in the values of $\omega_e x_e$ and ω_e found by them to be ± 0.5 and $\pm 5 \text{ cm}^{-1}$. Actually, however, the accuracy of the vibrational constants, found in these works, is less and depends greatly on the accuracy of determination of the values of $m_{\text{кант}}$. If the error in the constants B_e , α_1 and D_0 thus found is taken as ± 0.03 ; $\pm 1 \cdot 10^{-3}$ and $\pm 1 \cdot 10^{-6} \text{ cm}^{-1}$, which is probably less than the real error in these values, the magnitude of $m_{\text{кант}}$ can be calculated only with an accuracy of ± 4 and the beginning of the band

1-0 with an accuracy of about 10 cm^{-1} . Because the band edges in the spectrum of lithium fluoride were measured with an accuracy of $\pm 2 \text{ cm}^{-1}$, the uncertainty in the determination of the wave number of the band head cannot be less than 12 cm^{-1} , and the error in the vibration frequency is $10\text{-}15 \text{ cm}^{-1}$.

It should be pointed out that the vibrational constants of LiF, found by Vasilevskiy and Baykov, cannot be used for calculating the thermodynamic functions of lithium fluoride at high temperatures without introducing into the equation for $G(v)$ a term, which is proportional to v^3 or v^4 , because the dissociation energy of LiF corresponding to it, calculated by means of the relation (1.13) is approximately $27,000 \text{ cm}^{-1}$, while the Handbook adopted (see page 1847) 137 kcal/mole or about $49,000 \text{ cm}^{-1}$. Hence, the authors of the Handbook calculated the vibrational constants of this molecule by solving the system of equations $D_e = \omega_e^2/4\omega_e x_e$ and $\omega_e = \Delta G_{1/2} + 2\omega_e x_e$, taking $D_e = 49,000 \pm 500$ and $\Delta G_{1/2} = 882.7 \pm 10 \text{ cm}^{-1}$ (according to the data of [19a, 118a]). The corresponding values of the constants $\omega_e = 891 \pm 10$ and $\omega_e x_e = 4 \pm 2 \text{ cm}^{-1}$ were adopted in the Handbook and are given in Table 262. They agree satisfactorily with those proposed by Vasilevskiy and Baykov and with the results of the theoretical calculation of Koryazhkin and Tatevskiy [52, 250].

After the calculations of the thermodynamic functions of LiF on the basis of the constants, given in Table 262 had been completed, the work of Vidale [4105a] was published, in which as in the work of Vasilevskiy and Baykov, the infrared spectrum of lithium fluoride vapor had been investigated. However, in contrast to Vasilevskiy and Baykov, Vidale investigated both the emission and absorption spectrum of LiF, carrying out the investigation on an apparatus with a grating with higher dispersion, which enabled the rotational structure of the

bands 1-0 and 2-1 in the region $962-757 \text{ cm}^{-1}$, to be resolved. Like Vasilevskiy and Baykov, Vidale could not determine the wave numbers of the beginnings of the bands because of their overlapping and carried out the numbering of the lines of the rotational structure on the basis of the relative intensity of these lines, the estimated value of $B_e = 1.377 \text{ cm}^{-1}$ and the furnace temperature, which was known to him. The accuracy of his numbering of the lines, Vidale estimated to be ± 2 . The constants of LiF found by him ($\omega_e = 906.2 \pm 1.5$, $\omega_e x_e = 7.90 \pm 0.01$, $B_e = 1.378 \pm 0.010$, $\alpha_1 = 0.01971 \pm 0.00016$ and $r_e = 1.545 \pm 0.006 \text{ cm}^{-1}$) agree with those proposed by Vasilevskiy and Baykov within the limits of the measurement error. However, because of the use of an apparatus with high dispersion, the rotational constants found by Vidale, are slightly more accurate than those given in Table 262 on the basis of the data of [19a and 118a]. The vibrational constants, calculated by Vidale, like those obtained by Vasilevskiy and Baykov, are not suitable for calculating the thermodynamic functions of LiF because the levels of the vibrational energy calculated by means of them converge in the region $29,450 \text{ cm}^{-1}$. The values of ω_e and $\omega_e x_e$, calculated by simultaneous solution of the equations $D_e = \omega_e^2 / 4\omega_e x_e$ and $\omega_e = \Delta G_{1/2} = 890.4 \pm 1.5 \text{ cm}^{-1}$ (on the basis of the data of [4105a]), are 898.2 and 4.1 cm^{-1} , respectively.

In 1961, the work of Linevsky [2619a] was published, who investigated the infrared spectrum of the molecule LiF, frozen in matrices. The band frequencies, observed during the work with argon, krypton and xenon matrices, were situated in the region $823-840 \text{ cm}^{-1}$, and in the work with nitrogen matrices, in the region 778 cm^{-1} .

LiCl. The continuous absorption spectrum of lithium chloride in the region 2400 \AA was first observed by Muller [2968]. The spectrum of LiCl was not observed in subsequent investigations on the spectra of

the halogen compounds of the alkali metals [648, 2596, 3811] in which diffuse bands in the visible region and in the ultraviolet were observed for several molecules. Likewise, attempts to investigate the microwave spectrum of LiCl [2116] were also unsuccessful.

Hence, Rittner [3444], Honig et al [2116], Krasnov [260] and the authors of the Handbook carried out approximate estimates of the vibration frequency and interatomic distance of this molecule and obtained similar values of $\omega_e = 700 \pm 25 \text{ cm}^{-1}$ and $r_{\text{LiCl}} = 2.00 \pm 0.03 \text{ \AA}$. The last value is in good agreement with the length of the bond Li-Cl in the molecule Li_2Cl_2 , found by Akishin and Rambidi et al [65] during their investigations on lithium chloride vapor by the electron diffraction method, $2.17 \pm 0.08 \text{ \AA}$ (see page 1800).

Klemperer and Rice [2444] in 1957 were the first to obtain the infrared emission spectrum of the halogen compounds of lithium in the gas state, including the infrared spectrum of LiCl. The spectrum was obtained in the region 650 cm^{-1} on a low-dispersion apparatus and had the appearance of a single band, although in reality, because of the high temperature of the lithium chloride, it consisted of several bands corresponding to transitions from $\Delta v = 1$. The shortwave boundary of the observed band at $\omega_e x_e > 0$ is the edge of the band 1-0. Having measured the bands of LiBr and Lil and knowing the rotational constants of these molecules [2116], the authors of the work [2444] calculated the wave numbers of the beginnings of the bands and the values of ω_e of these molecules.*

As the rotational constants of LiCl are unknown, an analogous determination of the vibration frequency of this molecule was impossible. Hence, Klemperer and Rice determined it on the basis of a comparison of the difference in the wave numbers of the edges and the vibration frequencies ($\nu_{\text{max}} - \omega_e$) of LiBr and Lil and extrapolated this

value to LiCl. Using for LiCl $\nu_{\text{max}} = 688.9$ and $\nu_{\text{max}} - \omega_e = 26.5 \text{ cm}^{-1}$, the authors of the work [2444] found the vibration frequency of LiCl to be equal to 662.4 cm^{-1} .

Later on, Berry and Klemperer [772] and Koryazhkin, Tatevskiy and Kharitonov repeated the investigation of the absorption spectrum of lithium chloride in the ultraviolet and in the region $\lambda 2800 \text{ \AA}$, against the background of the continuous spectrum, were the first to obtain several diffuse bands. The intervals between these bands were of the order of $550\text{-}620 \text{ cm}^{-1}$. The authors of these two works interpreted these bands as being connected with a transition into an unstable excited state of the molecule LiCl with high vibrational levels ($v'' \approx 10$) of the ground state $X^1\Sigma$ and arrived at the conclusion that the data obtained by them confirmed the vibration frequency of LiCl, found by Klemperer and Rice. This value, whose uncertainty may attain 10 cm^{-1} because of several additional errors, has been adopted in the Handbook.

In the work of Koryazhkin and Tatevskiy [250] (see above, page 1802) the values 659 and 4.73 cm^{-1} , respectively, were found for the constants ω_e and $\omega_e x_e$ of the molecule LiCl on the basis of theoretical calculations.

The rotational constant of LiCl, adopted in the present Handbook and given in Table 262, has been calculated for $r_{\text{LiCl}} = 2.02 \text{ \AA}$ according to the estimate given by Honig et al [2116]. It should be pointed out that a similar value of the interatomic distance (2.018 \AA) has been adopted in the work of Klemperer and Rice [2444] but the rotational constant (0.625 cm^{-1}) given in the paper of these authors corresponds to an interatomic distance of 2.15 \AA . The authors of the work [2444] obviously made a mistake in the calculation of B_0 or a printing error occurred in the paper.

Table 262, in addition to the above-mentioned constants, gives the

values for the anharmonicity constant $\omega_e x_e$, the interaction of the rotation and vibration α_1 and the centrifugal stretching of the molecule LiCl. The corresponding constants were calculated by means of the relations (1.13), (1.38) and (1.36) on the basis of the adopted values $\omega_e = 662$, $B_e = 0.71$, and $D_0 = 38,600 \text{ cm}^{-1}$ (see page 1854).*

LiO. The spectrum of the molecule LiO has not yet been observed and experimental data on its vibrational and rotational constants are lacking in the literature. Also unknown are the analogous data for the diatomic oxygen compounds of the other alkali metals. Hence, a sound estimate of the molecular constants of LiO is a very difficult task.

In the present Handbook, the estimate of the constants of LiO was made on the basis of the assumption, that the interatomic distance in this molecule is 0.02 Å less than the bond length Li-O in the molecule Li_2O^{**} and equal to 1.80 Å. The rotational constant of this molecule and its vibration frequency, given in Table 262, were calculated by the authors of the Handbook, using $r_{\text{LiO}} = 1.80 \text{ Å}$. The value of ω_e was calculated by the method, proposed by Guggenheimer (see page 88). The constants thus obtained were also estimated by other methods, in particular, by a comparison of the force constants of the molecular bond in the oxides of the elements of group II of the Periodic System as a function of the number of valency electrons of the element and an analogous comparison of the rotational constants of these molecules. It may be assumed that the uncertainty in the adopted interatomic distance of the molecule LiO does not exceed 0.03 Å and that of the vibration frequency, 100-150 cm^{-1} .

The problem of the type of electronic ground state of LiO cannot be resolved without a study of the spectra of this molecule or the diatomic oxide of some other alkali metal. According to the correlation

rules of Wigner-Witmer, the ground state of LiO should be the state $^2\Sigma$ or $^2\Pi$. Because the molecule LiO is isoelectronic with the sodium atom, which has the ground state 2S , it has been assumed in the present Handbook that the ground state of this molecule is the state $^2\Sigma$.

Following the completion of the calculations of the thermodynamic functions of the lithium compounds for the present Handbook, the work of Berkowitz [758] on the investigation of the heat of sublimation of lithium oxide was published. The vibration frequency of LiO was estimated in this work on the basis of the assumption that the molecule LiO is isoelectronic with the molecule BeF and that the force constants and interatomic distances in the two molecules are the same. This assumption, however, is based on a misunderstanding, because the molecule LiO has seven valency electrons and BeF nine. Hence the authors of the work [758] obtained values for the force constant and the vibration frequency of LiO which were much too high and a lower value for the interatomic distance.

Li₂O and LiOH. The spectra of lithium oxide and hydroxide in the gaseous state have not yet been observed. The structure of the lithium oxide molecule has been investigated by the electron diffraction method by Akishin, Rambidi et al [64]. It was found in this work that the length of the Li-O bond in the molecule Li₂O is 1.82 Å and the angle Li-O-Li is 110°. The last value was determined by the position of the maximum, corresponding to the distance Li-Li, on the radial distribution curve and was thus determined with insufficient accuracy. Table 263 gives the product of the principal moments of inertia of Li₂O calculated on the basis of the above-mentioned structural parameters. The same Table gives the analogous values for LiOH, obtained by the authors of the Handbook for $r_{LiO} = 1.82$ Å (as in Li₂O), $r_{OH} = 0.95$ Å (as in H₂O) and $\angle Li-O-H = 105^\circ$. The uncertainty in the adopted geome-

trical parameters of the molecules Li_2O and LiOH does not exceed ± 0.03 A for the bond Li-O, ± 0.01 A for the bond O-H and 10° for the angles Li-O-Li and Li-O-H.

TABLE 263

Adopted Values of the Molecular Constants of Li_2O and LiOH

Молекула A	ν_1	ν_2	ν_3	$I_A I_B I_C$	σ
	cm ⁻¹			10^{-21} (g·cm ³) ²	
Li₂O	780	350	830	44,53	2
LiOH	3750	900	1100	1,065	1

A) molecule

Owing to the absence of experimental data, the fundamental frequencies of the molecules Li_2O and LiOH were calculated by means of the equations for the field of the valency forces [equations (P4.30) and (P4.27)] and the estimated values of the force constants. The force constants of the Li-O and O-H bond used in the calculation were the same as in the molecules LiO and OH ($1.82 \cdot 10^5$ and $7.76 \cdot 10^5$ dyne·cm⁻¹, respectively). The value of the constant f_α/d^2 in the molecule Li_2O was taken to be $0.1 f_d$ or $0.18 \cdot 10^5$ dyne·cm⁻¹ as for other nonlinear molecules of the type XY_2 (see [152], Table 40). The deformation constant f_α of the molecule LiOH was taken as $0.73 \cdot 10^{-11}$ dyne·cm/degree as a result of a comparison of the values of this constant in the molecules HOCl , DOCl and HDO .

A valid estimate of the probable errors in the calculated values of the fundamental frequencies of Li_2O and LiOH is very difficult. It may be assumed that it does not exceed 10-20%.

Following the completion of the calculations on the thermodynamic functions of the lithium compounds by the authors of the Handbook, the

work of Berkowitz, Chupka, Blue and Margrave [758] became known, in which an estimate of the constants of Li_2O had been made, using a force constant of the Li-O bond, equal to that of the molecule BeF_2 and the constant f_α/d^2 as $0.65 \cdot 10^5$ dyne $\cdot\text{cm}^{-1}$. The values $\nu_1 = 1270$, $\nu_2 = 740$ and $\nu_4 = 1650$ cm^{-1} , found by these authors, are wrong, because the force constants of the molecule Li_2O should be much smaller than the values adopted in the work [758].

§112. THERMODYNAMIC GAS FUNCTIONS

The thermodynamic functions of lithium and its compounds were calculated for temperatures from 293.15 to 6000°K. The calculations were carried out on the basis of the constants, adopted in the preceding paragraph, without allowing for the difference in the constants of the isotope modifications of the molecules of the respective gases.

The calculated values of the thermodynamic functions of lithium and its compounds in the ideal gas state are given in Tables 346, 347, 348, 349, 351, 353, 355, 357 and 359 of Volume II of the Handbook. In view of the absence of the corresponding experimental data, corrections for the molecular interactions of gaseous lithium and its compounds are not discussed in the Handbook.

Li. The thermodynamic functions of monatomic lithium in the gas state, given in Table 346 (II), were calculated by means of Eqs. (II.22) and (II.23). The translational components in the values of the thermodynamic functions of monatomic lithium were calculated by means of the relations (II.8)-(II.9), using $A_\phi = 1.5088$ and $A_S = 3.4595$ cal/g-atom \cdot degree and the electronic components, by means of Eqs. (II.20) and (II.21) by direct summation on the basis of the data given in Table 261. In order to simplify the calculation over the entire temperature range up to 3600°K, all the states corresponding to the electronic configurations with main quantum numbers of the valency electron of

$n \leq 10$ were allowed for. Because at these temperatures the contribution of the levels with the values $n = n_{\max} + a$, where n_{\max} has been found by means of the relation (II.18) and $a \leq 3$, is negligibly small, the simplification does not introduce any error into the calculated values. At higher temperatures, the contribution of the level corresponding to the value $n = 11$ was taken into account.

At temperatures under 5000°K the accuracy of the calculated thermodynamic functions is determined by the accuracy of the physical constants used in the calculations. The corresponding errors in the values of Φ_T^* and S_T° do not exceed 0.003 - 0.004 cal/g-atom·degree. The errors connected with the approximate estimation of the excitation energies of several levels, become noticeable at temperatures over 5000°K , but do not exceed 0.001 - 0.002 cal/g-atom·degree.

The thermodynamic functions of monatomic lithium have been calculated by several authors. Tables of the thermodynamic functions were calculated for a wide range of temperatures by Overstreet [3158] (Φ_T^* to 3500°K), Huff, Gordon and Morrell [2142] (to 6000°K), Evans, Munson et al [1513] (to 3500°K), Kolsky [2462] (to 8000°K). The data of [1513] were given in the Handbook of the U. S. Bureau of Standards [3680] and in the book [3894]. At low temperatures (up to 3000°K), the data of all these authors are in good agreement. At temperatures over 3000°K , the calculation results of the authors of the work [2142] are not accurate because they allowed only for the five lower levels of the lithium atom. The corresponding errors in the values of Φ_{6000}^* and S_{6000}° are 0.043 and 0.478 cal/g-atom·degree. The data of [2462] are in good agreement with those given in Table 346 (II) at temperatures under 5000°K . However, at higher temperatures, systematic differences between them are observed. These differences are due, on one hand, to the fact that all the energy levels of the Li atom, given by Moore

[2941], including those with $n > 11$, were allowed for, and, on the other hand, all the experimentally not observed levels with values of L from 5 to 10 were not taken into account.

The differences between the data in the first and present editions of the Handbook become important above 4000°K and attain 0.02 and 0.02 cal/g-atom·degree in the values of Φ_{6000}^* and S_{6000}° .

Li⁺. The thermodynamic functions of Li^+ , given in Table 347 (II), were calculated by means of Eqs. (II.22) and (II.23). The translational components were calculated by means of the same relation, which was used for the calculation of the functions of monatomic lithium. The electronic components are zero at all considered temperatures because the ground state of the Li^+ ion is the state ^1S , and the excitation energies of the other states exceed $100,000\text{ cm}^{-1}$. The thermodynamic functions of Li^+ have been calculated in the works of Huff et al [2462] for temperatures to 6000°K and Green et al [1851] to $50,000^\circ\text{K}$. The values, presented in Table 347 (II), agree with those obtained in these works and in the first edition of the Handbook within limits, determined by the differences in the physical constants.

Li₂. The thermodynamic functions of diatomic lithium, given in Table 348 (II), were calculated by means of Eqs. (II.34) and (II.35). The translational components in the values of the thermodynamic functions were found on the basis of the relations* (II.8)-(II.9), using $A_\phi = -0.8199$ and $A_S = 4.1482$ cal/mole·degree. The intramolecular components were calculated by means of the constants of the molecule Li_2^7 , given in Table 262. Neglecting the differences in the constants of the individual isotope modifications of the molecules Li_2 (Li_2^6 , Li_2^7 , Li^6Li^7) do not cause errors exceeding those of the calculated values of the thermodynamic functions.

The molecule Li_2 has a low dissociation energy and a low vibration

frequency in the ground state and excited states with low energies. Hence, when calculating the thermodynamic functions of Li_2 even at relatively low temperatures, one must take into account several effects which play an important part with other gases only at temperatures of the order of 8000-12,000°K. The intramolecular components in the values of Φ_T^* and the entropies were calculated by means of Eqs. (II.121)-(II.132). The statistical sum Q^X of the vibrational and rotational energy levels of the ground state and its derivative with respect to the temperature were calculated by direct summation of the energy levels, found on the basis of the constants, given in Table 262. The maximum quantum numbers ν and J are given in Fig. 21. The values of M_A , N_A , M_B and N_B in Eqs. (II.131) and (II.132) were calculated by the method of Gordon and Barnes [relations (II.137)-(II.138)]. Because the states $A^1\Sigma$ and $B^1\Pi$ of the molecule Li_2 have low dissociation energies, corrections were applied in the calculation of these values to limit the number of rotational levels, calculated by means of Eqs. (II.59)-(II.60).

The components of the third excited state of the molecule Li_2 with an energy of about $32,000 \text{ cm}^{-1}$ were calculated by means of the relations (II.126) and (II.127). The state $D^1\Pi$ was not included in the calculation (the calculation was carried out prior to publication of the work [662a]).

The main error in the calculated values of the thermodynamic functions of diatomic lithium is due to the insufficient accuracy of the adopted constants of Li_2 in the states $X^1\Sigma$, $A^1\Sigma$ and $B^1\Pi$ and also the approximate method of calculating the components of the two following states. However, all these errors, show up only at high temperatures. The uncertainty in the values Φ_{3000}^* does not exceed $\pm 0.02 \text{ cal/mole}\cdot\text{degree}$, but at 6000°K it can attain $\pm 0.5 \text{ cal/mole}\cdot\text{degree}$.

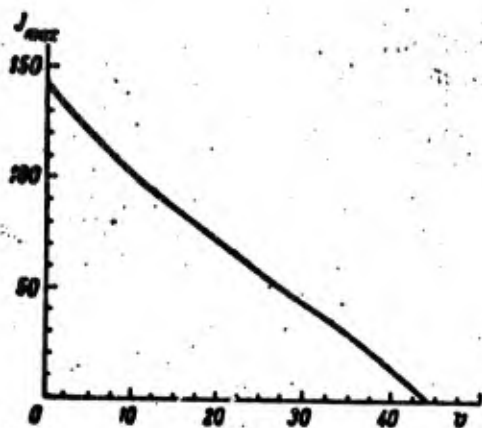


Fig. 21. J_{\max} as a function of v for the electronic state $X^1\Sigma_g^+$ of the molecule Li_2

The thermodynamic functions of Li_2 had been calculated previously by Gordon [1805] up to 2000°K , Evans and coworkers [1513] and the U. S. Bureau of Standards [3680] to 1500°K . The best agreement is observed between the calculation results of the present Handbook and the data [1805]

obtained by the tabular method of Gordon and Barnes. The maximum difference in the values of Φ_T^* occurs at 2000°K , attaining $0.02 \text{ cal/mole}\cdot\text{degree}$. The differences between the data of the present Handbook and the results of two other calculations, carried out by the method of Mayer and Goeppert-Mayer, are considerably greater, amounting to 0.12 - $0.15 \text{ cal/mole}\cdot\text{degree}$ in the value S_{1500}^0 . The results of Gordon's calculation are given in the book of Zeise [4384]; the book of Stull and Sinke [3894] gives the data of Evans et al. [1513].

In the first edition of the Handbook, the thermodynamic functions of diatomic lithium were calculated by the method of Gordon and Barnes. The difference between the values, given in the present and first edition, is due to the difference in the calculation methods for the state $X^1\Sigma$, the limitation of the number of rotational levels of the state $A^1\Sigma$ and $B^1\Pi$ and the slight change in the vibrational constants of the state $X^1\Sigma$. Although at low temperatures the corresponding differences are slight, not exceeding $0.02 \text{ cal/mole}\cdot\text{degree}$ in the values of Φ_T^* at $T \leq 3000^\circ\text{K}$, they increase rapidly at higher temperatures and attain 1.6 and $3.8 \text{ cal/mole}\cdot\text{degree}$ in the values Φ_{6000}^* and S_{6000}^0 .

TABLE 264

Constants for the Calculation of the Thermodynamic Functions of LiH, LiO, LiF and LiCl

Вещество A	θ	$x \cdot 10^3$	$\beta_1 \cdot 10^3$	$\beta_2 \cdot 10^4$	$\frac{q_0}{T}$	$\frac{d_0 \cdot 10^6}{T}$	$\frac{f_0 \cdot 10^{10}}{T^2}$	C_Φ	C_S
	град B				град ⁻¹ B		град ⁻² B	кал/моль·град C	
LiH	2022,1	16,5048	2,8733 ^a	9,054 ^a	0,093859	21,6	-7,86	-5,8048	1,1507
LiO	1150,8	—	—	—	—	—	—	2,5674	9,5229
LiF	1282	4,4893	1,2096	1,4632	0,512634	9,54	—	1,0037	8,0492
LiCl	952,48	3,3233	0,4935	0,243	0,979948	9,12	—	3,8458	10,8013

A) Substance; B) degree; C) cal/mole·degree; a) calculated without allowing for α_2 in the expression for B_V . Calculation by means of Eqs. (II.58) gives values of $\beta_1 = 2.8786 \cdot 10^{-2}$ and $\beta_2 = 9.299 \cdot 10^{-4}$.

LiH. The thermodynamic functions of gaseous lithium hydride, given in Table 353 (II), were calculated by means of Eqs. (II.161) and (II.162). The values $\ln \Sigma$ and $T \partial/\partial T \ln \Sigma$ were calculated by the method of Gordon and Barnes [Eqs. (II.137) and (II.138)] on the basis of the above indicated molecular constants. Table 264 gives the values of C_Φ and C_S and also the values of the coefficients in Eqs. (II.137) and (II.138), calculated by means of the molecular constants of LiH in the state $X^1\Sigma$.

The components of the state $A^1\Sigma$ of the molecule LiH were calculated by means of Eqs. (II.126) and (II.127); the components of the state $B^1\Pi$ were not taken into account because their contribution is fairly small at temperatures up to 6000°K. As in the calculation of the thermodynamic functions of the other lithium compounds the difference between the constants of the individual isotope modifications of LiH was not taken into account in the calculation of the thermodynamic functions of LiH. This simplification does not give errors in the va-

lues of the thermodynamic functions of an equilibrium isotope mixture of lithium hydride of more than 0.02 cal/mole·degree. The molecule LiH has a relatively low dissociation energy (see page 184), hence it is essential in the calculation of the thermodynamic functions of lithium hydride by approximate methods, to introduce a correction for the limitation of the number of the rotational energy levels. The corresponding correction was calculated by means of the relations (II.59) and (II.60) and amounts to 0.17 and 0.78 cal/mole·degree at 6000°K for the values of Φ_T^* and S_T^c , respectively.

The main error in the calculated values of the thermodynamic functions of lithium hydride is due to the absence of experimental data on the energies of the vibrational levels of the ground state of LiH with $v > 3$.* The error in the values of Φ_T^* at 298.15; 3000 and 6000°K is of the order of 0.02; 0.05 and 0.2 cal/mole·degree, respectively.

The thermodynamic functions of lithium hydride have been calculated earlier by Kelley [2363] to 2000°K and Huff et al. [2142] to 6000°K. These two calculations were carried out, using the approximate model of the rigid rotator-harmonic oscillator, which causes important differences between the data of the Handbook and the calculation results of these authors at high temperatures. The corresponding differences from the data [2142] in the values of Φ_{6000}^* and S_{6000}^c attain 0.7 and 1.5 cal/mole·degree.

The differences between the values of the thermodynamic functions of lithium hydride, given in the first and present Handbook editions, attain a magnitude of the order of 0.2 and 0.8 cal/mole·degree in the values Φ_T^* and S_T^c and are due to the fact that a correction for the limited number of rotational levels of the molecule LiH has not been applied in the first edition.

LiO. The thermodynamic functions of lithium monoxide, given in

Table 349 (II), were calculated by means of Eqs. (II.161) and (II.162) on the approximate model of the rigid rotator-harmonic oscillator; the values of $\ln \Sigma$, $\ln \Delta$ and their derivatives were taken as zero. The calculation was carried out by means of the constants, adopted in Table 262. Table 264 gives the values of C_ϕ and C_S , which were used in the calculation of the characteristic temperature for the interpolation by means of the harmonic oscillator Table. The components $R \ln 2$ were included in the constants C_ϕ and C_S to allow for the multiplet nature of the ground state $^2\Sigma$ of the molecule LiO.

The thermodynamic functions of LiO were calculated by means of estimated values of the molecular constants. The main errors in the functions, given in Table 349 (II), are due to the inaccuracy of the vibration frequency (up to 0.3-0.4 cal/mole·degree in Φ_T^* at 2000-6000°K) and the use of an approximate calculation method because of the lack of data on the anharmonicity constants and the vibrational and rotational interaction constants of LiO (up to 0.2 and 0.5 cal/mole·degree in Φ_T^* at 3000 and 6000°K, respectively). The total error in the calculated values $\Phi_{298.15}^*$, Φ_{3000}^* and Φ_{6000}^* is ± 0.1 , ± 0.5 and ± 2 cal/mole·degree, if the ground state of the molecule LiO is the state $^2\Sigma$, as assumed in the Handbook.

Data on the thermodynamic functions of lithium monoxide over a wide range of temperatures are lacking in the literature. The work of Berkowitz, Chupka et al. [758] gives the values of the thermodynamic functions of LiO at 1300, 1400 and 1500°K, but the authors of the work [758] used incorrect values of the molecular constants of LiO for the calculation of these magnitudes (see page 1809). Even after the application of the corrections for the erroneous value of r_{LiO} , used in the work, the differences exceed 2 cal/mole·degree in S_{T1}° . These differences are due to the incorrect value of ω_e , used by the authors of the

work [758] and the fact that the statistical weight of the ground state of LiO has been neglected.

The thermodynamic functions of lithium monoxide, given in the first and present editions of the Handbook, are identical.

LiF. The thermodynamic functions of gaseous lithium fluoride, given in Table 357 (II), were calculated by means of Eqs. (II.161) and (II.162). The values of $\ln \Sigma$ and $T \partial/\partial T \ln \Sigma$ in these equations were calculated by the method of Gordon and Barnes [Eqs. (II.137) and (II.138)] on the basis of the above indicated constants without application of corrections for the limited number of rotational energy levels of the molecule LiF. Table 264 gives the values of C_p , C_s , the values of the coefficients in the Eqs. (II.137) and (II.138) and also the values of θ and x for the interpolation by means of the anharmonic oscillator Tables, calculated on the basis of the adopted values of the molecular constants. Allowance was not made in the calculation for the excited electronic states of the molecule LiF.

The main errors in the calculated values of the thermodynamic functions of lithium fluoride are due to the lack of reliable data on the values of the vibrational constants of LiF. The differences in the values of Φ_T^* , calculated by means of the constants, obtained in the work of Vasilevskiy and Baykov (see page 1804) and adopted in the present Handbook, amount to 0.05 and 0.12 cal/mole·degree in Φ_{3000}^* and Φ_{6000}^* . Analogous differences from the values, calculated by means of the constants, found by Vidale [4105a], amount to 0.15 and 0.28 cal/mole·degree.

It should be pointed out that the errors due to neglecting the necessary limitation of the number of rotational levels and the need to take into account the components of the electronic states of LiF are an order of magnitude smaller than the above given values. The total

error in the calculated values of $\Phi_{298.15}^*$, Φ_{3000}^* and Φ_{6000}^* is estimated to be ± 0.93 , ± 0.1 and ± 0.2 cal/mole·degree, respectively.

The thermodynamic functions of lithium fluoride were calculated by Huff, Gordon and Morrell [2142] to 6000°K, using the approximate model of the rigid oscillator-harmonic rotator, by means of estimated constants (see page 180); by Altman [526] to 6000°K by means of the constants, found by Braunstein and Trischka [912] (see page 863) and as a result of estimates, on the basis of the approximate model of the rigid rotator-harmonic oscillator with subsequent application of corrections for the anharmonicity of the vibrations and the centrifugal stretching by the method of Pennington and Kobe; by Wilkins [4269a] to 6000°K by the method of Pennington and Kobe on the basis of constants similar to those found by Vidale [4105a]; by Brewer and Brackett [918a] (Φ_T^* to 2000°K) on the basis of the approximate rigid rotator-harmonic oscillator model; by Licirek and Papousek [2497b] to 3500°K by the method of Mayer and Goeppert-Mayer. The results of the calculations [2142, 526], carried out on the basis of incorrect molecular constants of LiF, contain considerable errors and differ from those given in the Handbook by 1.6 and 0.4 cal/mole·degree, respectively, in the values of Φ_{6000}^* . The data of Wilkins agree with those given in Table 357 (II) within the limits of 0.1 cal/mole·degree in the values of Φ_T^* and 0.17 cal/mole·degree in the values of S_T° , the data of the work [2497b] within the limits of 0.03 cal/mole·degree.

The differences between the values of Φ_T^* and S_T° , given in the first and present editions, attain 0.6 and 1.0 cal/mole·degree because of the circumstances that the calculations in the first issue were carried out on the basis of estimated constants for LiF.

LiCl. The thermodynamic functions of gaseous lithium chloride, given in Table 359 (II), were calculated by means of Eqs. (II.161) and

(II.162). The values $\ln \Sigma$ and $T \partial/\partial T \ln \Sigma$ in these equations were calculated by the method of Gordon and Barnes [Eqs. (II.137) and (II.138)] on the basis of the molecular constants, given in Table 262. The calculation was carried out without introducing corrections for the limited number of rotational energy levels. Table 264 gives the values of the constants C_ϕ and C_S , the coefficients in Eqs. (II.137) and (II.138) and also the values of θ and x for interpolation on the basis of the anharmonic oscillator Tables, calculated by means of the molecular constants of LiCl.

The main errors in the calculated values of the thermodynamic functions of lithium chloride are connected with the lack of reliable data on the constants of the molecule LiCl. The discrepancies between the values of Φ_T^* , calculated by means of the adopted vibrational constants and the vibrational constants, found in the work [2443b], amount to 0.11 and 0.22 cal/mole·degree in Φ_{3000}^* and Φ_{6000}^* . The total error in the values $\Phi_{298.15}^*$, Φ_{3000}^* and Φ_{6000}^* , given in Table 359 (II), is estimated as ± 0.05 ; ± 0.1 and ± 0.2 cal/mole·degree, respectively.

The thermodynamic functions of lithium chloride were calculated in the works of Altman [526] and Wilkins [4269a] (for $T \leq 6000^\circ\text{K}$) on the basis of the rigid-harmonic oscillator model, with corrections being applied for the anharmonicity of the vibrations and the centrifugal stretching by the method of Pennington and Kobe, and also in the work of Brewer and Brackett [918a] (Φ_T^* for $T \leq 2000^\circ\text{K}$) on the basis of the rigid rotator-harmonic oscillator model. The data of [526] differ from those given in Table 359 (II) by 0.2-0.3 cal/mole·degree, mainly because of the use of the value $B_e = 0.625 \text{ cm}^{-1}$, calculated in the work of Klemperer and Rice (see page). The discrepancy between the data of [4269a] and the values given in Table 359 (II), increases with increase in temperature, attaining 0.15 and 0.23 cal/mole·degree in

the values of Φ_{6000}^* and S_{6000}° . The values of Φ_T^* , calculated in the work [918a], agree with the data of Table 359 (II) within 0.01-0.06 cal/mole·degree.

The differences between the thermodynamic functions of lithium chloride, given in the first and present edition of the Handbook, attain 0.7 cal/mole·degree and are due to the fact that the calculation in the first edition was carried out on the basis of estimated constants of LiCl and the approximate rigid rotator-harmonic oscillator model.

Li₂O and LiOH. The thermodynamic functions of gaseous lithium oxide and hydroxide, given in Tables 351 (II) and 355 (II), were calculated by means of Eqs. (II.243) and (II.244) on the basis of the approximate rigid rotator-harmonic oscillator model, using the above given constants. Table 265 gives the values C_ϕ and C_S in the Eqs. (II.243) and (II.244) and also the characteristic temperatures, found by means of the vibrational constants of Li₂O and LiOH for interpolation on the basis of the harmonic oscillator Tables.

The calculations of the thermodynamic functions of lithium oxide and hydroxide were carried out by means of estimated values of the fundamental frequencies of the molecules Li₂O and LiOH; the structural parameters of LiOH were also chosen on the basis of estimates. The main error in the calculated thermodynamic functions of Li₂O and LiOH is connected with the absence of reliable data on the fundamental frequencies of these molecules. The corresponding errors in the values of Φ_T^* at 298.15; 3000 and 6000°K may attain ± 0.2 ; ± 1.0 and ± 2.0 cal/mole·degree. Nevertheless, the errors due to the inaccuracy of the adopted structural parameters of the two molecules do not exceed 0.1 cal/mole·degree over the entire temperature range. The total error in the calculated values of $\Phi_{298.15}^*$, Φ_{3000}^* and Φ_{6000}^* , taking into account

the errors due to the use of an approximate calculation method, attains ± 0.4 , ± 2 and ± 3 cal/mole·degree.

Following completion of the calculation of the thermodynamic functions of the lithium compounds, the paper of Gordon [1817] was published, in which Tables of the thermodynamic functions of Li_2O are given for $T \leq 6000^\circ\text{K}$. Because indications concerning the calculation method and the constants which had been used, are absent in the work [1817], it is impossible to judge the causes of the marked discrepancies between the data obtained by Gordon and those given in the present Handbook. However, these discrepancies, amounting to 2-3 cal/mole·degree at $T \approx 1000^\circ\text{K}$ and attaining 5.5 cal/mole·degree at 6000°K , considerably exceed the possible magnitude of error in the thermodynamic functions of lithium oxide, given in Table 351 (II). This obviously attests to the fact that the calculation results of Gordon contain gross errors. The results of the calculations of the values Φ_T^* and S_T° of lithium oxide in the work of Berkowitz, Chupka and others [758] ($T = 1300, 1400$ and 1500°K), differ from those given in the Handbook by approximately 2.5 and 3.5 cal/mole·degree, respectively. The discrepancies are due to the incorrect estimation of the fundamental frequencies of Li_2O in the work [758] (see page 1811).

TABLE 265

Constants for the Calculation of the Thermodynamic Functions of Gaseous Li_2O and LiOH

Вещество A	θ_1	θ_2	θ_3	C_D	C_S
	град B			кал/моль·град C	
Li_2O	1122,03	503,48	1193,9	2,2243	10,1730
LiOH	5394,4	1294,7	1582,4	-0,7674	7,1813

A) Substance; B) degree; C) cal/mole·degree

Calculations of the thermodynamic functions of lithium hydroxide are lacking in the literature. The thermodynamic functions of lithium oxide and hydroxide, given in the first and present editions of the Handbook, are identical.

§113. THERMODYNAMIC PROPERTIES OF Li, Li₂O, LiH, LiOH, LiF and LiCl
IN THE SOLID AND LIQUID STATE

The thermodynamic functions of Li, Li₂O, LiH, LiOH, LiF and LiCl in the solid and liquid state (Tables 345, 350, 352, 354, 356 and 358 of Volume II) were calculated by means of Eqs. (III.9)-(III.11) on the basis of the thermodynamic values given in Table 266. The errors in the calculated values of Φ_T^* at the temperatures 298, 1000, 1500, 2000 and 3000°K, estimated on the basis of an analysis of the possible uncertainties in the initial data, are presented in Table 267.

Li. Lithium is known in the form of the cubic face-centered modification (structural type Cu), stable at temperatures under 78°K and in the form of the cubic body-centered modification (structural type Na), which is stable at all other temperatures up to the melting point.

The table of thermodynamic functions for solid and liquid lithium (Table 345 (II)) up to 1500°K was borrowed from the work of Evans, Jacobson, Munson and Wagman [1513]. The authors of the work [1513] examined all works published until the middle of 1955 on the measurement of the heat capacity and enthalpy of lithium and calculated, on the basis of the most reliable data [3735, 3409, 677, 1389] the thermodynamic functions of solid and liquid lithium to 1500°K. The data of Roberts [3451] (1957) on the heat capacity of lithium within the temperature range of 1.5-20°K do not introduce important changes in the values of the thermodynamic functions because the value $S_{15}^{\circ} = 0.015$ cal/g-atom-degree, found in the work [2364] by extrapolation of the

heat capacity of lithium below 15°K, differs only slightly from the value, calculated by means of the data of [3451] ($S_{15}^{\circ} = 0.0185$ cal/g-atom-degree). The slight improvement in the melting point of lithium in the work of Crawford and Montgomery [1211] ($453.35 \pm 0.1^{\circ}\text{K}$) does not alter the calculated values of the thermodynamic functions of molten lithium.

TABLE 226

Adopted Values of the Thermodynamic Magnitudes of Lithium and its compounds in the Solid and Liquid State

Вещество A	Состояние B	$H_{298.15}^{\circ} - H_0^{\circ}$	$S_{298.15}^{\circ}$	$C_{p298.15}^{\circ}$	E Коэффициенты в уравнении для C_p°			Интервал температур ^F	T_m	ΔH_m
		кал/моль C	кал/моль-град D	кал/моль-град	a	b·10 ⁶	c·10 ⁻⁶	°K	°K	кал/моль G
Li	Крист.	1092	6,753	5,91	6	—	—	298,15—453,7	453,7	723
Li	Жидк. H	—	—	—	6	—	—	453,7—1500	—	—
Li	» H	—	—	—	6,82	—	—	1500—2900	—	—
Li ₂ O	Крист.	1732	9,056	12,95	14,939	6,08	3,38	298,15—1700	1700	11700
Li ₂ O	Жидк.	—	—	—	24,0	—	—	1700—5300	—	—
LiH	Крист.	903	4,79	6,89	5,275	9,824	1,345	298,15—961	961	7000
LiH	Жидк.	—	—	—	16,0	—	—	961—2500	—	—
LiOH	Крист.	1772	10,231	11,90	11,988	8,24	2,267	298,15—744,3	744,3	5010
LiOH	Жидк.	—	—	—	20,74	—	—	744,3—3500	—	—
LiF	Крист.	1548	8,53	10,02	—	—	—	298,15—1121,3	1121,3	6470
LiF	Жидк.	—	—	—	15,17	—	—	1121,3—3400	—	—
LiCl	Крист.	2020	12,3	11,51	10,20	5,21	0,22	298,15—883	883	4670
LiCl	Жидк.	—	—	—	16,25	-1,29	—	883—1100	—	—
LiCl	»	—	—	—	14,83	—	—	1100—3200	—	—

A) Substance; B) state; C) cal/mole; D) cal/mole-degree; E) coefficient in the equations for C_p° a); F) temperature range; G) cryst.; H) liq.;

a) $C_p^{\circ} = a + bT - cT^{-2}$ (cal/mole-degree).

b) Table data [1513]

c) $C_p^{\circ} = 13.552 + 3.035 \cdot 10^{-3} \cdot T - 5.308 \cdot 10^{-6} \cdot T^2 + 4.177 \cdot 10^{-9} \cdot T^3 - 1216.6 \cdot T^{-1}$ [1388].

When estimating the accuracy of the thermodynamic functions of lithium it must be taken into account that the accuracy of the measure-

ment of the heat capacity of lithium at low temperatures, carried out by Simon and Swain [3735] is not very high; in particular, the existence of a phase transformation of the "martensite" type [639a] has not been observed* in the work [3735].

The data on the heat capacity of lithium at 298 to 1200°K and the heat of melting of lithium, used in the work [1513] for the calculation of the thermodynamic functions, are highly accurate.

The heats of melting of lithium $\Delta H_{453.7} = 723$ cal/g-atom are also in good agreement with the literature data (717 cal/g-atom [1389], 690 ± 30 cal/g-atom [2439]). The heat capacity of molten lithium, according to [1389] decreases with increase in temperature from $C_{p453.7}^{\circ} = 7.27$ to $C_{p506}^{\circ} = 6.91$ and $C_{p1200}^{\circ} = 6.87$ cal/g-atom-degree. These data are intermediate between the data of Bates and Smith [677] and Redmond and Jones [3409] and, evidently describe best the temperature dependence of the heat capacity of lithium which reflects the decrease in heat capacity within the temperature range of 300-400° above the melting point, which has been established for several metals. At higher temperatures, the decrease in the heat capacity of molten metals is less rapid. Hence, the heat capacity of liquid lithium above 1500°K has been assumed to be constant and amounting to $C_{p1500}^{\circ} = 6.82$ cal/g-atom-degree [1513].

The thermodynamic functions of lithium in the solid and liquid states, calculated for the temperatures 293.15-2900°K, are given in Table 345 (II). The magnitude of the uncertainty in the calculated values Φ_T^* are given in Table 267.

The thermodynamic functions of solid and liquid lithium [Table 345 (II)] differ from those given in the first Handbook edition by approximately 0.1 and 0.4 cal/g-atom degree in the values Φ_{2900}^* and S_{2900}° , mainly because of the improvement in the heat capacity values

of molten lithium.

Li₂O. Lithium oxide is known in the form of the cubic modification (structural type CaF₂). The heat capacity of Li₂O within the range of 16-298°K has been measured by Johnston and Bauer [2269] who calculated the values of $S_{298.15}^{\circ} = 9.056 \pm 0.03$ cal/mole·degree and $H_{298.15}^{\circ} - H_0^{\circ} = 1732 \pm 5$ cal/mole. The entropy obtained by extrapolation of the heat capacity below 16°K is only 0.003 cal/mole·degree.

The enthalpy of crystalline lithium oxide above 298°K has been measured by Shomate and Cohen [3712] (425-1045°K), Rodigina, Gorn'kiy et al. [31] (476-1124°) and Kandyba, Kantor et al. [17] (985-1650°K). The data of the works [3712, 31] agree over the entire temperature range within 0.5% of each other. The equations for the heat capacity of Li₂O, derived in the works [3712, 31] give heat capacity values differing by not more than 1.2% (at 1000°K). At higher temperatures, the only data on the enthalpy of crystalline Li₂O are the measurements, carried out by the authors of the work [17]. Unfortunately, these data cannot be considered to be entirely reliable, because the equation proposed in the work [17] $C_p^{\circ} = 6.82 + 15.53 \cdot 10^{-3}T$ (574 - 1650°K) give improbably high values for the heat capacity of Li₂O* at temperatures over 1000°K. The cause of the high values given in [17] for the enthalpy and heat capacity of Li₂O are probably due to insufficient purity of the investigated sample of Li₂O.** Hence, until more measurements are carried out on the enthalpy of lithium oxide above 1000°K, it is preferable to use the heat capacity equation obtained by Shomate and Cohen (see Table 266) for the calculation of the thermodynamic functions of Li₂O, whose extrapolation to the melting point temperature of Li₂O gives more reasonable heat capacity values than the calculation by means of the equation, given in the work [17].

The data of Rodigina, Gomel'skiy et al. [31] are not inferior to the data of [3712] with regard to accuracy. The discrepancies between the thermodynamic functions, calculated on the basis of these data, are slight (at 1000 and 1700°K, the difference in the value Φ_T^* is less than 0.01 and 0.07 cal/mole·degree, respectively).

The results of different determinations of the melting point of Li_2O are very contradictory: 1848°K [4053] and $1700 \pm 15^\circ\text{K}$ [927 and 17]; the causes of these discrepancies remain unknown. The Handbook adopts the value 1700°K because it has been obtained in the only work [17] on the measurement of the enthalpy of liquid Li_2O . This value is possibly too low because of the presence of moisture in the investigated Li_2O samples.

The heat of melting of lithium oxide $\Delta H_{1700} = 11.7 \pm 2$ kcal/mole has been calculated from the enthalpies of solid and liquid Li_2O at the melting point, calculated on the basis of the adopted values. The heat capacity of molten Li_2O at temperatures to 2000°K is 24.0 cal/mole·degree according to the data of [17] (five enthalpy measurements within the temperature range 1723-1998°K). This heat capacity was also used in the calculation of the thermodynamic functions above 2000°K.

The thermodynamic functions of lithium oxide in the solid and liquid state, calculated for the temperatures 293.15-5300°, are given in Table 350 (II). The magnitudes of the uncertainties in the calculated values of Φ_T^* are given in Table 267.

The differences between the thermodynamic functions of liquid Li_2O given in the present and first editions of the Handbook, amount to 3.5 cal/mole·degree for the entropies and to 2 cal/mole²degree for the value of Φ_{3000}^* . This is due to the considerable improvement in the accuracy of the thermodynamic functions of liquid Li_2O as a result of

the measurement of the enthalpy and heat of melting of lithium oxide in the work of Kandyba, Kantor et al. [17].

LiH. Only the cubic modification of lithium hydride is known (NaCl structure type).

The heat capacity of lithium hydride at low temperatures was measured by Gunther [1890] and Kostyukov [251]. On the basis of the data from the first one of these works, in which 10 measurements of the heat capacity of a contaminated sample of LiH was carried out within the temperature range 74-96°K and at 293°K, Kelley [2364] recommended the figure $S_{298.15}^{\circ} = 5.9 \pm 0.5$ cal/mole·degree. Careful measurements of the heat capacity on a sample of lithium hydride with a purity of 99.8% (as determined by the active hydrogen content) carried out over the range 3.7-295.5°K by Kostyukov, gave a lower value of $S_{298.15}^{\circ} = 4.79 \pm 0.03$ cal/mole·degree and the value $H_{298.15}^{\circ} - H_0^{\circ} = 903 \pm 5$ cal/mole, which have been adopted in the Handbook.

Rodigina, Gomel'skiy, Luginina and Akchurina [33] carried out enthalpy measurements on the same LiH sample within the temperature range 393-664°K with an accuracy of $\pm 0.1\%$. The equation derived by the authors of the work [33] for the heat capacity of LiH (see Table 266) was used* for the calculation of the thermodynamic functions of solid lithium hydride to the melting temperature of LiH, equal, according to the data of Messer et al. [2857], $961 \pm 1^{\circ}\text{K}$.

The heat of melting of lithium hydride, estimated on the basis of thermographic data (heating and cooling curves), is 7 ± 2 kcal/mole [2857]. The heat capacity of liquid lithium hydride was estimated as 16 cal/mole·degree.

The thermodynamic functions of lithium hydride in the solid and liquid state, calculated for temperatures from 293.15-2500°K, are given in Table 352 (II). The errors in the calculated values of Φ_T^* are given

in Table 267.

The differences between the values of the thermodynamic functions of lithium hydride, given in the first and present editions of the Handbook, attain about 1 cal/mole-degree and are due to the more accurate determination of the heat capacity of LiH at lower temperatures [251] and the enthalpy of solid LiH at high temperatures [33, 4557].

LiOH. Lithium hydroxide is known in the form of the tetragonal modification (SnO structure type). There are indications [450] that lithium hydroxide undergoes a phase transformation at 686°K. However, the heat of this transformation is evidently small because it was not observed in the enthalpy measurements on LiOH and was therefore not allowed for in the calculation of the thermodynamic functions of LiOH.

Bauer, Johnston and Kerr [685] measured the heat capacity of lithium hydroxide from 16 to 302°K and calculated the values $S_{298.15}^{\circ} = 10.231 \pm 0.05$ cal/mole-degree and $H_{298.15}^{\circ} - H_0^{\circ} = 1772$ cal/mole. Extrapolation of the heat capacity below 16°K gives a value of only $S_{16}^{\circ} = 0.028$ cal/mole-degree.

The enthalpies of solid and liquid LiOH were measured by Shomate and Cohen [3712] (400-900°K). The equation for the heat capacity of solid LiOH (see Table 266), the melting point of 744.3°K, the heat of melting $\Delta H_{744.3} = 5.01$ kcal/mole and the heat capacity of liquid LiOH $C_p^{\circ} = 20.74$ cal/mole-degree, were adopted on the basis of the data [3712]. The last values was used for the calculation of the thermodynamic functions of liquid LiOH at temperatures below 900°K.

The values of the thermodynamic functions of lithium hydroxide in the solid and liquid state, calculated for the temperatures 293.15-3500°K, are given in Table 354 (II). The estimated errors in the calculated values of Φ_T^* are given in Table 267.

The Tables of the thermodynamic functions of LiOH, given in the

first and present edition of the Handbook, are identical.

LiF. A cubic modification of lithium fluoride is known (NaCl structure type). By the method of differential thermal analysis, phase transformations were detected in LiF at $1035 \pm 5^\circ\text{K}$ [231] and at 1100°K [231, 324]. The heat of these transformations are apparently small; they were not observed during the enthalpy measurements and were not taken into account in the calculation of the thermodynamic functions of LiF.

The heat capacity of LiF at low temperatures has been measured by Strelkov, Kostryukov and Alikhanyants [45] ($18-300^\circ\text{K}$), Clusius, Goldmann and Perlick [1135] ($18-273^\circ\text{K}$), Clusius and Eichenauer [1134] ($10-111^\circ\text{K}$), Jones and Martin [2297, 2785] ($2-30^\circ\text{K}$). There is satisfactory agreement between the data of all these works. The Handbook adopted the values $S_{298.15}^\circ = 8.53 \pm 0.05$ cal/mole·degree and $H_{298.15}^\circ - H_0^\circ = 1548$ cal/mole, found in the work [45]* (extrapolation of the heat capacity below 16.75°K gives a value of $S_{16.75}^\circ = 0.0032$ cal/mole·degree).**

For the heat capacity of solid LiF above 298°K , the values obtained by Douglas and Dever [1388] were used, who carried out 40 enthalpy measurements from 298°K to the melting temperature 1121.3°K ($\pm 1^\circ\text{K}$) and derived a five-term equation for the heat capacity (see Table 260). The research results of Voskresenskay, Sokolov, Banashek and Shmidt [134] who carried out measurements of the true heat capacity ($318-658^\circ\text{K}$, 9 measurements) and enthalpy ($298-1120^\circ\text{K}$, 14 measurements) and who derived the following equation for the heat capacity of LiF:

$$C_p^\circ = 10.32 + 3.90 \cdot 10^{-3} T - 1.36 \cdot 10^{-6} T^2 \quad (318 - 658^\circ\text{K}), \quad (\text{XXVIII.1})$$

$$C_p^\circ = 10.0 + 4.434 \cdot 10^{-3} T - 1.223 \cdot 10^{-6} T^2 \quad (293 - 1121^\circ\text{K}). \quad (\text{XXVIII.2})$$

agree with these data. The differences between the heat capacities cal-

culated by means of these equations and the equation, obtained in the work [1388], do not exceed 2%.

The heat of melting of LiF $\Delta H_{1121.3} = 6470$ cal/mole has been adopted in accordance with the data [1388] (6471 cal/mole) and [134] (6477 cal/mole). The uncertainty in the adopted value does not exceed 10-20 cal/mole).

The values 15.17 [134] (1123-1411°K) and 15.51 cal/mole·degree (1121-1173°K) were obtained for the heat capacity of molten LiF. We adopted the first value because it is based on measurements over a wider temperature range. The difference of 2% between the above given values is evidently due to the fact that the heat capacity of molten inorganic substances in the temperature range close to the melting point decreases slightly with increase in temperature, (see further on heat capacity of molten LiCl).

In the estimate of the uncertainties in the thermodynamic functions calculated by means of the adopted thermodynamic values one must allow for the fact that in spite of certain differences in the heat capacities of solid LiF, obtained in the works [1388, 134] and due mainly to the difference in the methods of processing of the experimental data, the values $H_{1121.3}^{\circ} - H_{298.15}^{\circ}$ practically coincide, while the values S_T° and Φ_T^* differ by less than 0.1 cal/mole·degree. This value can characterize the accuracy of the values Φ_T^* at 1000-1100°K. Up to 1400-1500°K, the possible errors in the values of Φ_T^* increase slowly (up to $\pm 0.15-0.2$) and at higher temperatures, when estimated heat capacities must be used for the calculation, the errors in the thermodynamic functions of LiF increase rapidly and amount to 0.4 and 1.0 cal/mole·degree in the values of Φ_{2000}^* and Φ_{3000}^* .

The thermodynamic functions of lithium fluoride in the solid and liquid state, calculated for temperatures of 293.15-3400°K, are given

in Table 356 (ii). The magnitude of the possible errors in the calculated values of Φ_T^* are given in Table 267.

The tables of the thermodynamic functions of LiF, given in the first and present edition of the Handbook, are identical.

LiCl. The structure of LiCl, like that of LiF, belongs to the structural type NaCl.

Shirley [3708b] carried out careful measurements of the heat capacity of LiCl within the range of 14-320°K and calculated the values $S_{298.15}^\circ = 14.17 \pm 0.02$ cal/mole·degree and $H_{298.15}^\circ - H_0^\circ = 2224$ cal/mole; extrapolation of the heat capacity to below 14°K results in the values $S_{14}^\circ = 0.016$ cal/mole·degree.*

Rodigina, Gomel'skiy and Luginina [32, 349], carried out 39 enthalpy measurements on solid LiCl (367-878°K) and 10 measurements of the enthalpy of liquid LiCl (883-1075°K) and derived an equation for the heat capacity of LiCl (see Table 266) and also determined the heat of melting $\Delta H_{883} = 4.67 \pm 0.1$ kcal/mole at $T_m = 883^\circ\text{K}$.**

The heat capacity of molten LiCl, according to the data [32] decreases with increase in temperature from $C_{p883}^\circ = 15.11$ to $C_{p1100}^\circ = 14.83$ cal/mole·degree. Above 1100°K, the constant value of $C_{p1100}^\circ = 14.83$ cal/mole·degree is used for the heat capacity of molten LiCl.

The thermodynamic functions of lithium chloride in the solid and molten state, calculated for temperatures of 293.15-3200°K, are presented in Table 358 (II). The uncertainties in the calculated values of Φ_T^* are indicated in Table 267.

The thermodynamic functions of LiCl, given in the first edition of the Handbook, were calculated on the basis of approximate estimates of the thermodynamic values and differ from those in the present edition (Table 358 (II)) by 2-3 cal/mole·degree.

TABLE 267

Uncertainties (in cal/mole·degree) of the Values Φ_T^* of Lithium and its Compounds in the Solid and Liquid States

T°K	Li	Li ₂ O	LiH	LiOH	LiF	LiCl
298	±0,2	±0,03	±0,03	±0,05	±0,05	±0,03
1000	±0,3	±0,1	±0,1	±0,1	±0,10	±0,1
1500	±0,4	±0,2	±0,7	±0,5	±0,2	±0,3
2000	±0,6	±0,4	±1,2	±1,0	±0,4	±0,6
3000	±1,0	±1,2	—	±2,0	±1,0	±1,2

§114. THERMOCHEMICAL VALUES

The standard state of lithium is Li (cryst.).

Li (gas). The pressure of saturated lithium vapor has been measured by the effusion method by Lewis [2605] (852-926.5°K), Bogros [858] (732-845°K) and Maucherat [2814] (735-925°K) and also by the method of boiling point determination by Hartmann and Schneider [1967] (1204-1353°K). The vapor pressures of lithium found by Lewis [2605] are much too high (3-4 times) as compared with the data of the other authors and the data of Bogros [858] have a large scatter and an incorrect trend of the pressure-temperature curve for the saturated vapor. Calculation of the heat of sublimation of lithium at 0°K allowing for the formation of Li₂ molecules in the vapor, gives the following values (in kcal/g-atom): 38.00 ± 0.05 [1967], 38.1 ± 0.2 [2814] and 38.5 ± 0.4 [858].

A value has been adopted in the Handbook for the heat of sublimation of lithium which is the average of the two first values:

$$\Delta H_s(\text{Li, cryst.}) = \Delta H_f^0(\text{Li, gas}) = 38,05 \pm 0,1 \text{ kcal/g-atom.}$$

The adopted value coincides with that recommended by Evans et al. [1513] and Stull and Sinke [3894].*

Li⁺(gas) The ionization potential of atomic lithium is 43487.19

cm⁻¹ or

$$I(\text{Li}) = 124,341 \text{ kcal/g-atom,}$$

adopted in correspondence with the recommendation of Moore [2941]. The uncertainty in this value is about 1 cal/g-atom. To the adopted value of the ionization potential corresponds

$$\Delta H_f^\circ(\text{Li}^+, \text{gas}) = 162,391 \pm 0.1 \text{ kcal/g-atom.}$$

Li₂ (gas). On the basis of a graphic extrapolation of the vibrational levels of the excited state B¹Π, in which the Li₂ molecule dissociates into Li(²S) + Li(²P), Loomis and Nusbaum [2647] proposed the value D₀(Li₂) = 1.14 ± 0.05 ev (26.29 ± 0.69 kcal/mole). By means of a more elaborate form of graphic extrapolation, Gaydon [141], based on the measurement results of the same authors, obtained the lower value D₀(Li₂) = 1.12 ± 0.05 ev (25.83 ± 1.15 kcal/mole). Because the potential curves can have small maxima in the excited states, the real dissociation energy of Li₂ may prove to be less than the values corresponding to the convergence limit of the bands. On the basis of these concepts, Gaydon recommended in the later edition of his monograph [1668] D₀(Li₂) = 1.10 ± 0.05 ev (25.34 ± 1.15 kcal/mole).

Lewis [2605] investigated the equilibrium of the reaction



by the molecular beam method and found on the basis of the data thus obtained D₀(Li₂) = 23.4 ± 1.0 kcal/mole. In the monograph by Herzberg [2020] and the Handbook [649] the value 23.75 kcal/mole is recommended with a reference to the work of Lewis [2605]. Evans et al. [1513], based on the measurement results of Lewis [2605] calculated D₀(Li₂) = 25.68 ± 0.15 kcal/mole. However, Evans et al. finally recommended the value D₀(Li₂) = 25.76 ± 0.1 kcal/mole, the average of the value calculated by them on the basis of the data of Lewis and the older value 25.83 ± 1.15 kcal/mole, calculated by Gaydon [141].

Analysis of the results of Lewis' work [2605] shows that the equilibrium of the reaction (XXVIII.3) has been measured by him fairly accurately,* but the vapor pressures of saturated lithium vapor measured in this work and used in the subsequent calculations is wrong (see page 1834). Recalculation of the data of Lewis by means of the vapor pressures of lithium given in the Handbook gives $D_0(\text{Li}_2) = 25.54 \pm \pm 0.12$ kcal/mole.

The round value

$$D_0(\text{Li}_2) = 25,5 \pm 0,2 \text{ kcal/g-atom.}$$

has been adopted in the Handbook. To this value corresponds

$$\Delta H_f^\circ(\text{Li}_2, \text{gas}) = 50,6 \pm 0,25 \text{ kcal/mole.}$$

LiO (gas). Brewer and Margrave [928] based on the results of the studies on the vapor pressure of lithium oxide (see page 1838) found $D_0(\text{LiO}) < 93$ kcal/mole. On the assumption that the energy of dissociation of LiO into ions is equal to the average of the corresponding values for LiF and LiCl, these authors calculated $D_0(\text{LiO}) = 97$ kcal/mole. Recalculation of this value, using the dissociation energy and electron affinity, as adopted in the present Handbook, gives the much lower value $D_0(\text{LiO}) = 73$ kcal/mole.

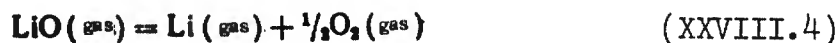
An approximate estimate of the dissociation energy of LiO, based on the ratio

$$\frac{D_0(\text{LiO})}{D_0(\text{Li}_2\text{O})} = \frac{D_0(\text{OH})}{D_0(\text{H}_2\text{O})},$$

gives the value $D_0(\text{LiO}) = 79$ kcal/mole.

Berkowitz, Chupka, Blue and Margrave [758] carried out a mass-spectrometric investigation on the composition of the evaporation products of lithium oxide. Using as a standard the partial pressures of the lithium atoms and oxygen molecules over lithium oxide, calculated on the basis of the thermodynamic data, the authors of the work [758]

found the coefficients required for the determination of the partial pressure of LiO on the basis of the measured LiO^+ ion current. On the basis of the equilibrium constant (1346-1470°K) thus calculated



and the thermodynamic functions of LiO estimated by the authors of the work [758] it was found that the heat of this reaction is $\Delta H_0 = 24.1$ kcal/mole and $D_0(\text{LiO}) = 83$ kcal/mole. In the comments on their work, the authors of [758] used the data of Akishin and Rambidi [64] on the electron diffraction study of the structure of the molecule Li_2O . The more accurate values of the thermodynamic functions calculated on the basis of these data gave $\Delta H_0 = 22.5$ kcal/mole and $D_0(\text{LiO}) = 81.5$ kcal/mole.

Recalculation of the experimental data, obtained by Berkowitz et al. [758], using the thermodynamic properties of lithium and its oxygen compounds* as adopted in the Handbook, led to $\Delta H_0 = 19.7$ kcal/mole and $D_0(\text{LiO}) = 78.7$ kcal/mole. The difference between this and the calculation results of Berkowitz et al. [758] are due mainly to the difference in the thermodynamic functions of LiO (see page 1818).

Berkowitz et al. [758] assumed that the value of $D_0(\text{LiO})$ found by them may be slightly too high owing to the formation of some LiO^+ ions as a result of a dissociation of Li_2O during ionization. The value

$$D_0(\text{LiO}) = 78 \pm 5 \text{ kcal/mole.}$$

To this value corresponds

$$\Delta H_f^\circ(\text{LiO, gas}) = 19,037 \pm 5,0 \text{ kcal/mole.}$$

Li_2O (cryst.). The most reliable method of determining the heat of formation of crystalline lithium oxide is the measurement of the heat of solution of lithium oxide and metallic lithium in water. The heat of solution of lithium in water is discussed in the section on the choice of the heat of formation of crystalline lithium hydroxide

(see page 1842).

The heat of solution of lithium oxide in water has been measured by Beketov [87] and Forcrand [1580]. The insufficient purity of the original lithium oxide samples and the absence of a sufficiently detailed description of the experimental method lowers the value of these works.

A more accurate measurement of the heat of solution of lithium oxide in water was carried out by Kolesov, Skuratov and Zaykin [235, 40]. These authors found $\Delta H_{298.15} = -31.74 \pm 0.04$ kcal/mole (infinite dilution), to which corresponds

$$\Delta H_{\text{sol}}^{\circ}(\text{Li}_2\text{O, cryst.}) = -142,8 \pm 0,2 \text{ kcal/mole.}$$

This value has been adopted in the Handbook.

Li₂O (gas). The vapor pressure of lithium oxide was measured at the MGU under the direction of Ya. I. Gerasimov and Academician N. Nesmeyanov [13]. Nesmeyanov and Belykh [13, 309, 315] used an integral variant of the effusion method (1383-1506°K), while Yevseyev and Pozharskaya [13] determined the weight change of the effusion vessel (1337-1513°K). Comparison of the pressure of saturated lithium oxide vapor found in the work [13] with the value calculated by means of the thermodynamic data on the partial pressure of atomic lithium over lithium oxide shows that under the experimental conditions of [13], about 10% of the lithium oxide in the vapor dissociates into atoms. The heat of sublimation of lithium oxide, calculated with allowance being made for the dissociation on the basis of the data of Academician N. Nesmeyanov and Belykh is 98.1 ± 0.3 kcal/mole and on the basis of the data of Yevseyev and Pozharskaya 98.5 ± 0.7 kcal/mole.

The measurement of the vapor pressure of lithium oxide in the work of Brewer and Margrave [9128] was also carried out by the effusion method (1541-1669°K). The authors of the work [928] found that the

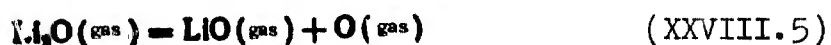
rate of evaporation of lithium oxide, recorded on the basis of the total weight loss of the effusion vessel, is 20-25 times greater than that calculated on the basis of the quantity of lithium, collected from the target. Comparison between the vapor pressure of lithium oxide calculated by means of these data and the partial vapor pressure of atomic lithium over lithium oxide shows that if the calculation is based on the measurement of the total weight loss, the main evaporation product should be lithium oxide with a heat of sublimation of 101.2 kcal/mole; however, if it is based on measurement of the quantity of lithium collected on the target, the main evaporation product should be atomic lithium and oxygen. Brewer and Margrave did not offer any explanation of this contradiction. It should be pointed out that the lithium-oxygen ratio in the condensate, found by these authors, varied from one test to another within wide limits (from 4 to 1) which could not be explained either.

The authors of the work [928] tried to clarify the problem of the principal evaporation product of lithium oxide by special investigations into the volatility of lithium oxide in a stream of argon and oxygen*. A marked decrease in the volatility of lithium oxide in a stream of oxygen was not found. However, the powerful influence of water vapor on the volatility of lithium oxide and the difficulty of preparing really dry gases led to a large scatter in the results and the authors of the work [928] could not arrive at a definite conclusion concerning the nature of the main evaporation product of lithium oxide under neutral conditions.

Berkowitz, Chupka, Blue and Margrave [758] determined the composition and measured the partial pressure of the evaporation products of lithium oxide, using a mass-spectrometric method (see page 1836). The authors of [758] found that the main evaporation products of lithium

oxide are atomic lithium and oxygen and that the concentration of Li_2O molecules in the vapor is about 10%. Based on the relation of the Li_2O ion current and the temperature, Berkowitz et al. found that $\Delta H_{s_{1391}}(\text{Li}_2\text{O}, \text{cryst.}) = 108.3$ kcal/mole and $\Delta H_{s_0} = 115.9$ kcal/mole. In these calculations, the authors of [758] used estimated values of the thermodynamic functions of lithium oxide. The work of Akishin and Rambidi [64] enabled the authors of the work [758] to improve the accuracy of the thermodynamic functions of the lithium compounds which resulted in a slightly higher value for the heat of sublimation $\Delta H_{s_0}(\text{Li}_2\text{O}) = 106.1$ kcal/mole. However, even after this improvement, the thermodynamic functions of Li_2O , calculated in the work [758], contained a considerable error, because incorrect values had been used in the calculation for the fundamental frequencies of the Li_2O molecule (see page 1811). Calculation of the heat of sublimation of Li_2O on the basis of the data of the work [758], using the thermodynamic functions calculated in the present Handbook, leads to $\Delta H_{s_0} = 114.2$ kcal/mole (on the basis of the Li_2O ion current as a function of temperature) and $\Delta H_{s_0} = 109.6 \pm 0.3$ kcal/mole (on the basis of the partial pressure of Li_2O in the vapor). The value calculated by means of the partial pressure of Li_2O is the more accurate.

Brewer and Margrave [928] pointed out that the system lithium-oxygen close to the composition corresponding to Li_2O , has a narrow homogeneity range which can lead to errors in the calculation of the equilibrium heat effects, including the condensed phase Li_2O . The work of Berkowitz et al. [758] gives the values of the vapor pressures of Li_2O , LiO and O at 1400°K . These data enable the heat effect of the reaction



and the corresponding heat of sublimation of lithium oxide $\Delta H_{s_0} = 110$

kcal/mole, which does not depend on the composition of the condensed phase, to be calculated. This value is in good agreement with that calculated from the equilibrium reaction of the evaporation of lithium oxide. Thus, the existence of a homogeneity region evidently does not affect the evaporation of lithium oxide to any marked degree.

The discrepancy between the heat of sublimation of lithium oxide, obtained by the effusion and mass-spectrometric methods, exceeds the usual fortuitous errors of these methods. The mass-spectrometric measurements are the more reliable because the assignment of the measurement results to the Li_2O , Li etc., does not cause any doubt in this case. Moreover, the main qualitative result of the mass-spectrometric investigation [758], the predominance of elemental lithium in the lithium oxide vapor, is undeniable, whereas the results of the effusion measurements can be accounted for only on the assumption that the main component of the vapor is Li_2O .

The heat of sublimation of lithium oxide

$$\Delta H_s(\text{Li}_2\text{O, cryst.}) = 110 \pm 3 \text{ kcal/g-atom,}$$

to which correspond

$$\Delta H_f^\circ(\text{Li}_2\text{O, gas}) = -31,311 \pm 3 \text{ kcal/g-atom,}$$

$$D_0(\text{Li}_2\text{O}) = 166,398 \pm 3 \text{ kcal/mole.}$$

has been adopted in the Handbook.

LiH (cryst.). The heat of solution of crystalline lithium hydride in water, required for the calculation of its heat of formation, has been measured by Guntz [1900], Moers [2932] and Messer, Fasolini and Thalmayer [2859]. Values of -31.96; -31.62 and -31.76 ± 0.10 kcal/mole, respectively, were found in these works (all values were recalculated for infinite dilution [2859]). The most accurate value is that found by Messer et al. [2859]. On the basis of this value and the heat of solution of Li in water, found by the same authors (see further on),

the value

$$\Delta H_f^{\circ}(\text{LiH, cryst.}) = -21,34 \pm 0,4 \text{ kcal/mole.}$$

has been adopted in the Handbook.

LiH (gas). The dissociation energy of LiH was estimated by Gaydon [1668] and Herzberg [2020] on the basis of different extrapolations of the vibrational levels of the states $X^1\Sigma$ and $A^1\Pi$ and was taken as being equal to 57.7 kcal/mole. A similar value has been obtained in the work of Klemperer [2439] by means of an estimate of the equilibrium constant of the dissociation of lithium hydride made on the basis of the intensity of the infrared spectrum of LiH.

Velasko [4076] determined the dissociation energy of LiH on the basis of an analysis of the rotational structure of the bands in the system $B^1\Pi \rightarrow X^1\Sigma$. Because the discontinuity in the rotational structure in different branches of the bands was observed at the same value of J , and the maximum value of J decreases with increasing v' , the author of the work [4076] ascribed the discontinuity to the existence of a predissociation in the rotation of the state $B^1\Pi$ and determined the energy of the dissociation limit of the state $B^1\Pi$ as $34,495 \pm 2 \text{ cm}^{-1}$. Because the dissociation limit of this state of the molecule LiH corresponds to $\text{Li}(2p^2P) + \text{H}(1s^2S)$, the dissociation energy of lithium hydride is $34,495 - 14,904 = 19,591 \text{ cm}^{-1}$, or

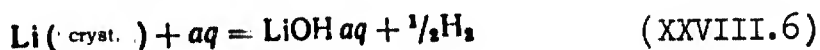
$$D_0(\text{LiH}) = 56,015 \pm 0,05 \text{ kcal/mole.}$$

To the adopted value of the dissociation energy corresponds

$$\Delta H_f^{\circ}(\text{LiH, gas}) = 33,667 \pm 0,1 \text{ kcal/mole.}$$

$$\Delta H_f^{\circ}(\text{LiH, cryst.}) = 53,809 \pm 0,4 \text{ kcal/mole.}$$

LiOH (cryst.). The heat of formation of crystalline lithium hydroxide can be calculated from the cycle into which enter the heats of metallic lithium [3981, 1900, 4391, 2932].



and of lithium hydroxide [1580, 1581, 4033, 4032]



Rossini et al. [3493, 3495, 813, 3508] recalculated the results of these works to standard conditions and obtained the value $\Delta H_{298.15}$ of the reaction (XXVIII.6) = -53.194 kcal/mole and of the reaction (XXVIII.7) = -5.061 kcal/mole (the heats are related to infinite dilution in both cases). To these values corresponds the heat of formation of crystalline lithium hydroxide $\Delta H^{\circ}f_{298.15} = -116.45$ kcal/mole.

Johnston and Bauer [2269] carried out a more accurate recalculation of the measurement results of Moers [2932], Forcrand [1581] and Ueta [4033, 4032] and found for the heat effect of the reaction (XXVIII.6) a value of -53.349 kcal/mole and for the reaction (XXVIII.7) -5.077 kcal/mole (recalculated to 298.15°K on the basis of the heat capacities of the components, taken from Johnston and Bauer and to infinite dilution on the basis of the data [3508]). By means of these data Johnston and Bauer calculated the value $\Delta H^{\circ}f_{298.15}(\text{LiOH, cryst.}) = -116.589 \pm 0.090$ kcal/mole.

Popov, Skuratov, Kolesov and Zaykin [235, 27, 40] carried out a recalculation of the data of Guntz [1900], Zhukovskiy [4391] and Moers [2932] on the heat of solution of lithium in water. By means of the most reliable literature data [3508, 1871] on the heat capacity of the components of the reaction (XXVIII.6), its heat effect was calculated as -53.17 ± 0.2 kcal/mole.

A new determination of the heat of solution of metallic lithium in water was carried out in the work of Messer, Fasolino and Thalmayer [2859] and a heat effect of the reaction (XXVIII.6) of -53.10 ± 0.11 kcal/mole was obtained which is in good agreement with the result of the recalculation [235, 27, 40] of the data of previous works.

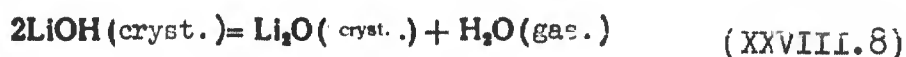
The heat of formation of crystalline lithium hydroxide adopted in

the Handbook

$$\Delta H^\circ_{298.15}(\text{LiOH, cryst.}) = -116.34 \pm 0.3 \text{ kcal/mole.}$$

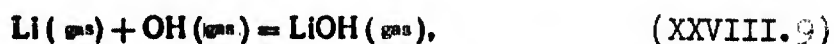
was calculated on the basis of the data [2859] by means of the heat effect of the reaction (XXVIII.6)* and the data of [2269] on the heat effect of the reaction (XXVIII.7).**

The equilibrium



has been investigated in the work [2288, 685, 2269, 1352, 1853]. The most accurate data are evidently those of Gregory and Mohr [1853]. Recalculation of the equilibrium constants found by them, using the thermodynamic functions of the reaction components as adopted in the Handbook, leads to a value for the heat of this reaction of $\Delta H_{298.15} = 32.65$ kcal, which corresponds to a heat of formation of $\Delta H^\circ_{298.15}(\text{LiOH, cryst.}) = -116.6$ kcal/mole. This value agrees within the possible limits of error with that adopted in the Handbook.

LiOH. (gas). The heat of formation of lithium hydroxide in the gaseous state can be calculated on the basis of the measurements of the equilibrium constants of several reactions. Smith and Sugden [3782] found in a study of the ionization, occurring when salts of alkali metals are introduced into a hydrogen-air flame, that the electron concentration in the flame is less than that calculated on the assumption that lithium is present in the flame only in the form of atoms and Li^+ ions. On the assumption that this decrease in the electron concentration and the corresponding concentration of lithium atoms occurs in consequence of the reaction



Smith and Sugden found the equilibrium constant of this reaction and its heat effect, equal to -102 kcal/mole. Recalculation of the data obtained in the work [3782] using the thermodynamic values adopted in

the Handbook gives a heat of the reaction (XXVIII.9) of $\Delta H_0 = -98$ kcal/mole and $\Delta H^\circ f_0(\text{LiOH, gas}) = -51$ kcal/mole. The accuracy of determination of this value is reduced by the possibility of the occurrence of side reactions which reduce the quantity of lithium atoms in the combustion products.

Berkowitz, Meschi and Chupka [758a] carried out a mass-spectrometric study of the reaction



The lithium oxide was heated (1100-1400°K) in an effusion vessel into which water vapor was introduced (pressure 0.04 and 0.16 mm Hg). The molecules LiOH , $(\text{LiOH})_2$ and $(\text{LiOH})_3$ were observed to be present in the vapor. The main component of the vapor was LiOH . The partial pressure of $(\text{LiOH})_2$ was about 40 times and that of $(\text{LiOH})_3$ about 1000 times less than the partial pressure of LiOH . Its heat effect was found by means of the relation between the equilibrium constant of the reaction (XXVIII.10) and temperature as $\Delta H_{1300} = 78 \pm 3$ kcal/mole, or $\Delta H_0 = 83.7$ kcal/mole, to which corresponds $\Delta H^\circ f_0(\text{LiOH, gas}) = -57.4 \pm \pm 1.5$ kcal/mole.

Using the thermodynamic functions of the reaction components calculated by them and the measured equilibrium constants of the reaction (XXVIII.10), Berkowitz et al. found the heat effect of this reaction $\Delta H_{1300} = 80 \pm 2$ kcal/mole or $\Delta H_0 = 85.7$ kcal/mole which gives $\Delta H^\circ f_0(\text{LiOH, gas}) = -56.3 \pm 1.0$ kcal/mole. Calculation, using the thermodynamic functions adopted in the Handbook, gave $\Delta H_0 = 87.3$ kcal/mole and $\Delta H^\circ f_0(\text{LiOH, gas}) = -55.6$ kcal/mole.

The value

$$\Delta H^\circ f_0(\text{LiOH, gas}) = -56.5 \pm 2 \text{ kcal/mole,}$$

which is the average of two values calculated on the basis of the measurement results of Berkowitz, Meschi and Chupka [758a] has been

opted in the Handbook. To this value corresponds

$$D_s(\text{LiOH}) = 205,169 \pm 2 \text{ kcal/mole,}$$
$$\Delta H_s(\text{LiOH, cryst.}) = 58,473 \pm 2 \text{ kcal/mole.}$$

LiF (cryst.). The heat of neutralization of a lithium hydroxide solution with hydrofluoric acid has been measured by Petersen [3227] who found $\Delta H_{292} = -16.4$ kcal/mole (final concentration $\text{LiF} \cdot 400\text{H}_2\text{O}$). The relatively high final concentration of the solution caused a precipitation of lithium fluoride, which disturbed the measurement during the final period of the calorimetric experiment and the determination of the correction for the heat exchange. Moreover, the purity of the original solutions, used by Petersen, is open to doubt.

Popov, Skuratov and Kolesov [27, 234a] carried out a more accurate determination of the heat of neutralization of hydrofluoric acid by lithium hydroxide solution (slight excess). After application of a correction for the impurities in the original solution of lithium hydroxide (0.5% NaOH), the authors of the work [27] found $\Delta H_{294.6} = -16.45 \pm 0.03$ kcal/mole (final concentration of the solution $\text{LiF} \cdot 3800 \text{H}_2\text{O}$).

The authors of the works [27, 234a] also measured the heat of solution of crystalline lithium fluoride in water by adiabatic and diathermic methods. Both methods gave the same value $\Delta H_{294.6} = 1.25 \pm 0.02$ kcal/mole (dilution $\text{LiF} \cdot 3800 \text{H}_2\text{O}$). The heat of solution of lithium fluoride had been measured previously by Forcrand [1582] and by Kapustinskiy and Stakhanova [212]. In the work of Forcrand, the value $\Delta H = 1.04$ kcal/mole; however, several important details of the experiment remained unclear. Kapustinskiy and Stakhanova obtained the value $\Delta H_{298.15} = 1.33$ kcal/mole (infinite dilution). Recalculation to the dilution $\text{LiF} \cdot 3800 \text{H}_2\text{O}$ gives a value which is identical with those of the works [27, 234a] within the limits of experimental error. On the basis

of the heat of solution and neutralization from the works of Popov, Skuratov and Kolesov [27, 234a] (the correction for bringing it up to 25°C is relatively small and can be neglected [27]) the Handbook adopted

$$\Delta H^{\circ}_{298,15} (\text{LiF, } \text{crist.}) = -146,4 \pm 0,5 \text{ kcal/mole.}$$

The magnitude of the supposed error is determined primarily by the inaccuracy of the heat of formation of hydrogen fluoride.

LiF (gas). Table 268 gives the results of the calculation of the heat of sublimation of lithium fluoride on the basis of measurements of its saturated vapor. When carrying out such calculations, it is essential to allow for the complex composition of the saturated vapor. In spite of the fact that the association of the molecules of the halogen compounds of the alkali metals in the vapor has been investigated in numerous works (see [3469, 198, 3108, 2502, 3124, 2087, 2914, 1472, 62]), this problem cannot be considered to have been solved as yet.

Eisenstadt, Rothberg and Kusch [1472] investigated the velocity distribution of the molecules in a molecular beam of lithium fluoride and found that at 1127°K the vapor contains 56.2% LiF, 37.4% Li₂F₂ and 6.4% Li₃F₃. Akishin, Gorokhov and Sidorov [62, 63] carried out a mass-spectrometric investigation on LiF vapor using a double effusion chamber and found that at 1098°K the vapor consists of 48.8% LiF, 35.6% Li₂F₂ and 15.6% Li₃F₃ (the calculations were carried out by the method proposed by Milne [2921]. Based on the relation of the LiF⁺ and Li₂F₂⁺ ion currents and temperature, Akishin, Gorokhov and Sidorov calculated the heat of sublimation of lithium fluoride in the form of the monomeric molecules $\Delta H_{s0} = 65.3$ kcal/mole and the dissociation energy of the dimer into monomer $D_0 = 58.7 \pm 2.7$ kcal/mole.

TABLE 268

Results of the Calculations of the Heat of Sublimation of LiF

Авторы A	Год B	Метод C	Интервал температур, °K D	Число измерений E	$\Delta H_{20}(\text{LiF})^a$ ккал/моль F	$\frac{P_{\text{LiF}} \cdot 100\%}{P_{\text{LiF}} + P_{\text{Li}_2\text{F}_2}}$	$\Delta H_{20}(\text{LiF})^b$ ккал/моль F
Варгелс:рг, Шульц [4170] G	1921	Кипения	1626—1820	5	63,6±0,2	75—80	64,7
Руфф и др. [3562] H	1922	"	1671—1939	14	63,1±0,2	75—80	64,2
Сенс, Стюи [3687] I	1958	Протока	1124—1333	—	63,1	60—70	65,3
Пью, Барроу [3344] K	1958	Эффузионный ^a	1001—1126	68	64,1±0,3	57—63	65,4
Эйзенштадт, Ротберг Куш [1472] L	1958	Эффузионный	1072—1158	6	66,7±0,3	59—63 ^r	67,8
Евсеев и др. [182] M	1959	"	926—1053	12	61,0±0,2	40—43	63,4
Акишин, Горохов, Сидоров [62, 63] N	1959	Масс-спектрометрический	1096	—	—	58	65,3

- A) Authors; B) year; C) method; D) temperature interval, °K;
 E) number of measurements; F) kcal/mole
 a) On the assumption of an absence of association in lithium fluoride vapor
 b) with allowance being made for the complex composition of lithium fluoride vapor
 c) variant of the method - torsion method
 d) found by Eisenstadt, Rothberg and Kusch [1472].

In order to allow for the association of lithium fluoride vapor over a broad range of temperatures, the authors of the present Handbook calculated the equilibrium constants of the dissociation of the dimer into the monomer. The thermodynamic function of Li_2F_2 (and also of Li_2Cl_2 and Na_2Cl_2) were calculated for temperatures of 800-2000°K on the basis of the approximate model of the harmonic oscillator-rigid rotator by means of the molecular constants, given in Table 269. A flat rhombic configuration and the interatomic distances given in Table 269 were used for the dimeric molecules in correspondence with the data of Akishin and Rambidi [65]. The vibration frequencies of the dimers were estimated, according to Milne and Cubicciotti [2922] in the following manner: the four valency vibrations were assumed to be

equal to the vibration frequency of the monomer molecule (rounded off) and for the two deformation frequencies, values were used which were three times smaller.* The calculated values of Φ_T^* and the enthalpies of the three dimerized gases are given in Table 270.

The most reliable measurements of the pressure of saturated lithium fluoride vapor were carried out by Sense and Stone [3687] and Pugh and Farrow [3344]. On the basis of these measurements and the thermodynamic functions of Li_2F_2 , on the assumption that the partial pressure of Li_3F_3 under the experimental conditions is 15% of the total pressure [62, 63] the authors of the Handbook calculated the dissociation energy



as 61 kcal/mole at 0°K. The values 60.3 ; 58.7 ± 2.7 and 63.8 ± 2.3 kcal/mole, calculated by means of the second law of thermodynamics in the works [1472, 62 and 2650] respectively, and calculated to 0°K, agree within the limits of error with this value. Milne and Cubicciotti [2922] obtained 54.2 kcal/mole as a result of an estimate of this value, using the ionic model [3202].

TABLE 269

Molecular Constants of Li_2F_2 , Li_2Cl_2 and Na_2Cl_2 Adopted for the Calculation of the Thermodynamic Functions

Вещество A	$\omega_1(4)$	$\omega_2(2)$	r_{M-X}	r_{X-X}	$I_A I_B I_C$	σ
	cm^{-1}		\AA		$10^{-21} (\text{g}\cdot\text{cm}^2)^2$	
Li_2F_2	900	300	1,68	2,67	367,7	4
Li_2Cl_2	660	220	2,17	3,68	5216	4
Na_2Cl_2	365	120	2,45	4,04	44210	4

A) substance

TABLE 270

Thermodynamic Functions of Gaseous Li_2F_2 , Li_2Cl_2 and Na_2Cl_2 (Φ_T^* in cal/mole·degree, $H_T^\circ - H_0^\circ$ in cal/mole)

T, °K	Li_2F_2		Li_2Cl_2		Na_2Cl_2	
	Φ_T^*	$H_T^\circ - H_0^\circ$	Φ_T^*	$H_T^\circ - H_0^\circ$	Φ°	$H_T^\circ - H_0^\circ$
800	62,157	11302	69,163	12266	77,333	13707
1000	68,208	15011	72,670	16092	81,214	17634
1200	69,007	18802	75,655	19964	84,461	21577
1400	71,461	22644	78,252	23864	87,253	25525
1600	73,648	26517	80,550	27787	89,701	29485
1800	75,620	30411	82,611	31714	91,881	33446
2000	77,413	34324	84,478	35656	93,845	37412

- a) The thermodynamic functions of Li_2Cl_2 were calculated in the work [684a]. The values thus obtained agree with those given in Table 270 within the limits of 1 cal/mole·degree.

The composition of the evaporation products of lithium fluoride, calculated on the assumption that the dissociation energy of the dimer is 61 kcal/mole and that the trimer concentration at 900-1300°K is 15% [62, 63] and at 1600-1900°K is about 5%, is given in the penultimate column of Table 268. It can be seen from these data that the proportion of the dimer in lithium fluoride vapor decreases with increase in temperature by 60-40% (over solid LiF) to 20% (over liquid LiF near the boiling point). The last column of Table 268 gives the heats of sublimation of the monomeric LiF molecules, calculated under the assumption of a complex composition of the vapor.

The heat of sublimation of lithium fluoride, calculated on the basis of the data obtained in the works [3344, 3687, 3562, 4170, 62] are in satisfactory agreement and give a mean value of $\Delta H_{s,0} = 65$ kcal/mole.

The composition of lithium fluoride vapor and not its saturation

pressure was primarily studied in the work of Eisenstadt, Rothberg and Kusch [1472]. The vapor pressures obtained in this work are evidently not accurate and the heat of sublimation $\Delta H_{s_0} = 67.8$ kcal/mole calculated by means of them is too high as for several other halogen compounds of the alkali metals (see NaF, KF). The reasons for the fact that the vapor pressure values for lithium fluoride, obtained by the differential and integral variants of the effusion method by Yevseyev et al. [182] are much greater than those found by other authors, are unclear.

The value

$$\Delta H_{s_0}(\text{LiF, cryst}) = 65 \pm 1 \text{ kcal/mole,}$$

to which corresponds

$$\begin{aligned} \Delta H^{\circ}_f(\text{LiF, gas}) &= -80,801 \pm 1.1 \text{ kcal/mole,} \\ D_0(\text{LiF}) &= 137,351 \pm 1.2 \text{ kcal/g-atom.} \end{aligned}$$

has been adopted in the Handbook for the heat of sublimation of LiF.

Page and Sugden [3164] made attempts to determine the dissociation energy of the molecule LiF on the basis of a study on the influence of fluorine addition on the absorption of centimeter waves in lithium-containing flames. Under these conditions, the decrease in the damping of the centimeter waves is due to the formation of LiF molecules and also of F^- ions in the flame. Page and Sugden found the value $D_0(\text{LiF}) = 141$ kcal/mole, but this value is less accurate than that calculated on the basis of the heat of formation and the heat of sublimation of lithium fluoride.*

LiCl (cryst.). An analysis and recalculation of numerous data on the heat of neutralization of lithium hydroxide solution with hydrochloric acid and on the heat of solution of lithium chloride has been carried out by Bykhovskiy, Rossini et al. [3508, 813, 3493, 3494].

The heat of formation of crystalline lithium chloride, -97.9

kcal/mole, was calculated on the basis of these data. Recalculation of this value, allowing for the heat of solution of lithium in water as adopted in the Handbook (see page 1844), gives the value -97.6 kcal/mole.

Ueda [4033, 4032] investigated the e.d.s. of a lithium chloride element and determined the free energy of formation of lithium chloride $\Delta G_{298.1} = -91,612$ cal/mole. By means of the entropies adopted in the present Handbook for lithium chloride, metallic lithium and elemental chlorine it was found that this value corresponds to a heat of formation $\Delta H^{\circ}f_{298.15}(\text{LiCl, cryst.}) = -97.9$ kcal/mole.

H. Siemonsen and U. Siemonsen [3721] found the heat of formation of lithium chloride $\Delta H^{\circ}f_{298.15} = -94.8 \pm 0.6$ kcal/mole from the combustion of metallic lithium in chlorine. The authors of the work [3721] believe that the relationship between the heats of formation of NaCl and LiCl and the ionic radii of the corresponding metals confirm the correctness of the value found by them.

The reasons for the discrepancy between the values, found by different methods, are unclear. The Handbook adopted

$$\Delta H^{\circ}f_{298.15}(\text{LiCl, cryst.}) = -97.6 \pm 0.5 \text{ kcal/g-atom,}$$

because this value has been obtained by the most accurate method of determination of the heats of solution and because it is confirmed by the e.d.s. data.

LiCl (gas). The results of the calculations of the heat of sublimation of lithium chloride on the basis of measurements of the pressure of its saturated vapor are given in Table 271. Saturated lithium chloride vapor, like lithium fluoride vapor, contains associated molecules.

Miller and Kush [2914] investigated the velocity distribution of the molecules in a molecular beam of lithium chloride and found that at

870°K the vapor contains 20.4% LiCl, 74.3% Li₂Cl₂ and 5.3% Li₃Cl₃. Akishin, Gorokhov and Khodeyev [3] carried out a mass-spectrometric study of the composition of lithium chloride vapor and confirmed the high concentration of the dimer Li₂Cl₂ and the trimer Li₃Cl₃ in lithium chloride vapor. In this work the authors used a conventional effusion chamber but after [62] they showed that more accurate results can be obtained by means of a double effusion chamber. Hence, the results of the work [3] must be considered as preliminary.

TABLE 271

Results of the Calculation of the Heat of Sublimation of LiCl

	А	В	С	Д	Е	Г	Ж	З
	Авторы	Год	Метод	Интервал температур, °К	Число измерений	$\Delta H_{so}(\text{LiCl})^a$, ккал/моль	$\frac{P_{\text{LiCl}} \cdot 100\%}{P_{\text{LiCl}} + P_{\text{Li}_2\text{Cl}_2}}$	$\Delta H_{so}(\text{LiCl})^b$, ккал/моль
Г	Нива [3097]	1938	Эффузионный	783—823	5	48,3±0,1	14	52,0
Н	Несмеянов, Сазонов [314]	1958	О	808—889	19	49,3±0,1	19	52,6
И	Миллер, Куш [2914] . .	1956	П	878—911	3	49,9±0,1	28	52,3
К	Кангро, Викинг [2321] .	1938	Протока	1133—1263	3	48,3±0,3	40	51,7
Л	Руфф, Мугдан [3562] . .	1921	Кипения	1318—1598	7	48,6±0,2	55	50,5
М	Вартенберг, Шульц [4170]	1921	Q	1442—1657	12	49,7±0,2	65	51,1
Н	Акишин и др. [3] . . .	1958	Масс-спектрометрический	816—863	—	—	—	52,2

- A) authors, B) year; C) method; D) temperature range, °K; E) number of measurements; F) kcal/mole; G) Niwa; H) Nesmeyanov, Sazonov; I) Miller, Kush; K) Kangro and Wicking; L) Ruff, Mugdan; M) Wartenberg, Schulz; N) Akishin et al.; O) effusion; P) channel; Q) Boiling; R) mass-spectrometric.
- a) The data of Maier [2744] (1154-1525°K), obtained by the static method, differ markedly from all other data and are thus not considered here.
- b) on the assumption of an absence of association in lithium chloride vapor.
- c) taking into account the complex composition of lithium chloride vapor.

The dissociation energy $D_0 = 53$ kcal/mole and the equilibrium constants of the dissociation of the dimer Li₂Cl₂ into monomer were calculated on the basis of the ratio of the components of saturated lithium chloride vapor, found by Miller and Kush [2914], the pressure

of the saturated vapor at 870°K on the basis of the data [314] and the thermodynamic functions of Li_2Cl_2 from Table 270. The concentration of the monomeric molecules of lithium chloride in the saturated vapor, given in Table 271, were calculated on the basis of these equilibrium constants under the assumption that the concentration of the trimer in the vapor is 5% and is independent of temperature. The last column of Table 271 gives the heats of sublimation of lithium chloride in the form of monomeric molecules. The values calculated on the basis of the results of the effusion measurements, the measurements by the channel method and the boiling method, are in good agreement.

The heat of sublimation of lithium chloride which is the average of the calculation results on the basis of the data [3097, 314, 2914, 2321, 3562, 4170]; has been adopted for the Handbook:

$$\Delta H_s(\text{LiCl, cryst.}) = 51.7 \pm 1.0 \text{ kcal/mole,}$$

to this value correspond

$$\begin{aligned} \Delta H^{\circ} f_0(\text{LiCl, gas}) &= -45,935 \pm 1.1 \text{ kcal/mole,} \\ D_0(\text{LiCl}) &= 112,535 \pm 1.1 \text{ kcal/mole,} \end{aligned}$$

Gurvich and Veyts [170] investigated the dissociation equilibrium of LiCl in $\text{H}_2 + \text{Cl}_2$ flames by the method of measuring the intensity of the resonance line of the lithium atom. The value found by them $D_0(\text{LiCl}) = 118 \pm 4 \text{ kcal/mole}$ corresponds to a heat of sublimation of lithium chloride of 46 kcal/mole. The discrepancy between this value and that calculated on the basis of the results of the vapor pressure measurements of Lithium chloride, somewhat exceeds the possible error due to the inaccuracy of the thermodynamic functions or the association of the LiCl molecules in the flame ($\pm 2.0 \text{ kcal/mole}$).*

TABLE 272

Values adopted (cal/mole) for thermochemical quantities of lithium and its compounds

Вещество A	Состояние B	D_0, I_0 или ΔH_{s_0} C	ΔH°_0	$\Delta H^\circ_{293.15}$	$\Delta H^\circ_{298.15}$	$H^\circ_{293.15} - H^\circ_0$	$H^\circ_{298.15} - H^\circ_0$
Li	Крист. D	38 050 ^a	0	0	0	1083	1092
Li	Газ E	—	38 050	38 443	38 439	1456	1481
Li ⁺	• E	124 341 ^b	162 391	164 240	164 261	1456	1481
Li ₂	•	25 590	50 600	50 745	50 730	2271	2314
LiO	•	78 000	19 037	19 039	19 031	2085	2123
Li ₂ O	Крист. D	119 000 ^a	-141 311	-142 789	-142 800	1668	1732
Li ₂ O	Газ E	166 398	-31 311	-31 817	-31 839	2640	2694
LiH	Крист. D	53 809 ^a	-20 142	-21 327	-21 340	870	903
LiH	Газ E	56 015	33 667	33 652	33 641	2040	2075
LiOH	Крист. D	58 473 ^a	-114 973	-116 335	-116 340	1713	1772
LiOH	Газ E	205 169	-56 500	-57 199	-57 219	2376	2420
LiF	Крист. D	65 000 ^a	-145 801	-146 403	-146 400	1498	1548
LiF	Газ E	137 351	-80 801	-80 828	-80 838	2073	2111
LiCl	Крист. D	51 700 ^a	-97 635	-97 608	-97 600	2167	2224
LiCl	Газ E	112 535	-45 935	-45 957	-45 967	2118	2157

^aHeat of sublimation. ^bIonization potential of Li atom.

A) Substance; B) state; C) or D) cryst. E) gas.

1794 Below we give the vibrational constants of Li_2 in the state $X^1\Sigma$ on the basis of the data from several authors:

	[4346]	[1979]	[2647]	[2712]	[834]	[3020]
ω_e	359.3	349.9	351.00	351.374	351.300	351.435
$\omega_e x_e$	2.4	2.5	2.500	2.5	2.503	2.502
$\omega_e y_e$	-	-	0.0007	---	-0.0003	-0.0008

1795 The analysis of the rotational structure in the work [524] was carried out on the 10-12 first lines of each branch.

1797 The state $A^1\Sigma$ of the molecule LiH , like that of the molecules of the other alkali metals, is of an anomalous nature because of the influence of the ionic states of these molecules. As a result, $\Delta G_{v+1/2}$ and B_v pass through maxima with increase in v . A theoretical analysis of this phenomenon has been given by Mulliken [2993, 2994].

1799 The work of Moran and Trischka [2945a] was published in 1961, in which these measurements had been repeated by means of an improved apparatus. $\omega_e = 930 \pm 19 \text{ cm}^{-1}$ and the ratio of the vibration frequencies of Li^6F and Li^7F of 1.06 ± 0.01 were found in the work [2945a].

1800 An estimate of r_{LiF} has also been made in the work of Huff, Gordon and Morrell [2142] but the value of $r_{\text{LiF}} = 1.35 \text{ \AA}$ adopted by them can be excluded as being totally unreasonable.

1802 In the works [52, 250] the interatomic distances in the molecules of the alkali metal halogenides were chosen on the basis of the data [2116] obtained in studies of the microwave spectra.

1806 Klemperer and Rice [2444] calculated the anharmonicity constant $\omega_e x_e$ required for the calculation of the vibration frequencies of LiBr and LiI from the relation $\omega_e x_e = [\omega_e^2 / \nu + \nu] / \nu^2$. The authors of the work [2444] believe that the uncertainty in the frequencies determined by them do not exceed several cm^{-1} .

1808 Following the completion of the calculations of the thermodynamic functions of LiCl , the authors of the Handbook learned of the new work of Klemperer and coworkers [2443b], in which the absorption spectra of LiCl , LiBr and LiI had been obtained on an apparatus with large dispersion. The wave numbers of the band edges 1-0, 2-1 and 3-2 are given in the work [2443b] for LiCl and it is pointed out that in the band

1-0 the displacement of the edge from its head, as for all halogen compounds of lithium, is about 50 cm^{-1} . The vibrational constants of LiCl, recommended in the work [2443b] are $\omega_e = 641.1 \pm 3$ and $\omega_e x_e = 4.2 \pm 0.3 \text{ cm}^{-1}$. The authors of this work do not explain the reasons for the marked change in the value of ω_e as compared with that found in the previous work by Klemperer and Rice [2444]; analogous changes, which are somewhat smaller but exceed the possible error, indicated in the preceding works, refer to the vibrational constants of LiBr and LiI.

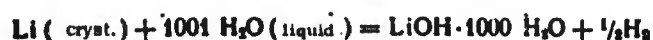
- 1808 Comparison of the interatomic distances in the molecules XY_2 and XY shows that in the divalent elements X the bond length in the molecule XY is normally a few hundredth A shorter than in the molecule XY_2 .
- 1813 The constants A_ϕ and A_S include the value $R \ln 2$ for taking into account the symmetry of the molecule Li_2 .
- 1817 The second anharmonicity coefficient in the equation for the energy of the vibrational levels of LiH in the state $X^1\Sigma$ has not been allowed for in the calculation. The two-term equation should evidently approximate the energy of the higher vibrational levels of this state more closely because the energy levels, calculated by means of this equation, converge near the dissociation limit of LiH.
- 1826 Following the completion of the calculation of the thermodynamic functions of lithium, the work of Martin [2786] on the measurement of the heat capacity of a lithium sample with an atomic weight of 6.945 within the temperature range 20-300°K was published. The "martensite" transformation of lithium (78°K [639a]) discovered earlier is apparent in the heat capacity curve at 100-120°K; the heat of transformation has been found to be 14 cal/g-atom. Martin calculated for lithium $S_{298.15}^\circ = 6.95 \pm 0.04 \text{ cal/g-atom}\cdot\text{degree}$; calculation of $H_{298.15}^\circ - H_0^\circ$ by means of Martin's data gives 1105 cal/g-atom.
- 1827 Thus, for example, at the melting point of Li_2O , the value $C_{p1700}^\circ = 33.22 \text{ cal/mole}\cdot\text{degree}$, calculated by means of the above equation, considerably exceeds the value of 21.0 cal/mole·degree calculated by means of the equations, proposed in the works [31, 3712], respectively.
- 1827 The authors of the work [17] assumed the presence of lithium hydroxide as an impurity in the Li_2O sample, which has a much lower melting point than Li_2O .
- 1829 In good agreement with the data of Rodigina et al [33] are the results of the enthalpy measurements on lithium hydride

carried out over the range 413-840°K by Lang [2557], who recommended for the heat capacity of LiH the linear equation $C_p = 3.82 \pm 11.42 \cdot 10^{-3} T$.

- 1831 Calculation of $S_{298.15}^\circ$ for LiF on the basis of the data [1135] gives values (in cal/mole·degree) of 8.37 [3508], 8.53 [134], 8.52 [1388], and 8.50 ± 0.05 [2494]. The recommendation of Kelley [2364] $S_{298.15}^\circ = 8.32 \pm 0.08$ cal/mole·degree is erroneous because it is based on the data of the preliminary report of Clusius [1132] on the heat capacity of LiF at constant volume.
- 1831 On the basis of the data [2785] at $T < 20^\circ\text{K}$ the characteristic Debye temperature of LiF is $737 \pm 9^\circ\text{K}$, giving $S_{16.75}^\circ = 0.0036$ cal/mole·degree.
- 1833 The measurements of the mean heat capacity of LiCl within the range 85-270°K carried out earlier by Slonin and Haetig [3766] are inaccurate and give a low entropy ($S_{298.15}^\circ = 12.3$ cal/mole·degree). Kelley [2364] calculated $S_{298.15}^\circ = 13.9 \pm 0.5$ cal/mole·degree on the basis of the heat of formation and isobaric potential of LiCl.
- 1833 Dworkin and Bredig [1431] obtained for the heat of melting of LiCl the similar value $\Delta H_m = 4.76 \pm 0.1$ kcal/mole. The value $\Delta H_m = 3.85 \pm 0.15$ kcal/mole, determined by Blanc [838a] by the cryometric method, is erroneous.
- 1834 The more precise determination of the heat capacity of lithium at low temperatures in the work [2786] (see page 1825) published after the completion of the work on this chapter, gives slightly different heats of sublimation for lithium, namely (in kcal/g-atom): 37.8 [1967], 38.0 [2814], and 38.3 [858].
- 1836 The data of Lewis are confirmed by the works of Meissner and Scheffers [2846, 2847].
- 1837 Calculation of the partial pressures of lithium and oxygen over Li_2O , based on the thermodynamic properties of the lithium compounds, adopted in the Handbook, gives values which are 7% smaller than those calculated in the work [758]. This difference was taken into account in the calculations because the partial pressures of lithium and oxygen were used in the work [758] for the calculation of the pressure of LiO .
- 1839 The decrease in the volatility of lithium oxide in presence of oxygen would mean that the evaporation takes place mainly in the form of lithium atoms.
- 1842 In the work of Gunn and Green [1887] published after completion of the work on the materials of the present Chapter, the heat of solution of lithium in water (see page 843) and of lithium hydride in water were measured again (-31.597 ± 0.018 kcal/mole, recalculated to infinite dilution). To

the heats of these reactions found by the authors of the work [1887] corresponds a heat of formation of lithium hydride $\Delta H^{\circ}_f_{298.15} = -21.666 \pm 0.026$ kcal/mole, which exceeds considerably that adopted in the Handbook.

1844 Following completion of the work on the materials of the present chapter, the work of Gunn and Green [1887] was published, in which another determination of the heat of solution of metallic lithium in water has been carried out. As a result of very careful measurements, these authors found for the heat of the reaction



the value -53.142 ± 0.019 kcal/mole. To this value corresponds a heat effect of the reaction (XXVIII.6) of -53.263 ± 0.019 kcal/mole (the heats of dilution from the work [3508] were used in the recalculations). The difference between this value and that adopted in the Handbook is within the limits of possible error.

1844 In the work of Reshetnikov [344b] published after the completion of the work on the materials of the present chapter the results of measurements of the heat of solution of LiOH are given. According to his data, the heat of solution of LiOH is $\Delta H_{298.15} = -5.47 \pm 0.007$ kcal/mole (concentration LiOH $\cdot 400 \text{ H}_2\text{O}$). Recalculation of this value to infinite dilution, based on the data from the Handbook [3508] gave $\Delta H_{298.15} = -5.622$ kcal/mole. On the basis of these and other data of Kolesov and Skuratov [18] it is believed that the heat effect of the reaction (28.7) is $-5.0 \pm$ kcal/mole. This value agrees within the limits of error with that adopted in the present Handbook (-5.077 kcal/mole).

1849 Following the completion of the corresponding calculations by the authors of the Handbook, the works [684a, 756a] became known, in which an estimate of the constants of the molecules of several dimers had been made and also the work [2443a] in which two bands had been detected in the spectra of Li_2X_2 which are ascribed to the vibrations B_{2n} and B_{3n} . The fundamental frequencies, obtained in these works, differ considerably from each other and also from those adopted by the authors of the Handbook.

1851 In the work [1010a], which appeared after the present chapter had been written, the value $D_0(\text{LiF}) = 136 \pm 8$ kcal/mole, which is in good agreement with that adopted in the Handbook, was obtained on the basis of flame photometric measurements.

1854 The value $D_0(\text{LiCl}) = 110.5 \pm 3$ kcal/mole which is closer to that adopted in the Handbook, was found as a result of flame photometric measurements in the work [1010a].

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script
Page
No.

[Transliterated Symbols]

- 1803 $v_{\text{конт}} = v_{\text{kant}} = v_{\text{edge}}$
- 1852 э.д.с. = e.d.s. = elektrodizhushchaya sila = electromotive
force (emf).
- 1838 МГУ = MGU = moskovskiy gosudarstvenny universitet = Moscow
State University

Chapter 29

ALKALI METALS AND COMPOUNDS OF SODIUM AND

POTASSIUM WITH FLUORINE AND CHLORINE

(Na, Na⁺, NaF, NaCl, K, K⁺, KF, KCl, Rb, Rb⁺, Cs, Cs⁺)

The thermodynamic properties of the alkali metals (with the exception of lithium and its compounds, which were treated in a separate chapter (and the fluoride of sodium and potassium will be dealt with in the present chapter of the Handbook. In the condensed state the alkali metals form oxides of several types. The normal oxides of sodium, potassium, rubidium and cesium with the general stoichiometric formula M₂O are not typical for these metals. Much more typical are the peroxides of these metals of the type M₂O₂ and MO₂, whose crystal lattices contain O₂²⁻ and O₂⁻ ions. Oxides of potassium, rubidium and cesium having the general stoichiometric formula M₂O₃ are also known. In addition to the above-listed types of compounds, cesium can form the suboxides Cs₇O₂, Cs₄O, Cs₃O and Cs₂O under certain conditions. At high temperature, the most stable oxides are those of type M₂O. The temperatures at which the pressures of the evaporation products of the alkali metal oxides attain atmospheric, are [917] (in °K):

Na₂O 2040 ± 60,

K₂O 1750 ± 100,

Rb₂O 1600 ± 100,

Cs₂O 1550 ± 100.

For an accurate thermodynamic calculation of systems, contains large quantities of alkali metals and oxygen, at lower temperatures than those given above, the formation of solid oxides must be taken into ac-

count.

The gaseous oxygen compounds of sodium, potassium, cesium and rubidium are evidently not stable [95, 915, 926, 928]. Thus, for example, at a temperature of 1740°K, when the total vapor pressure over molten K_2O is nearly 1 atmosphere, the partial pressure of K_2O in the vapor is only 10^{-4} at [917]. It was shown in the work [95] that the gaseous oxides KO , RbO and CsO are possibly more stable than the corresponding oxides of the type M_2O , but their partial pressures are nonetheless much lower than the partial pressures of the free metals and oxygen. Hence, the gas phase of the systems sodium, potassium, rubidium and cesium with oxygen can be represented with sufficient accuracy by the corresponding metals in the form of monatomic gases and oxygen.

The fluorides and chlorides of the alkali metals are considerably more stable than their oxides. The polymeric molecules of the compounds of the alkali metals with halogens of the type $(MX)_n$ will not be considered in the Handbook; the concentration of these molecules in the vapor, particularly at elevated temperatures, is comparatively low and neglecting them practically does not affect the thermodynamic calculations of the corresponding systems. The fluorides and chlorides of rubidium and cesium are not discussed in the Handbook.

An important class of compounds which is not discussed in the Handbook, are the hydroxides of the alkali metals. The literature data show that for some metals these compounds have considerable stability in the gaseous state. Hence, the thermodynamic calculations of the compositions of systems, containing alkali metals, hydrogen and oxygen, cannot be carried out with complete accuracy on the basis of the data, given in the Handbook.

§115. MOLECULAR CONSTANTS

Na. The sodium atom in the ground state has the electronic configuration $1s^2 2s^2 2p^6 3s$, to which corresponds one 2S term. The series boundary of the states $1s^2 2s^2 2p^6 ({}^1S)n\ell$ is situated $41,449.65 \text{ cm}^{-1}$ above the lower level 2S of this atom. The states $1s^2 2s^2 2p^6 ({}^1S)n\ell$ consist of doublet terms with $L = \ell$. The terms which arise upon excitation of the electrons of the atomic residue, are situated $265,000 \text{ cm}^{-1}$ above the ground state 2S and are thus not examined in the present Handbook.

Table 273 gives the energy levels of the sodium atom, corresponding to the states $1s^2 2s^2 2p^6 ({}^1S)n\ell$ with values of $n \leq 11$. The excitation energies of these levels were adopted on the basis of the values, recommended by Moore [2941]. As in other instances, states with similar excitation energies are combined into a single level with a summary statistical weight and mean excitation energy.

The review of Moore gives only data for a relatively small number of terms ; at $n \leq 11$ these are mainly the terms S, P, D and F. The other terms have not been observed experimentally and their energy has been estimated in correspondence with the general rules (see page).

Thus, for a given n , the excitation energy of the term with $L = 3$ i.e., the energy of the last known term is ascribed to all absent terms with $L > 3$. The only exception is the terms $\dots 6h^2G$, to which is ascribed the excitation energy of the term $\dots 6g^2H$.

The maximum uncertainty in the estimation of the energy of the levels is assumed for the state $1s^6 2s^2 2p^6 7i^2I$ and amounts to approximately 90 cm^{-1} . The excitation energy of these terms is of the order of $39,000 \text{ cm}^{-1}$, hence a similar inaccuracy in the determination of the energy of the level does not give important errors in the results of the subsequent calculations.

TABLE 273

Energy Levels of the Sodium Atom

Номер уровня A	Состояние B		Статисти- ческий вес E	Энергия, см ⁻¹ F
	электронная конфигура- ция C	терм D		
0	1s ² 2s ² 2p ⁶ 3s	¹ S	2	0
1	1s ² 2s ² 2p ⁶ 3p	³ P	6	16967,65
2	1s ² 2s ² 2p ⁶ 4s	¹ S	2	25739,86
3	1s ² 2s ² 2p ⁶ 3d	³ D	10	29172,87
4	1s ² 2s ² 2p ⁶ 4p	³ P	6	30270,6
5	1s ² 2s ² 2p ⁶ 5s	¹ S	2	33200,7
6	1s ² 2s ² 2p ⁶ 4d, 4f	³ D, ³ F	24	34572
7	1s ² 2s ² 2p ⁶ 5p	³ P	6	35042
8	1s ² 2s ² 2p ⁶ 6s	¹ S	2	36372,7
9	1s ² 2s ² 2p ⁶ 5d, 5f, 5g 1s ² 2s ² 2p ⁶ 6p	³ D, ³ F, ³ G ³ P	48	37085
10	1s ² 2s ² 2p ⁶ 7s 1s ² 2s ² 2p ⁶ 6d — 6h 1s ² 2s ² 2p ⁶ 7p	¹ S ³ D, ³ F, ³ G*, ³ H ³ P	72	38400
11	1s ² 2s ² 2p ⁶ 8s 1s ² 2s ² 2p ⁶ 7d — 7l 1s ² 2s ² 2p ⁶ 8p	¹ S ³ D, ³ F, ³ G*, ..., ³ I* ³ P	98	39210
12	1s ² 2s ² 2p ⁶ 8d — 8k 1s ² 2s ² 2p ⁶ 9s, 9p	³ D, ³ F, ³ G*, ..., ³ K* ³ S, ³ P	128	39734
13	1s ² 2s ² 2p ⁶ 9d — 9l 1s ² 2s ² 2p ⁶ 10s, 10p	³ D, ³ F, ³ G*, ..., ³ L* ³ S, ³ P	162	40093
14	1s ² 2s ² 2p ⁶ 10d — 10m	³ D, ³ F, ³ G*, ..., ³ M*	194	40356
15	1s ² 2s ² 2p ⁶ 11s — 11n	¹ S, ³ D, ..., ³ N*	242	40535

*The terms in Table 273, which have not been given in the summary [2941] are designated by an asterisk.

A) Number of level; B) state; C) electronic configuration; D) term; E) statistical weight; F) energy.

K. The potassium atom in the ground state has the electronic configuration $1s^2 2s^2 2p^6 3s^2 3p^6 4s$, to which corresponds the single term 2S . The series limit of the states $1s^2 2s^2 2p^6 3s^2 3p^6 nl$ is situated $35,009.78 \text{ cm}^{-1}$ above the lower level 2S of this atom. The states $\dots 3s^2 3p^6 nl$ are doublet terms with $L = l$. The terms connected with an excitation of the electrons of the residual atom, have energies over

150,000 cm^{-1} and will thus not be considered in the present Handbook.

TABLE 274

Energy Levels of the Potassium Atom

Номер уровня A	Состояние		Статисти- ческий вес	Энергия, см^{-1}
	электронная конфигура- ция C	терм D		
0	...3s ² 3p ⁴ 4s	¹ S	2	0
1	...3s ² 3p ⁴ 4p	³ P	6	13023,65
2	...3s ² 3p ⁴ 5s	¹ S*	2	21026,8
3	...3s ² 3p ⁴ 3d	¹ D	10	21535,35
4	...3s ² 3p ⁴ 5p	³ P	6	24713,95
5	...3s ² 3p ⁴ 4d ...3s ² 3p ⁴ 6s	¹ D ¹ S	12	27406,32
6	...3s ² 3p ⁴ 4f	³ F	14	28127,7
7	...3s ² 3p ⁴ 6p	³ P	6	29004,9
8	...3s ² 3p ⁴ 5d ...3s ² 3p ⁴ 7s	¹ D ¹ S	12	30200
9	...3s ² 3p ⁴ 5f ...3s ² 3p ⁴ 5g	³ F ¹ G	32	30615
10	...3s ² 3p ⁴ 7p	³ P	3	31073,0
11	...3s ² 3p ⁴ 6d ...3s ² 3p ⁴ 8s	¹ D ¹ S	12	31707
12	...3s ² 3p ⁴ 6f — 6h ...3s ² 3p ⁴ 8p	³ F, ¹ G, ³ H ¹ P	60	31985
13	...3s ² 3p ⁴ 7d ...3s ² 3p ⁴ 9s, 9p ...3s ² 3p ⁴ 7f — 7i	¹ D ³ S, ¹ P ³ F, ¹ G*, ..., ¹ I*	98	32760
14	...3s ² 3p ⁴ 8d — 8k ...3s ² 3p ⁴ 10s, 10p	¹ D, ³ F, ¹ G*, ..., ¹ K* ³ S, ¹ P	128	33285
15	...3s ² 3p ⁴ 9d — 9l ...3s ² 3p ⁴ 11s, 11p	¹ D, ³ F, ¹ G*, ..., ¹ L* ³ S, ¹ P	154	33647
16	...3s ² 3p ⁴ 10d — 10m	¹ D, ³ F*, ..., ¹ M*	192	33850
17	...3s ² 3p ⁴ 11s — 11n	³ S, ¹ P, ¹ D, ³ F*, ..., ¹ N*	242	34060

*In Table 274 the terms which are not given in the summary [2941] are marked by an asterisk.

A) Number of level; B) state; C) electronic configuration; D) term; E) statistical weight; F) energy.

Table 274 gives the energy levels of the potassium atom and the corresponding states $1s^2 2s^2 2p^6 3s^2 3p^6 nl$ with values of $n \leq 11$. The excitation energies of these levels have been adopted on the basis of

the values, recommended by Moore [2941]; states with similar excitation energies are combined into a single level with a summary statistical weight and a mean excitation energy.

The summary of Moore gives data only for a relatively small number of terms of the potassium atom with $n \leq 11$; these are mainly the terms 2S , 2P , 2D and 2F . The other terms have not been observed experimentally and their energy has been estimated to correspond to the general rules (see page 56). Thus, in the absence of experimental data, an excitation energy of the term with $L = 3$, i.e., the energy of the last known term, has been ascribed to all terms with $L > 3$ for a given n .

The maximum uncertainty in the estimation of the energy of the levels is assumed for the state $1s^2 2s^2 2p^6 3s^2 3p^6 7i^2 I$ and amounts to approximately $\pm 170 \text{ cm}^{-1}$. The excitation energy of this term is of the order of $33,000 \text{ cm}^{-1}$ so that such an inaccuracy in the determination of the energy of a level leads to practically unimportant errors in the results of subsequent calculations.

Rb. The rubidium atom in the ground state 2S has the electronic configuration $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 5s$. The series limit of the states $\dots 4s^2 4p^6 nl$ is $33,691.02 \text{ cm}^{-1}$ above the lower level 2S of this atom. The states $\dots 4s^2 4p^6 (^1S)nl$ are doublet terms with $L = l$. The terms arising upon excitation of the electrons of the residual atom, have excitation energies over $120,000 \text{ cm}^{-1}$ and will therefore not be considered in the Handbook.

Table 275 gives the energy levels of the rubidium atom, corresponding to the states $\dots 4s^2 4p^6 (^1S)nl$ with values of $n \leq 11$. The excitation energies of these levels were adopted on the basis of the values, recommended by Moore [2941]. As in the other cases, the states with similar excitation energies are combined into a single level with

a summary statistical weight and mean excitation energy.

TABLE 275

Energy Levels of the Rubidium Atom

Номер уровня A	Состояние Bm		E Статисти- ческий вес	F Энергия, см ⁻¹
	электронная конфигура- ция C	терм D		
0	...4s ⁴ p ⁵ s	¹ S	2	000
1	...4s ⁴ p ⁵ p	¹ P _{1/2}	2	12578,96
2	...4s ⁴ p ⁵ p	¹ P _{3/2}	4	12816,56
3	...4s ⁴ p ⁵ d	¹ D _{3/2}	10	19355,19
4	...4s ⁴ p ⁵ s	¹ S	2	20133,6
5	...4s ⁴ p ⁵ p	¹ P	6	23767
6	...4s ⁴ p ⁵ d	¹ D	10	25702
7	...4s ⁴ p ⁵ s	¹ S	2	26311
8	...4s ⁴ p ⁵ f	¹ F	14	26792
9	...4s ⁴ p ⁵ p	¹ P	6	27858
10	...4s ⁴ p ⁵ d	¹ D	10	28689
11	...4s ⁴ p ⁵ s	¹ S	2	29047
12	...4s ⁴ p ⁵ f, 5g	¹ F, ¹ G	32	29287
13	...4s ⁴ p ⁵ p	¹ P	6	29848
14	...4s ⁴ p ⁵ d	¹ D	10	30281
15	...4s ⁴ p ⁶ s ...4s ⁴ p ⁶ f — 6h	¹ S ¹ F, ¹ G, ¹ H	56	30632
16	...4s ⁴ p ⁶ p	¹ P	6	30967
17	...4s ⁴ p ⁶ d ...4s ⁴ p ⁶ 10s ...4s ⁴ p ⁶ 7l — 7i	¹ D ¹ S ¹ F, ¹ G [*] , ..., ¹ I [*]	92	31415
18	...4s ⁴ p ⁶ 10p ...4s ⁴ p ⁶ 9d	¹ P ¹ D	16	31761
19	...4s ⁴ p ⁶ 8l — 8k ...4s ⁴ p ⁶ 11s, 11p	¹ F, ¹ G [*] , ..., ¹ K [*] ¹ S, ¹ P	110	31969
20	...4s ⁴ p ⁶ 10d ...4s ⁴ p ⁶ 9l — 9l	¹ D ¹ F, ¹ G [*] , ..., ¹ L [*]	154	32420
21	...4s ⁴ p ⁶ 10f — 10m ...4s ⁴ p ⁶ 11d	¹ F [*] , ¹ G [*] , ..., ¹ M [*] ¹ D [*]	192	32665
22	...4s ⁴ p ⁶ 11f — 11n	¹ F [*] , ¹ G [*] , ..., ¹ N [*]	224	32838

*The terms which are not given in the summary [2941] have been marked in Table 275 by an asterisk.

A) Number of level; B) state; C) electronic configuration; D) term; E) statistical weight; F) energy; G) Table 275 (continuation)

The summary of Moore [2941] gives data only for a relatively small

number of terms of the rubidium atom, mainly for the terms S, P, D and F. The terms with $L > 3$ and $n \leq 11$, have not been observed experimentally, as a rule, and their excitation energies were estimated in accordance with the general rules (see page 56), the excitation energy of the term with $L = 3$, i.e., the energy of the last known term, being ascribed to all the absent terms with $L > 3$, for a given n .

The maximum error in the estimation of the energy of the levels has been committed for the states $\dots 4s^2 4p^6 (1S) 7i^2 I$ and amounts to approximately $\pm 200 \text{ cm}^{-1}$. The excitation energy of this term is of the order of $31,000 \text{ cm}^{-1}$ so that such an inaccuracy in the determination of the energy gives practically unimportant errors as a result of subsequent calculations.

Cs. The cesium atom in the ground state 2S has the electronic configuration $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 4d^{10} 5s^2 5p^6 6s$. The series limit of the states $\dots 5s^2 5p^6 (1S) nl$ is $31,406.71 \text{ cm}^{-1}$ above the lower level 2S of this atom.

The states $\dots 5s^2 5p^6 (1S) nl$ consist of doublet terms with $L = l$. The terms arising upon excitation of the electrons of the atomic residue have excitation energies over $130,000 \text{ cm}^{-1}$ and are not considered in the Handbook for this reason.

Table 276 gives the energy levels of the cesium atom with the above indicated electronic configuration and values of $n \leq 11$. The excitation energies of these levels were adopted on the basis of the values recommended by Moore [2941]. The states with similar excitation energies were combined into a single level with a summary statistical weight and mean excitation energy.

The summary of Moore [2941] does not give the terms with large values of L and these were therefore estimated in accordance with the general rules (see page 56). Thus, for all terms with $L > 3$, (for a

given n) the excitation energy of the term with $L = 3$, i.e., the energy of the last known term is given.

The maximum error in the estimate of the energy levels was committed for the states $\dots 5s^2 5p^6 7d^2 I$ and amounts to not more than 100 cm^{-1} . The excitation energy of this term is of the order of $29,000 \text{ cm}^{-1}$ hence such an inaccuracy in the determination of the energies gives only slight errors in the results of the subsequent calculations.

Na^+ , K^+ , Rb^+ , Cs^+ . The positive ions of sodium, potassium, rubidium and cesium have closed electron shells, analogous to the electron shells of the noble gases. The ground states of these ions are consequently singlet states 1S . The energy levels of the ions of the alkali metals, connected with excitation of the s- and p-electrons, are $100,000$ - $200,000 \text{ cm}^{-1}$ above their ground states 1S ; hence, they will not be considered in the present Handbook.

NaF and KF . The experimental determination of the molecular constants of NaF and KF is very difficult because of the low volatility of these substances and the diffuse nature of their electronic spectra. The only work in which band spectra of sodium and potassium fluoride have been obtained is the work of Barrow and Caunt [648] who investigated the absorption spectra of the halogen compounds of the alkali metals in the ultraviolet region on a Hilger apparatus with low and medium dispersion. The spectra of these two molecules consisted of diffuse bands without edges. It was not possible to determine the constants of NaF from the spectrograms because the spectrum consisted only of five bands without any regularity in the magnitude of the intervals between the bands. The spectrum of KF , on the other hand, contained more than 20 bands which the authors of the work [648] interpreted as being connected with transitions from several consecutive vibrational levels of the ground state to the upper unstable excited state of this

molecule (or to a state with a low minimum potential energy). Because a strict sequence was also absent in the intervals between the bands of KF, Barrow and Caunt, in order to determine the vibration frequencies of the potassium fluoride molecule, estimated the anharmonicity constant, assuming that the derivative of $x_e \mu^{1/2}$ ($x_e = \omega_e x_e / \omega_e$ and μ the reduced mass) in the molecules of the halogen compounds of the alkali metals is of approximately the same magnitude as has been found earlier by Vago and Barrow [4044] for the molecules of other groups. On the basis of this hypothesis and the data of Levi [2596] the authors of the work [648] assumed that the magnitude of $x_e \mu^{1/2}$ in the halogen compounds of the alkali metals lies between 0.008 and 0.018 and calculated three sets of vibrational constants for KF:

$$\begin{array}{l} \text{I } \omega_e = 400, \omega_e x_e = 1.4; \quad \text{II } \omega_e = 430, \omega_e x_e = 2.3 \\ \text{and III } \omega_e = 380, \omega_e x_e = 0.9 \text{ cm}^{-1}. \end{array}$$

It can be seen from these values that it is impossible to determine sufficiently accurate constants for KF from the experimental data because of the diffuse nature of the spectrum. It should also be pointed out that the vibrational constants, found in the work [648] for the other halogen compounds of the alkali metals differ in several cases considerably from those obtained by other authors during studies of the infrared spectra of these molecules (see further on, constants of KCl).

Rittner [3444] adopting the ionic model for the molecules of the halogen compounds of the alkali metals and allowing for the polarizability of the ions, calculated several constants of these molecules (see page 1802). The vibration frequencies of NaF and KF, found by Rittner, are 477 and 410 cm^{-1} , respectively.

Similar values were calculated for these constants (450 and 378 cm^{-1}) for the ionic model and the same polarizabilities of the ions in

the work of Berkowitz [755a].

TABLE 276

Energy Levels of the Cesium Atom

Номер уровня A	Состояние		Статисти- ческий вес E	Энергия, см ⁻¹ F
	электронная конфигура- ция C	терм D		
0	...5s ² 5p ⁶ 6s	¹ S	2	0
1	...5s ² 5p ⁶ 6p	³ P _{1/2}	2	11178.24
2	...5s ² 5p ⁶ 6p	³ P _{3/2}	4	11732.35
3	...5s ² 5p ⁶ 5d	¹ D _{3/2}	4	14499.49
4	...5s ² 5p ⁶ 5d	¹ D _{5/2}	6	14597.08
5	...5s ² 5p ⁶ 7s	¹ S	2	19535.5
6	...5s ² 5p ⁶ 7p	³ P _{1/2}	2	21676
7	...5s ² 5p ⁶ 7p	³ P _{3/2}	4	21947
8	...5s ² 5p ⁶ 6d	³ D	10	22615
9	...5s ² 5p ⁶ 8s	¹ S	2	24317
10	...5s ² 5p ⁶ 4f	³ F	14	24472
11	...5s ² 5p ⁶ 8p	³ P	6	25764
12	...5s ² 5p ⁶ 7d	³ D	10	26060
13	...5s ² 5p ⁶ 9s ...5s ² 5p ⁶ 5f, 5g	¹ S ³ F, ³ G	34	26988
14	...5s ² 5p ⁶ 9p ...5s ² 5p ⁶ 8d	³ P ³ D	16	27762
15	...5s ² 5p ⁶ 10s ...5s ² 5p ⁶ 6f - 6h	¹ S ³ F, ³ G, ³ H	56	28345
16	...5s ² 5p ⁶ 10p ...5s ² 5p ⁶ 9d	³ P ³ D	16	28800
17	...5s ² 5p ⁶ 11s ...5s ² 5p ⁶ 7f - 7i	¹ S ³ F, ³ G*, ..., ³ I*	80	29148
18	...5s ² 5p ⁶ 11p ...5s ² 5p ⁶ 10d	³ P ³ D	10	29450
19	...5s ² 5p ⁶ 8f - 8k	³ F, ³ G*, ..., ³ K*	110	29679
20	...5s ² 5p ⁶ 11d ...5s ² 5p ⁶ 9f - 9l	³ D ³ F, ³ G*, ..., ³ L*	154	30033
21	...5s ² 5p ⁶ 10f - 10m	³ F, ³ G*, ..., ³ M*	182	30302
22	...5s ² 5p ⁶ 11f - 11n	³ F, ³ G*, ..., ³ N*	224	30470

*The terms in Table 276, which are not given in the summary [1914], are marked by an asterisk.

A) Number of level; B) state; C) electronic configuration; D) term; E) statistical weight; F) energy.

Grabner and Hughes [1836] carried out an investigation on the electronic resonance spectrum of potassium fluoride, and estimated the

vibration frequencies of KF on the basis of the relative intensity of the transitions corresponding to various vibrational states of this molecule. The value thus found was $390 \pm 40 \text{ cm}^{-1}$, but the accuracy of this method is low and analogous measurements for the molecule RbF and CsF led to incorrect vibration frequencies.

Because of the absence of reliable vibration frequency data on NaF and KF in the literature, the authors of the Handbook carried out an estimate of these values during the preparation of the present edition by the Guggenheimer method (see page 88) on the basis of the relation of the force constants of the molecules of the halogen compounds of the metals and the ionization potential of the metal and also the interatomic distances of the molecules. The constants thus found are 520 ± 30 and $430 \pm 30 \text{ cm}^{-1}$ for NaF and KF, respectively.

On the basis of these estimates and also the values obtained in the works [648, 3444, 1836] the values 520 ± 30 and $430 \pm 30 \text{ cm}^{-1}$ have been adopted in the Handbook for the vibration frequencies of the molecules of sodium and potassium fluoride.

Following completion of the calculations of the thermodynamic functions of sodium and potassium fluoride another estimate of the vibrational constants of NaF and KF was made in the works of Vasilevskiy and Baykov [118] and Koryazhkin and Tatevskiy [250] (see page 1802). The values of ω_e found by these authors for the molecules NaF (521 and $530 \pm 30 \text{ cm}^{-1}$) and KF (437 and $420 \pm 25 \text{ cm}^{-1}$) are in good agreement with those given above. It should be pointed out that the anharmonicity constants of NaF and KF were estimated in the work [250] in addition to their vibration frequencies (3.15 and 2.34 cm^{-1}).

The rotational spectra of the molecules NaF and KF have not been investigated until recently. Attempts to determine the interatomic distance in these molecules by the electron diffraction method (see

[324]) were not crowned with success owing to the marked dimerization of sodium fluoride and potassium fluoride vapor. Grabner and Hughes [1836] found as a result of a study of the electron resonance spectrum of potassium fluoride that the interatomic distance in this molecule is 2.55 Å. Honig et al [2116] and Krasnov [259, 260] estimated the interatomic distances in the molecules of NaF and KF on the basis of the interatomic distances in the molecules of the halogen compounds of alkali metals, found from their microwave spectra. The values found by them are in good agreement and are 1.84; 1.877 and 1.917 Å for NaF and 2.129; 2.142 and 2.166 Å for KF. The last value is in good agreement with that obtained in 1961 by Green and Lew [1850] during measurements of the rotational spectrum of KF in the radio frequency region of wavelengths ($r_e = 2.17144 \pm 0.00005$ Å). The rotational constant of NaF given in Table 277 was calculated by means of the interatomic distance, proposed by Krasnov [260], and that of KF, on the basis of the data of Green and Lew [1850]. The uncertainty in the adopted interatomic distance for NaF does not exceed ± 0.03 Å.

The molecules NaF and KF, like the molecules of other diatomic compounds of halogens and alkali metals, have the fundamental electronic states $^1\Sigma$. The known excited states of the molecules of the alkali metal halogenides are unstable states or states in which the potential energy curve of the molecule has a shallow minimum, and they are not considered in the present Handbook for this reason.

NaCl and KCl. The absorption band spectrum of sodium and potassium chloride in the near ultraviolet was first obtained in the work of Levi [2596]. On the basis of the measured intervals between the intensity maxima of the diffuse bands of these molecules and the assumption of the instability of their excited states, Levi calculated the vibrational constants of NaCl and KCl in the ground states $X^1\Sigma$ (in cm^{-1}):

$\omega_e = 380$, $\omega_e x_e = 1.0$ and $\omega_e = 280$, $\omega_e x_e = 0.9$, respectively. Barrow and Caunt [648] reprocessed the data of Levi for KCl and, estimating by means of the relation $x_e \mu^{1/2} = 0.013$ (see above, page 1870), the magnitude of $\omega_e x_e$, found $\omega_e = 305$ and $\omega_e x_e = 0.9 \text{ cm}^{-1}$.

Klemperer and Rice in 1957 [3429] investigated the infrared absorption spectra of these molecules in the region of their fundamental frequencies. As in the case of LiCl (see page 1806), the spectrum was obtained with a low-dispersion apparatus and had the form of a single band, although it really consisted of several bands, corresponding to the transition $\Delta v = 1$ between a large number of vibrational levels of the ground state of the investigated molecules, because of the low vibration frequency of NaCl and KCl and the high temperature of the vapor. It follows from the general formulas (see [2020]) that in the vibrational-rotational spectrum of a diatomic molecule at $\omega_e x_e > 0$ and $\alpha_1 > 0$ the shortwave limit of the bands with $\Delta v = 1$ is the edge of the R-branch of the band 1-0. On the basis of this fact and using for the rotational constants of NaCl and KCl the values found with great accuracy in the microwave spectrum investigations, Klemperer and Rice calculated the rotational quantum numbers J , to which corresponds the edge of the band 1-0 and the wave numbers of the beginning of this band. In the order to determine the anharmonicity constant of the halogenides of the alkali metals, the authors of the work [3429] used the relation

$$\omega_e x_e = \frac{\left[\omega_e \frac{\alpha_1}{6} + B_e^2 \right]^2}{B_e^2},$$

obtained from equation (1.38). The vibrational constants of NaCl and KCl thus found proved to be $\omega_e = 366.1 \pm 4$, $\omega_e x_e = 2.05$ and $\omega_e = 281 \pm 6$, $\omega_e x_e = 1.30 \text{ cm}^{-1}$, respectively.

It is evident from these data that the vibration frequencies of NaCl and KCl, found in the works [2596, 3429] are in satisfactory agreement, particularly in the case of KCl, although the anharmonicity constants differ by a factor of 1.5-2. This discrepancy may evidently

be partly accounted for by the fact that the anharmonicity constants in the work of Klemperer and Rice were determined, not from experimental data, but were calculated by means of a theoretical relation, whose application to the molecules of the alkali metal halogenides has little justification. It should be pointed out that the rotational energy levels of NaCl and KCl, calculated by means of the constants, proposed by these authors, converge in a region with an energy which is 50-60% of the dissociation energy of these molecules; hence calculation of the thermodynamic functions of the corresponding gases at high temperatures at these values of the vibrational constants should result in considerable errors.

TABLE 277

Adopted Values of the Molecular Constants₁ of NaF, NaCl, KF, KCl in the Ground States $X^1\Sigma$

Молекула A	ω_e	$\omega_e x_e$	B_e	$\alpha_1 \cdot 10^2$	$D_e \cdot 10^7$	r_e
	cm ⁻¹					Å
NaF	520 ^a	—	0,46 ^a	—	—	1,917 ^a
NaCl	365	1,0	0,21805 ^b	0,161 ^{b,c}	2,87	2,3606
KF	430	—	0,268	—	—	2,17144
KCl	280,5	1,0	0,12864 ^f	0,079 ^{f,A}	1,05 ^e	2,6666

- A) Molecule;
a) estimate
b) constant of the molecule NaCl³⁵
c) $\alpha_1 = 4.84 \cdot 10^{-4} \text{ cm}^{-1}$
d) constant of the molecule KCl³⁵
e) $\alpha_2 = 1.67 \cdot 10^{-4} \text{ cm}^{-1}$
f) calculated by means of the relation (1.36).

In the cases in which, in addition to the magnitude of $\Delta G_{1/2}$, the dissociation energy of the corresponding diatomic molecule is known, its vibrational constants can be found by solving the Eqs. (1.40). Using the values $\Delta G_{1/2}$, calculated by means of the data [3429]* and the

dissociation energies of NaCl and KCl, given further on (see page 1898 and 1904), the authors of the Handbook found in the following constants for these molecules (in cm^{-1}): $\omega_e = 365 \pm 5$, $\omega_e x_e = 1.0 \pm 0.2$ and $\omega_e = 280.5 \pm 7$, $\omega_e x_e = 1.0 \pm 0.2$. The vibrational constants of NaCl and KCl found in this manner are given in Table 277 and have been adopted in the present Handbook. The constants of KCl, and also the magnitude of $\omega_e x_e$ for NaCl, given in Table 277, agree with the data of Levi within the limits of the determination accuracy. The discrepancy between the adopted vibrational frequency of NaCl and the value obtained by Levi considerably exceeds the possible inaccuracy in the determination of the beginning of the band of this molecule in the infrared spectrum and attests to the presence of errors in the measurements of Levi or to an incorrect numbering of the bands in this work. The important difference between the adopted anharmonicity constants and those found by Klemperer and Rice attests to the inaccurate estimate of the constants of these molecules by the use of theoretical relations, based on Morse functions. The differences in the values of ω_e and $\omega_e x_e$ calculated by means of the relations (1.38) and (1.40), characterize the magnitude of error in the values of these constants, which is shown in Table 277.

The rotational constants of NaCl and KCl were determined with high accuracy as a result of studies of their rotational spectra in the work [2116] by radiospectroscopic methods. The values given in Table 277 were recalculated by means of the physical constants adopted in the Handbook on the basis of the data of [2116]. As has been shown in the works [342, 2116], the interatomic distances in the molecules NaCl and KCl, found earlier by Maxwell and coworkers [2818] by the electron diffraction method, were erroneous, evidently because of the dimerization of the vapor of these substances.

§116. THERMODYNAMIC GAS FUNCTIONS

The thermodynamic functions of the alkali metals and also of the compounds of sodium and potassium with fluorine and chlorine in the ideal gas state, calculated for temperatures of 293.15-6000°K, are given in the Tables 361, 362, 364, 366, 368, 369, 371, 373, 375, 376, 378, 379 of Volume II of the Handbook. The differences in the constants of the isotope modifications of their molecules were not taken into account in the calculation of the thermodynamic functions of these gases.

Na, K, Rb, Cs. The thermodynamic functions of gaseous Na, K, Rb and Cs, given in Tables 361, 368, 375 and 378 of Volume II, were calculated by means of Eqs. (II.22) and (II.23). The translational components were calculated by means of the relations (II.8)-(II.9); the corresponding values A_{ϕ} and A_S are given in Table 278. The electronic components of the thermodynamic functions were calculated, allowance being for a finite number of electronic states of their atoms (see page 136) on the basis of the values given in Tables 273-276. In order to simplify the calculation at temperatures to 6000°K, all the electronic states of the atoms, corresponding to main quantum numbers of the valency electron with $n \leq 11$, were taken into account.

The uncertainties in the calculated values of the thermodynamic functions of Na, K, Rb and Cs at $T \leq 3500^\circ\text{K}$ are mainly due to the inaccuracy of the physical constants and do not exceed 0.005 cal/g-atom-degree in the values of Φ_T^* and S_T° . At higher temperatures the errors due to the use of an approximate calculation method and the absence of data on energies of the levels with $L \geq 3$ become important. However, even at 6000°K, the errors due to inclusion of excess levels (levels with $7 < n \leq 11$ at $T \leq 4500^\circ\text{K}$) and also the approximate estimate estimate of the energies of unobserved levels do not exceed 0.008, 0.03

and 0.04 cal/g-atom·degree in the values of Φ_T^* for Na, K, Rb and Cs, respectively. The errors due to the inaccuracy of determination of the maximum main quantum number exceeds these values considerably when the chosen method (see page 133) is used. For $\Delta n = \pm 1$ they amount to ± 0.02 , ± 0.07 , ± 0.07 and ± 0.09 cal/g-atom·degree in the values of Φ_{6000}^* for Na, K, Rb and Cs.

TABLE 278

Values (in cal/g-atom·degree) of the Constants A_Φ and A_S for the calculation of the thermodynamic Functions of Gaseous Na, K, Rb, Cs and Their Ions.

Вещество A	A_Φ	A_S
Na, Na ⁺	2,06168	7,03006
K, K ⁺	3,64477	8,61297
Rb, Rb ⁺	5,97631	10,94451
Cs, Cs ⁺	7,29204	12,26024

A) Substance.

The thermodynamic functions of the alkali metals in the gas state have been calculated by several authors. Overstreet [3158] calculated the values of Φ_T^* for monatomic sodium and potassium at temperatures up to 3000°K. The thermodynamic functions of sodium were also calculated by Benton and Inatomi [751] ($T \leq 2600^\circ\text{K}$) and for potassium, by Griffel [1859]. Katz and Margrave [2334], Evans et al [1513] and Kolsky, Gilmer and Gilles [2462] calculated the thermodynamic functions of monatomic sodium, potassium, rubidium and cesium for temperatures up to 2000, 2500 and 8000°K, respectively. The data of [1513] were also incorporated into the Handbook of the U. S. Bureau of Standards [3680] and the book of Stull and Sinke [3894]. At temperatures to 3000°K, the differences between the calculation results of the different authors and the data, given in the present Handbook, are due mainly to the difference in the physical constants, used in the calculations and do not exceed 0.001-0.002 cal/g-atom·degree. (The data of Overstreet [3158] differ considerably more, by up to ± 0.04 cal/g-atom·degree, but because of the absence of this work in the libraries of the USSR, the reasons for the discrepancies

could not be determined). At temperatures over 3000-4000°K the differences between the calculation results of Kolsky et al [2462] and the data of the Handbook increase rapidly with temperature and attain magnitudes of 0.021, 0.064 and 0.078 and 0.277 cal/g-atom·degree in the values of Φ_{6000}^* for Na, K, Rb and Cs, respectively. These differences are due to the fact that in the work [2462] in the calculation of the thermodynamic functions only those energy levels of the gas atoms were taken into account, which are given in the Handbook of Moore [2941] and that the quantum number n was not limited in any way. For the atoms Na, K and Cs, Moore gives mainly data for the states 2P (up to values of $n \approx 50-80$). At the same time, the states 2D and 2F which correspond to large values of n , and also all the states with $L > 3$ are absent in the Handbook of Moore. Hence, the values of the the thermodynamic functions of Na, K and Cs, obtained in the work [2462] are lower than those calculated by the authors of the present Handbook in spite of the fact that the 2P states of the atoms of the alkali metals were taken into account in this work up to the states with $n = 77$. In addition to transitions to the states 2D and 2F with high values of n (up to $n = 53$) were observed in the spectra of rubidium; the corresponding data were given by Moore. Hence, the functions of atomic rubidium, calculated in the work [2462], proved to be too high (up to 0.078 cal/g-atom·degree).

The differences between the values of the thermodynamic functions of monatomic sodium and potassium, given in the present and first edition of the Handbook, are due to the fact that the levels with excitation energies over $31,000 \text{ cm}^{-1}$ had not been included in the calculations for the first edition. These differences attain 0.06 and 0.2 cal/g-atom·degree in the values of Φ_{6000}^* for Na and K, respectively.

Na⁺, K⁺, Rb⁺, Cs⁺. The thermodynamic functions of Na⁺, K⁺, Rb⁺

and Cs^+ , calculated for temperatures of 293.15-6000°K, are given in Tables 362, 369, 376 and 379 of the second volume. The ions of the alkali metals have electronic ground states ^1S and excited electronic states with large excitation energies. Hence, the electronic components in the values of the thermodynamic functions of these gases within the considered temperature range are zero and the thermodynamic functions are equal to the translational components. The corresponding values of the thermodynamic functions of Na^+ , K^+ , Rb^+ and Cs^+ were calculated by means of the relations (II.8)-(II.9) with the values of A_ϕ and A_S , given in Table 278.

The differences in the values of the thermodynamic functions of ionized sodium and potassium given in the first and present editions of the Handbook and also the thermodynamic functions of Na^+ , K^+ , Rb^+ and Cs^+ , given in the present Handbook and the work [1851a], are due to the difference in the physical constants and do not exceed 0.002 cal/g-atom·degree.

KF, NaF. The thermodynamic functions of gaseous sodium fluoride and potassium fluoride, given in Table 364 (II) and 371 (II), were calculated on the basis of the constants in §115, using the approximate model of the rigid rotator-harmonic oscillator. Table 279 gives the magnitudes of C_ϕ and C_S for the calculation of the translational components and the rigid rotator in the value of Φ_T^* and S_T° of these gases and also θ for the calculation of the vibrational components.

Because the excited states of NaF and KF, like those of the molecules of the other halogen compounds of the alkali metals, have flat potential curves and a very limited number of levels, the existence of these states has not been taken into account in the calculations of the thermodynamic functions.

The basic uncertainties in the values of the thermodynamic func-

tions of NaF thus calculated at low temperatures are due to the approximate estimation of the interatomic distance of this molecule (of the order of 0.05-0.07 cal/mole·degree in Φ_T^* and S_T^0). At temperatures over 1000°K, the errors due to the inaccurate estimation of the vibrational frequencies of NaF and KF (about 0.1-0.15 cal/mole·degree in the values of Φ_{3000}^* and Φ_{6000}^*) and the use of an approximate method of calculation because of the absence of data on the anharmonicity constants of the vibrations and the interaction constants of these molecules (± 0.15 and ± 0.25 cal/mole·degree in the values of Φ_{3000}^* and Φ_{6000}^*) become important.

TABLE 279

Numerical Values of the Constants for the Calculation of the Thermodynamic Functions of Gaseous NaF, NaCl, KF and KCl

Вещество A	θ	$\alpha \cdot 10^6$	$\beta_1 \cdot 10^6$	$\beta_2 \cdot 10^6$	$\frac{q_0}{T}$	$\frac{d_0 \cdot 10^6}{T}$	C_Φ	C_S
	град B				град ⁻¹ B		кал/моль·град C	
NaF	748,17	—	—	—	—	—	4,6776	11,6331
NaCl	525,16	0,7397	2,7405	0,327	3,21688	8,46	7,1650	14,1205
KF	618,68	—	—	—	—	—	6,6349	13,5004
KCl	403,58	3,5651	0,6127	0,247	5,45935	9,01	8,9418	15,8973

A) Substance; B) degree; C) cal/mole·degree.

The total error in the values of $\Phi_{298.15}^*$, Φ_{3000}^* and Φ_{6000}^* for NaF and KF is ± 0.1 , ± 0.3 and ± 0.5 cal/mole·degree.

Data on other calculations of the thermodynamic functions of sodium fluoride and potassium fluoride are not to be found in the literature.

NaCl and KCl. The thermodynamic functions of gaseous sodium chloride and potassium chloride, given in Tables 366 (II) and 373 (II), were calculated by means of Eqs. (II.161) and (II.162). The values

$\ln \Sigma$ and $T \partial/\partial T \ln \Sigma$ were calculated by the method of Gordon and Barnes [Eqs. (II.137) and (II.138)] by means of the constants which have been adopted in the preceding paragraph, without applying corrections for the limited number of rotational levels of the ground states of the molecules NaCl and KCl. Table 279 gives the values of C_ϕ and C_S for the calculation of the translational components and the components of the rigid rotator in the values of the thermodynamic functions of these gases, the values θ and x for the interpolation in accordance with the anharmonic oscillator Tables and also the values of the coefficients in the Eqs. (II.137) and (II.138), calculated by means of the adopted molecular constants. Because the rotational constants of NaCl and KCl were determined for the isotopes NaCl^{35} and KCl^{35} (see Table 277), the "effective" values of the molecular constants for the isotope mixture, calculated on the basis of the relations (1.43), were used in the calculation of the thermodynamic functions of the two gases.

The main errors in the thermodynamic functions of NaCl and KCl calculated in this manner are due to the insufficient accuracy of the vibrational constants of these molecules used in the calculations (see page 1875).

Some idea of the magnitude of these errors may be gained from a comparison of the calculations of the thermodynamic functions of these gases on the basis of the constants, adopted in the present Handbook and those found in the works of Rice and Klemperer [3429] and Levi [2956]. The differences in the values ϕ_T^* of these gases, calculated by the method of Gordon and Barnes and the corresponding constants at 298.15; 3000 and 6000°K, attain 0.01 and 0.3 cal/mole·degree for potassium chloride. On the basis of these differences, the errors in the values of ϕ_T^* calculated in the Handbook for 298.15; 3000 and 6000°K

for the two gases are assumed to be ± 0.02 , ± 0.15 and 0.3 cal/mole \cdot degree.*

The thermodynamic functions of sodium chloride and potassium chloride had been calculated earlier by Kelley [2363] (S_T° at $T \leq 2000^\circ\text{K}$) and Rice and Klemperer [3430] ($T \leq 2000^\circ\text{K}$). The calculation of Kelley was carried out by means of the rigid rotator-harmonic oscillator model using the vibrational constants, found by Levi and the moments of inertia, calculated on the basis of the interatomic distances, found by Maxwell and co-workers [2818]. The differences between the data of Kelley and the values given in the present Handbook, do not exceed 0.2 cal/mole \cdot degree.

The calculation of Rice and Klemperer was carried out by the method of Maier and Goeppert-Maier and the constants, found by Rice and Klemperer in the work [3429] (see page 1874). The differences between the data of these authors and the values, given in the present Handbook, do not exceed 0.02 - 0.04 cal/mole \cdot degree; the difference increases with temperature.

§117. THERMODYNAMIC PROPERTIES OF Na, NaF, NaCl, K, KF, KCl, Rb, AND Cs IN THE SOLID AND LIQUID STATES

The thermodynamic functions of Na, NaF, NaCl, K, KF, KCl, Rb and Cs in the solid and liquid states (Tables 360, 363, 365, 367, 370, 372, 374 and 377 of the 2nd volume) were calculated by means of Eqs. (III.9)-(III.11) on the basis of the thermodynamic quantities given in Table 280. The uncertainties in the values of Φ_T^* at temperatures of 298, 1000, 1500, 2000 and 3000°K , estimated on the basis of an analysis of the possible errors in the original data, are given in Table 281.

The alkali metals considered in this Section (Na, K, Rb and Cs) have a structure of the type of cubic body-centered packing (structural type of Na) at room temperature. At low temperatures sodium goes

over into the cubic dense packed modification. The fluorides and chlorides of sodium and potassium are known only in the form of cubic modifications (structural type of NaCl) and do not give polymorphous transformations.

Na. On the basis of the results of heat capacity measurements on sodium, carried out by Simon and Zeidler [3736] (16-118°K), Dauphinee, McDonald and Preston-Thomas [1267] (55-315°K), Parkinson and Quarrington [3190] (1.4-20°K), Stull and Sinke [3894] calculated the values $S_{298.15}^{\circ} = 12.21$ cal/g-atom·degree and $H_{298.15}^{\circ} - H_0^{\circ} = 1532$ cal/g-atom. The results of the heat capacity measurements on sodium, carried out later by Roberts [3451] (1.5-20°K) and Martin [2786a] (20-300°K) cannot modify these values to any great extent because the correction to be applied to the above-given entropy amounts to only 0.03 cal/g-atom·degree on the basis of the data of Roberts [3451] and to 0.03 cal/g-atom·degree* on the basis of Martin's data [2786a].

The values of $S_{298.15}^{\circ}$ and $H_{298.15}^{\circ} - H_0^{\circ}$, recommended by Stull and Sinke [3894] have been adopted in the present Handbook; the accuracy of these values is ± 0.05 cal/g-atom·degree and ± 5 cal/g-atom, respectively. The values obtained in the work of Evans, Jacobson, Munson and Wagman [1513], who re-examined all the experimental data on the thermodynamic properties of sodium published up to 1955, and who composed a Table of the thermodynamic functions up to 1200°K,** were used in establishing the Table of thermodynamic functions of solid and liquid sodium [Table 360 (2)] within the range of 298-1200°K. For the heat of melting of sodium, the authors of the work [1513] obtained the value $\Delta H_{370.97} = 622$ cal/g-atom. The specific heat of liquid sodium above 1200°K was assumed to be constant and equal to $C_{pf200}^{\circ} = 7.11$ cal/g-atom·degree [1513].

TABLE 280

Adopted Values of the Thermodynamic Properties of Sodium, Potassium, Rubidium, Cesium and their Compounds in the Solid and Liquid State

Вещество A	Состояние B	$H_{298.15}^{\circ} - H_0^{\circ}$	$S_{298.15}^{\circ}$	$C_{p,298.15}^{\circ}$	Коэффициенты в уравнении для C_p° в E			Интервал температуры F	T_m	ΔH_m
		кал/моль C	кал/моль-град D	кал/моль-град D	a	b · 10 ³	c · 10 ⁻⁵	°K	°K	кал/моль F
Na	Крист. G	1532	12,21	6,74	6	—	—	298,15—370,97	370,97	622
Na	Жидк. H	—	—	—	6	—	—	370,97—1200	—	—
Na	•	—	—	—	7,11	—	—	1200—2300	—	—
NaF	Крист.	2027	12,26	11,19	10,40	3,88	0,33	298,15—1285	1285	8030
NaF	Жидк.	—	—	—	16,4	—	—	1285—3600	—	—
NaCl	Крист.	2534	17,35	12,14	10,98	3,90	—	298,15—1073	1073	6850
NaCl	Жидк.	—	—	—	16,0	—	—	1073—3400	—	—
K	Крист.	1695	15,46	7,16	6	—	—	298,15—336,4	336,4	554
K	Жидк.	—	—	—	6	—	—	336,4—1000	—	—
K	•	—	—	—	7,26	—	—	1000—2300	—	—
KF	Крист.	2391	15,91	11,64	11,266	3,858	0,688	298,15—500	—	—
KF	•	—	—	—	11,885	2,07	—	500—1130	1130	6750
KF	Жидк.	—	—	—	16,0	—	—	1130—3200	—	—
KCl	Крист.	2710	19,70	12,31	9,89	5,20	-0,77	298,15—1043	1043	6100
KCl	Жидк.	—	—	—	16,0	—	—	1043—3100	—	—
Rb	Крист.	1790	18,22	7,38	6	—	—	298,15—312	312	560
Rb	Жидк.	—	—	—	7,50	—	—	312—2300	—	—
Cs	Крист.	1859	20,16	7,50	6	—	—	298,15—301,8	301,8	510
Cs	Жидк.	—	—	—	7,60	—	—	301,8—2400	—	—

- a) $C_p^{\circ} = a + bT - cT^{-2}$ (cal/mole·degree)
 b) Table data of [1513]
 c) Table data of [3894]
 A) Substance;
 B) state;
 C) cal/mole;
 D) cal/mole·degree;
 E) coefficients in the equation for C_p° ;
 F) temperature range;
 G) crystalline;
 H) liquid.

The thermodynamic functions of sodium in the solid and liquid state, calculated for the temperatures 293.15-2300°K, are given in Table 360 (II). The uncertainties in the calculated values of Φ_T^* are given in Table 281.

The thermodynamic functions of sodium [Table 360 (II)] do not differ from the values given in the Handbook of Stull and Sinke [3894]

for temperatures to 1100°K.

NaF. King [2402] measured the specific heat of sodium fluoride within the range 54-296°K and calculated the value $S_{298.15}^{\circ} = 12.26 \pm \pm 0.07$ cal/mole·degree. The value S_{51}° , calculated by extrapolation of the specific heat to below 51°K is 0.50 cal/mole·degree. The value $S_{298.15}^{\circ} - H_0^{\circ} = 2027$ cal/mole has been calculated on the basis of King's data [2402].

The most accurate measurements of the enthalpy of NaF at high temperatures have been made by O'Brien and Kelley [3106] (406-1747°K). The data obtained by them differ by about 3% from the results of the measurement, carried out earlier by Krestovnikov and Karetnikov [262] (288-1073°K) and Lyashenko [284] (290-1287°K). O'Brien and Kelley [3106] obtained an equation for the specific heat of solid NaF (see Table 280), the melting point (1285°K), the heat of melting (8.03 kcal/mole) and the specific heat of molten sodium fluoride ($C_p^{\circ} = 16.4$ cal/mole·degree). The last value has been used in the Handbook for the calculation of the thermodynamic functions of liquid sodium fluoride at temperatures above 1800°K.

The thermodynamic functions of sodium fluoride in the solid and liquid state, calculated for temperatures of 293.15-3600°K, are given in Table 363 (II). The uncertainties in the calculated values of ϕ_T^* are given in Table 281.

NaCl. Kelley [2364] recommended the value $S_{298.15}^{\circ} = 17.3 \pm 0.3$ cal/mole·degree on the basis of insufficiently reliable data on the specific heat of sodium chloride, obtained by Nernst [3042] (25-84°K) and McGraw [2707] (95-245°K). Recent measurements on the heat capacity of NaCl at low temperatures were carried out by Clusius, Goldmann and Perlick [1135] (11-268°K), Martin [2785] (2-30°K) and Morrison, Patterson and Dugdale [2955] (2.5-20°K). The results of these measure-

ments enable the entropy at 298°K to be calculated more precisely, giving $S_{298.15}^{\circ} = 17.35 \pm 0.1$ cal/mole·degree.* The value $H_{298.15}^{\circ} - H_0^{\circ} = 2534 \pm 15$ cal/mole was calculated on the basis of the data [1135]. The enthalpy of solid and liquid NaCl at high temperatures was measured by numerous authors; on the basis of these data, Kelley [2363] derived a linear equation for the specific heat of solid NaCl (see Table 280) and also calculated the heat of melting $\Delta H_{1073} = 6.8$ kcal/mole and the specific heat of molten NaCl (at $T \leq 1300^{\circ}\text{K}$) $C_p^{\circ} = 16.0$ cal/mole·degree. In 1951, Popov and Gal'chenko [333] carried out measurements of the specific heat of NaCl within the range of 423-973°K. The equation derived by them ($C_p^{\circ} = 10.3 \pm 5.10 \cdot 10^{-3}T$) gives lower values at temperatures under 560°K and higher values above 560°K for the specific heat of NaCl as compared with those recommended by Kelley [2363]. Within the temperature range 400-700°K, the difference between the data of [2363, 333] does not exceed about 1%. The increase in the difference at higher temperatures (approximately to 3% at 1000°K) is probably due to the lower accuracy of the Kelley equation [2363] and the accuracy of determination of the absolute values of the true specific heat in the work [333] at these temperatures.

The equation recommended by Kelley [2363] which agrees with the above-given heats of melting** and specific heat of molten NaCl has been adopted in the Handbook for the calculation of the thermodynamic functions of NaCl. It should be pointed out that the values of Φ_T^* , calculated by means of the data [333, 2363] differ by not more than 0.05 cal/mole·degree. The specific heat of molten NaCl $C_p^{\circ} = 16.0$ cal/mole·degree is also used at temperatures above 1300°K.

The thermodynamic functions of sodium chloride in the solid and liquid state, calculated for temperatures of 293.15-3400°K, are given in Table 365 (II). The uncertainties in the calculated values of Φ_T^* are

given in Table 281.

K. Stull and Sinke [3894], based on the results of the specific heat measurements on potassium, carried out by Dauphinee, Martin and Preston-Thomas [1268] (30-330°K) and Wallace, Krier and Craig [4133] recommended the value $S_{298.15}^{\circ} = 15.39$ cal/g-atom·degree, calculated on the assumption that $S_{12}^{\circ} = 0.33 \pm 0.02$ cal/g-atom·degree, (extrapolation of the heat capacity to 0°K). The work of Roberts [3451] published in 1957 on a study of the specific heat of potassium within the temperature range 1.5-20°K gives the higher value of $S_{12}^{\circ} = 0.40$ cal/g-atom·degree. Calculation on the basis of Robert's measurements gives $S_{298.15}^{\circ} = 15.46 \pm 0.05$ cal/g-atom·degree and $H_{298.15}^{\circ} - H_0^{\circ} = 1695 \pm 5$ cal/g-atom, which have been adopted in the Handbook. The results of the calculations of the thermodynamic functions of potassium, carried out by Evans et al [1513], have been used in the compilation of the Tables of the thermodynamic functions of solid and molten potassium within the range of 298-1000°K. For the heat of melting of potassium these authors calculated the value $\Delta H_{336.4} = 554$ cal/g-atom. The specific heat of liquid potassium above 1000°K has been assumed to be constant, amounting to $C_{p1000}^{\circ} = 7.26$ cal/g-atom·degree [1513].

The thermodynamic functions of potassium in the solid and liquid state, calculated for the temperatures 293.15-2300°K, are given in Table 367 (II). The magnitude of the uncertainties in the calculated values of Φ_T^* is given in Table 281.

The thermodynamic functions of potassium [Table 367 (II)] differ from the values, given in the Handbook of Stull and Sinke [3894] up to 1000°K by not more than 0.07 cal/g-atom·degree. The cause of these slight differences is the more precise entropy of potassium at low temperatures, obtained as a result of the measurements of Roberts [3451] which have been taken into account in the present Handbook.

KF. Westrum and Pitzer [4224] measured the specific heat of potassium fluoride within the range of 16-298°K and obtained the values $S_{298.15}^{\circ} = 15.91 \pm 0.1$ cal/mole·degree and $H_{298.15}^{\circ} - H_0^{\circ} = 2391$ cal/mole. The value S_{16}° , obtained by extrapolation of the specific heat to 0°K is 0.04 cal/mole·degree. The enthalpy of KF at high temperatures was measured by Westrum and Pitzer [4224] (325-530°K) and Lyashenko [284] (429-1187°K). The equation for the specific heat of KF, derived by Westrum and Pitzer [4224] has been used in the Handbook for the temperature range 298-500°K (see Table 280). At temperatures above 500°K, for solid potassium fluoride the authors of the Handbook derived a linear equation on the basis of the values $C_{p500}^{\circ} = 12.92$ cal/mole·degree [4224] and $H_{1130}^{\circ} - H_{500}^{\circ} = 8550$ cal/mole. The last value and also the heat of melting of KF $\Delta H_{1130} = 6.75 \pm 0.3$ kcal/mole were taken from the Table of Kelley's Handbook [2363] compiled on the basis of the data of Lyashenko [284]. The specific heat of molten potassium fluoride is assumed to be 16.0 cal/mole·degree.

The thermodynamic functions of potassium fluoride in the solid and liquid state, calculated for the temperatures 293.15-3200°K, are given in Table 370 (II). The uncertainties in the calculated values of Φ_T^* are given in Table 281.

KCl. The specific heat of potassium chloride at low temperatures has been investigated several times. Kelley [2364], based on several works, carried out before 1939, recommended the value $S_{298.15}^{\circ} = 19.76 \pm 0.07$ cal/mole·degree. In later years, the specific heat of KCl was measured at low temperatures by Clusius, Goldman and Perlick [1135] (11-268°K), Strelkov, Itskevich, Kostryukov and Mirskaya [396, 3097] (12-300°K), Keesom and Pearlman [2342] (1.5-4.3°K) and Berg and Morrison [753] (2.5-270°K). The data of all these authors are in good agreement and give values of $S_{298.15}^{\circ} = 19.70 \pm 0.05$ cal/mole·degree and

$H_{298.15}^{\circ} - H_0^{\circ} = 2710 \pm 10$ cal/mole. An equation derived by Kelley [2363] on the basis of several experimental investigations on the enthalpy of KCl (see Table 280) was used in the Handbook for the specific heat of solid KCl at higher temperatures. An investigation on the enthalpy of KCl carried out by Skuratov and Lapushkin [382] in the range 675-933°K was published in 1951. The linear equation for the specific heat of KCl derived by these authors gives lower values for room temperature and somewhat excessive values at higher temperatures (by 2% in the range 700-1000°K) compared with the Kelley equation [2363]. The differences between the values of Φ^* for solid potassium chloride calculated by means of the data [2363, 382] do not exceed 0.08 cal/mole·degree (at the melting point of KCl).

The heat of melting of potassium chloride $\Delta H_{1043} = 6.1 \pm 0.3$ kcal/mole* and the heat capacity of molten KCl have been adopted in the Handbook on the basis of the recommendations of Kelley [2363].

The thermodynamic functions of potassium chloride in the solid and liquid state, calculated for the temperatures 293.15-3100°K, are given in Table 372 (II). The uncertainties in the calculated values of Φ_T^* are indicated in Table 281.

Rb and Cs. Dauphinee, Martin and Preston-Thomas [1268] investigated the specific heat capacity of rubidium (25-325°K) and cesium (20-310°K). Stull and Sinke [3894] used these data to calculate the values of $S_{298.15}^{\circ}$ and $H_{298.15}^{\circ} - H_0^{\circ}$ which are 18.22 cal/g-atom·degree and 1790 cal/g-atom for rubidium and 20.16 cal/g-atom·degree and 1859 cal/g-atom for cesium, respectively. The possible uncertainties in the entropy values at 298.15°K for Rb and Cs amount to approximately ± 0.3 and ± 0.4 cal/g-atom·degree and are due to the relatively high entropies obtained by extrapolation of the specific heat to 0°K on the basis of the Debye equation (for Rb $S_{25} = 3.44$, $\theta_D = 61^{\circ}\text{K}$, and for Cs $S_{20}^{\circ} =$

=3.91 cal/g-atom·degree, $\theta_D = 44^\circ\text{K}$) as well as the anomalies in the specific heats of rubidium and cesium.*

TABLE 281

Uncertainties (in cal/mole·degree) in the Values of Φ_T^* for Sodium, Potassium Rubidium, Cesium and Their Compounds in the Solid and Liquid State

T°K	298	1000	1500	2000	3000
Na	±0.05	±0.15	±0.25	±0.4	—
NaF	±0.07	±0.15	±0.25	±0.4	±1.0
NaCl	±0.10	±0.2	±0.3	±0.5	±1.2
K	±0.05	±0.15	±0.25	±0.4	—
KF	±0.10	±0.2	±0.3	±0.5	±1.2
KCl	±0.05	±0.15	±0.3	±0.5	±1.2
Rb	±0.3	±0.4	±0.7	±1.0	—
Cs	±0.4	±0.5	±0.8	±1.2	—

The melting points of Rb and Cs were taken as 312.0 [3508] and 301.8°K [1136] and the heats of melting $\Delta H_{312} = 560$ and $\Delta H_{301.8} = 510$ cal/g-atom [3894], respectively. According to the estimate of Stull and Sinke [3894], the specific heat of molten rubidium is 7.5 cal/g-atom·degree, and that of molten cesium 7.6 cal/g-atom·degree.

The thermodynamic functions of rubidium and cesium in the solid and liquid state, calculated for the temperature range 293.15-2300°K, are given in Tables 374 and 377 of the second volume. The uncertainties in the calculated values of Φ_T^* are given in Table 281.

The thermodynamic functions of rubidium and cesium do not differ from the values given in the Handbook of Stull and Sinke [3894] for temperatures to 900°K.

§118. THERMOCHEMICAL QUANTITIES

The standard state of sodium is Na (cryst.).

Na (gas). The results of the measurements on the pressure of saturated sodium vapor carried out prior to 1955, were re-examined by Evans, Jacobson, Munson and Wagman [1513]. Taking into account the dimerization of the sodium atoms in the vapor ($D_0(\text{Na}_2) = 17.53 \pm 0.15$ kcal/mole), the authors of the work [1513], based on some of the most reliable works [2513, 3963, 1463, 3468, 3471, 1915, 4193, 2748] calculated the heat of sublimation of sodium $\Delta H_{s_0} = 25.92 \pm 0.05$ kcal/g-atom. Application of a correction, allowing for the difference between the values for the specific heat of sodium at low temperatures, adopted in the work [1513] and in the present Handbook, gives the value

$$\Delta H_{s_0}(\text{Na, cryst.}) = \Delta H^\circ f_0(\text{Na, gas}) = 25.97 \pm 0.1 \text{ kcal/mole}^{\circ}$$

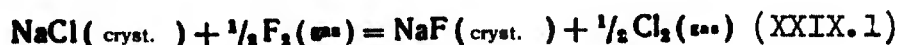
Na⁺ (gas). The ionization potential of the sodium atom is 41,449.65 cm^{-1} or

$$I(\text{Na}) = 118,515 \text{ kcal/g-atom}$$

and has been adopted on the basis of Moore's recommendation [2941]. The uncertainty in this value does not exceed $\pm 0.1 \text{ cm}^{-1}$ or $\pm 0.3 \text{ cal/g-atom}$. To the adopted ionization potential corresponds

$$\Delta H^\circ f_0(\text{Na}^+, \dots) = 144,485 \pm 0.1 \text{ kcal/g-atom.}$$

NaF (cryst.). The heat of the reaction

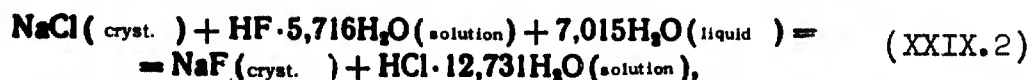


has been measured by Wartenberg and Fitzner [4162] and Schmitz and Schumacher [3641]. Similar values of -39.3 and -39.5 kcal/mole were found in these works. To these values correspond the heats of formation of crystalline sodium fluoride of -137.8 and -138.0 kcal/mole. However, taking into account the great difficulties in working with fluorine and also the fact that the preparation of sufficiently pure ele-

mental fluorine has only recently become possible, the uncertainties in the values found in the works [4162, 3641] must obviously be much greater (of the order of several kilocalories per mole).

The heat of formation of sodium fluoride can be calculated on the basis of measurements of the heat of neutralization of a solution of NaOH by HF solution. On the basis of these data Bichowsky, Rossini et al [813, 3508] calculated the value of $\Delta H^{\circ}f_{298.16} = -136.0$ kcal/mole. Based on these same data, Vorob'yev and Skuratov [130, 129] calculated $\Delta H^{\circ}f_{298.16} = -136.17 \pm 0.3$ kcal/mole. New measurements of the heat of solution of sodium fluoride in water were carried out in the works [2568, 1993]. The value thus obtained $\Delta H_{298.16} = 213 \pm 10$ cal/mole (infinite dilution) differs considerably from that adopted in the works of Rossini et al. [813, 3508] and Vorob'yev and Skuratov [130, 129]* amounting to $\Delta H_{298.16} = 60$ cal/mole, based on the obsolete measurements of Thomsen [3981]. Application of corrections which take into account the more accurate heat of solution of NaF and heat of solution of metallic sodium in water (see page 1896) give a value of -136.4 kcal/mole.

The work of Coughlin [1195] in which the heats of solution of crystalline sodium fluoride and chloride in solutions of hydrofluoric acid has been measured, appeared in 1958. On the basis of these data the authors of the work [1195] found the heat of the reaction



to be $\Delta H_{298.16} = -1410 \pm 6$ cal/mole and calculated for the heat of formation of sodium fluoride the value $\Delta H^{\circ}f_{298.16} = -136.3 \pm 0.2$ kcal/mole. Application of a correction allowing for the differences between the heats of formation of NaCl as adopted in the work [1195] and in the present Handbook, gives a value of

$$\Delta H^{\circ}_{\text{sub}}(\text{NaF, cryst.}) = -136.6 \pm 0.3 \text{ kcal/mole.}$$

This value, based on the measurements of Coughlin [1195], is more accurate than that calculated in the works [813, 3508, 130, 129] and has been adopted for the present Handbook.

NaF (gas). The results of the calculations of the heat of sublimation of sodium fluoride, carried out on the basis of measurements of the pressure of the saturated vapor, are given in Table 282.

Saturated sodium fluoride vapor contains dimeric Na_2F_2 molecules. Miller and Kusch [2914] investigated the velocity distribution of the molecules in the molecular beam of sodium fluoride and found that at 1146°K the saturated vapor contains 9% of the dimer. Later on, Eisenstadt, Rothberg and Kusch [1472] carried out more accurate measurements and found at 1170° the dimer contributes 19% to the total pressure. The results of the mass spectrometric studies of Berkowitz and Chupka [757] confirm the results of the work [1472]. The measurements of Pugh and Barrow [3344] and Porter and Schoonmaker [3307] are less accurate.

The last column of Table 282 gives the heats of sublimation of NaF molecules, calculated by means of an approximate allowance for the dimerization of the vapor. On the basis of the data [1472], the dimer concentration in the vapor was assumed to be 19% for the calculation and the assumption was made that the dimer concentration is constant over the entire temperature range.

The main object of the work of Kusch et al [2914, 1472] was the determination of the composition of the vapor and not of the pressure of the saturated vapor. These authors estimated the uncertainty in the measured vapor pressure as $\pm 50\%$. The results of the measurements of Naryshkin [305] are insufficiently accurate, and the heats of sublimation calculated on this basis show a large scatter. Hence, these

works were not taken into account in the selection of the value for the heat of sublimation of NaF.

The Handbook adopted a heat of sublimation of NaF which is the average of the calculation results on the basis of the data [3097, 3344, 3563, 3685, 4170]

$$\Delta H_{s_0}(\text{NaF, cryst.}) = 66,3 \pm 1 \text{ kcal/mole.}$$

The uncertainty in this value is mainly due to the insufficient accuracy of the data on the vapor pressure and the dimerization of sodium fluoride vapor.

To the adopted heat of sublimation corresponds

$$\Delta H^{\circ} f_0(\text{NaF, gas}) = -69,74 \pm 1 \text{ kcal/mole,}$$

$$D_0(\text{NaF}) = 114,21 \pm 1,2 \text{ kcal/mole.}$$

TABLE 282

Results of the Calculations of the Heat of Sublimation of NaF

	Авторы А	Год В	Метод С	Интервал тем-пературы D °K	Число изме-рений Е	$\Delta H_{s_0}^a, \text{ F}$ ккал/моль	$\Delta H_{s_0}^b, \text{ F}$ ккал/моль
1	Вартенберг, Шульд [4170]	1921	Кипения	1672—1835	5	$65,6 \pm 0,2$	66,3
2	Руфф и др. [3563]	1922	"	1699—1974	14	$65,3 \pm 0,2$	66,1
3	Ниwa [3097]	1938	Эффузи-онный	1053—1112	5	$65,1 \pm 0,05$	65,5
4	Нарышкин [305]	1939	Протока	11188—1273	4	$64,9 \pm 1,3$	65,4
5	Миллер, Кусх [2914]	1956	Эффузи-онный	1115—1189	4	$68,3 \pm 0,1$	68,7
6	Сенс и др. [3685]	1957	Протока	1220—1348	16	$65,8 \pm 0,1$	66,3
7	Пью, Барроу [3344]	1958	Эффузи-онный	1023—1166	63	$66,8 \pm 0,4$	67,3
8	Эйзенштадт, Ротберг, Кусх [1472]	1958	Эффузи-онный	1113—1189	6	$69,4 \pm 0,1$	69,8

a) Heats of sublimation of NaF, calculated without allowance for the dimerization of the vapor; b) heats of sublimation of NaF, calculated with allowance being made for the dimerization of the vapor; c) variant of the method ("torsion" method). A) Authors; B) year; C) method; D) temperature range, °K; E) number of measurements; F) kcal/mole.
 1) Wartenberg, Schulz [4170]; 2) Ruff et al [3563]; 3) Niwa [3097]; 4) Naryshkin [305]; 5) Miller, Kusch [2914]; 6) Sense et al [3685]; 7) Pugh, Barrow [3344]; 8) Eisenstadt, Rothberg, Kusch [1472]; 9) boiling; 10) effusion; 11) flow.

The dissociation energy of NaF has been measured in the work of Page and Sugden [3164] on the basis of a study of the radiowave ab-

sorption of sodium containing flames upon introduction of fluorine into these flames. The value of $D_0(\text{NaF}) = 120$ kcal/mole found by Page and Sugden greatly exceeds that obtained by calculation, using the heat of formation and sublimation of sodium fluoride. The causes of this difference, which exceeds the uncertainties of the values, adopted in the present Handbook, remain unclear. It should be pointed out that in the case of LiF and KF the values of D_0 found by the authors of the work [3134] are in satisfactory agreement with the values calculated by means of the heat of formation and sublimation of the corresponding substances (see page 1851 and 1901).*

NaCl (cryst.). Numerous data on the heats of neutralization of caustic soda solutions with hydrochloric acid,** the heats of solution of sodium chloride and the heats of dilution of its solutions have been examined in the works of Rossini et al. [3493, 3494, 813, 3508]. In the last work of Rossini et al [3508], the heat of formation of crystalline sodium chloride $\Delta H^\circ_f_{298.16} = -98.232$ kcal/mole was recommended on the basis of these data. When this calculation was carried out it was assumed that the heat of formation of NaOH solution (infinite dilution) is $\Delta H^\circ_f_{298.16} = -112.236$ kcal/mole. Ketchen and Wallace [2378, 2377] found the similar value of -112.37 kcal/mole. Messer, Fasolino and Thalmayer [2859] carried out a new, more accurate measurement of the heat of solution of metallic sodium in water and found $\Delta H_{298.16} = -44.23 \pm 0.13$ kcal/mole, which gives a value of $\Delta H^\circ_f_{298.16} = -112.547$ kcal/mole for the heat of formation of an infinitely diluted solution of caustic soda.***

Recalculation of the value, recommended by Rossini et al [3508] with more accurate data on the heat of formation of caustic soda solution being taken into account [2859] gives the value

$$\Delta H^\circ_{298.16}(\text{NaCl, cryst.}) = -98.54 \pm 0.2 \text{ kcal/mole,}$$

which has been adopted in the Handbook.

NaCl (gas). The most accurate data on the pressure of saturated sodium chloride vapor were obtained by the effusion method by An. N. Nesmeyanov and Sazonov [312] (745-948°K), Zimm and Mayer [4389] (760-897°K) and Niwa [3097] (883-973°K). Calculation of the heat of sublimation of sodium chloride on the basis of these data without allowing for the formation of polymeric molecules in the vapor gives values of 54.58 ± 0.13 ; 54.52 ± 0.14 and 54.44 ± 0.03 kcal/mole, respectively.

Numerous data on the pressure of the saturated vapor over molten NaCl have been examined by Kelley [2355]. Calculation of the heat of sublimation of NaCl, based on the pressure of the saturated vapor recommended by Kelley, gives 54.7 (1100°K) to 53.8 kcal/mole (1700°K). The later data published by Barton and Blum [669a], obtained within the range 1330-1540°K by the boiling method, gives a value of 54.0 kcal/mole.

The polymerization of the NaCl molecules in the vapor has been investigated by several authors. The data obtained by Berkowitz and Chupka [757] and Gorokhov, Khodeyev and Akishin [165, 3] are of a qualitative nature. More reliable data were obtained by Akishin, Gorokhov, and Sidorov [62] by the mass spectrometric method, and by Kusch and co-workers [2914, 1472] by the method of studying the velocity distributions of the molecules in a molecular beam. The data of these authors are in satisfactory agreement and show that at 1000°K the partial pressure of the Na₂Cl₂ dimer is about 25% of the total pressure, and that of the Na₃Cl₃ trimer, about 1% [62].

Using these ratios of the partial pressures of the monomer, dimer and trimer and the most reliable data on the pressure of the saturated vapor over solid NaCl [312, 4389, 3097], and also the thermodynamic

functions of Na_2Cl_2 (see page 1850), the authors of the Handbook calculated the heat of dissociation $\Delta H_0 = 50.5$ kcal/mole* and the equilibrium constants of the dissociation of the dimer into the monomer. According to these data, the dimer concentration in the vapor over solid NaCl increases with temperature from 23% to 700°K to 30% close to the melting point and for molten NaCl it decreases slowly (25% at 1300°K and 16% at the boiling point of 1738°K).**

Calculations of the heat of sublimation of NaCl, allowing for the complex composition of the vapor according to the data of An. N. Nesmeyanov and Sazonov [312], Zimm and Mayer [4389], and Niwa [3097] give the mean value $\Delta H_{s0} = 55.1 \pm 0.2$ kcal/mole. Analogous calculations on the basis of the pressure of the saturated vapor over molten sodium chloride [2355] gives the mean value $\Delta H_{s0} = 54.9 \pm 0.4$ kcal/mole, and on the basis of the data, found by Barton and Blum [669a] (1330-1540°K, boiling method), to the value $\Delta H_{s0} = 54.7$ kcal/mole.

The heat of sublimation of sodium chloride, based on the results of the vapor pressure measurements over solid NaCl

$$\Delta H_{s0}(\text{NaCl, cryst.}) = 55.1 \pm 0.5 \text{ kcal/mole,}$$

has been adopted in the Handbook.

To the adopted heat of sublimation correspond

$$\begin{aligned} \Delta H^\circ f_0(\text{NaCl, gas}) &= -43,345 \pm 0.55 \text{ kcal/mole,} \\ D_0(\text{NaCl}) &= 97,865 \pm 0.55 \text{ kcal/mole.} \end{aligned}$$

Beutler and Levi [804], based on an analysis of the electronic spectra of sodium chloride vapor, obtained by Levi [2596], found $D_0(\text{NaCl}) = 97.4$ kcal/mole, which is in good agreement with the value given above. However, because of the complexity of analysis of the electronic spectra of the alkali halides, the uncertainty in this value is much greater than that in the dissociation energy, calculated on the basis of the thermochemical data.

The intensities of the resonance lines of the sodium atom in the spectrum of chlorine - hydrogen flames, to which a certain quantity of sodium salts was added in the form of a solution, were measured in the work of Gurvich and Veyts [170]. By means of these data the authors of the work [170] calculated the dissociation constants of NaCl and its dissociation energy, equal to 98.5 ± 3.0 kcal/mole. Within the limits of the above-indicated error, this value also agrees with that calculated by means of the thermodynamic data.*

The standard state of potassium is K(cryst.).

K (gas). Evans et al [1513] by means of the most accurate data [1463, 3056, 2064, 2490, 1562] on the pressure of saturated potassium vapor, published prior to 1955, calculated the heat of sublimation of monatomic potassium $\Delta H_{s0} = 21.71 \pm 0.08$ kcal/g-atom. Allowance was made in the calculations for the formation of K_2 molecules, the value 11.85 ± 0.1 kcal/mole being assumed for the dissociation energy of this molecule. The recently carried out measurements of the specific heat of potassium at low temperatures (see preceding Section) made it possible to obtain more accurate values for the thermodynamic functions of solid and liquid potassium as compared with those adopted by the authors of [1513]. Introduction of the corresponding corrections gives the value

$$\Delta H_{s0}(K, \text{cryst.}) = \Delta H'_{f0}(K, \text{gas.}) = 21.59 \pm 0.1 \text{ kcal/g-atom,}$$

adopted in the Handbook.**

K⁺ (gas). The ionization potential of the potassium atom has been adopted in correspondence with the recommendation of Moore [2941] as being $35,009.78 \text{ cm}^{-1}$, or

$$I(K) = 100, 92 \text{ kcal/g-atom.}$$

The uncertainty in this value does not exceed $\pm 0.1 \text{ cm}^{-1}$ or $\pm 0.3 \text{ cal/g-atom}$. To the adopted value of the ionization potential corresponds

$$\Delta H^{\circ} f_0(\text{K}^+, \text{gas}) = 121,692 \pm 0,1 \text{ kcal/g-atom.}$$

KF (cryst.). The results of the investigations on the heat of solution of crystalline potassium fluoride in water, the heat of neutralization of caustic soda solution with hydrofluoric acid and the heat of solution of metallic potassium in water were examined critically by Bychowsky, Rossini et al [3508, 813]. On the basis of these data, Rossini et al [3508] calculated the heat of formation of crystalline potassium fluoride $\Delta H^{\circ} f_{298,16} = -134,46 \text{ kcal/mole}$. This value has been adopted in most reviews and original papers.

The introduction of a correction, taking into account the more accurate value of the heat of solution of metallic potassium in water (see Section on KCl), gives the value

$$\Delta H^{\circ} f_{298,16}(\text{KF, cryst.}) = -134,8 \pm 0,5 \text{ kcal/mole,}$$

which has been adopted in the Handbook.

KF (gas). Table 283 gives the heats of sublimation of KF, calculated on the basis of the results of the pressure measurements of the saturated vapor, carried out by different authors.

Saturated potassium fluoride vapor contains associated molecules. Eisenstadt, Rothberg and Kusch [1472] studied the velocity distribution in a molecular beam of potassium fluoride vapor and found that the partial pressure of the K_2F_2 dimer at 1000°F amounts to about 12% of the total vapor pressure. The results of the measurements of Berkowitz and Chupka [757] and Pugh and Barrow [3344] agree with these data. On the basis of the data of [1472] on the composition of potassium fluoride vapor, the heats of sublimation given in the last column of Table 283 have been calculated with allowance for the formation of the dimer.

The value

$$\Delta H_s(\text{KF, cryst.}) = 56,7 \pm 1,0 \text{ kcal/mole,}$$

which is the mean of the heats of sublimation of KF calculated on the basis of the measurements [4170, 3563, 3097, 305, 3344]* with the dimerization of the vapor of potassium fluoride being taken into account, has been adopted in the Handbook.

To this heat of sublimation correspond

$$\Delta H^{\circ} f_0(\text{KF}, \infty) = -77,741 \pm 1,1 \text{ kcal/mole,}$$

$$D_0(\text{KF}) = 117,831 \pm 1,2 \text{ kcal/mole.}$$

TABLE 283

Results of the Calculation of the Heat of Sublimation of KF

	Авторы A	Год B	Метод C	Интервал тем- пературы, °K D	Число изме- рений E	ΔH_{∞}^a , F kcal/mole	ΔH_{∞}^b , F kcal/mole
1	Вартенберг, Шульц [4170]	1921	Кипения	1624—1777	5	56,9±0,5	57,3
2	Руфф и др. [3563]	1922	а 7	1551—1773	6	56,55±0,2	56,9
3	Ниwa [3097]	1938	Эффузи- онный	913—973	5	55,1±0,05	55,3
4	Нарышкин [305]	1939	Протока	1023—1233	4	56,2±0,4	56,5
5	Пью, Барроу [3344]	1958	Эффузи- онный	902—1023		57,7±0,3	57,9
6	Эйзенштадт, Ротберг, Кусч [1472]	1958	Эффузи- онный	972—1050	6	59,0±0,1	59,2

- a) Heats of sublimation of KF, calculated without allowance for the dimerization of the vapor
- b) heats of sublimation of KF, calculated with allowance for the dimerization of the vapor
- c) variant of the effusion method ("torsion" method).
- A) Authors
- B) year
- C) method
- D) temperature range
- E) number of measurements
- F) kcal/mole
- 1) Wartenberg, Schulz
- 2) Ruff et al
- 3) Niwa
- 4) Naryshkin
- 5) Pugh, Barrow
- 6) Eisenstadt, Rothberg, Kusch
- 7) boiling
- 8) effusion
- 9) flow

The dissociation energy of KF given above practically coincides

with the value found by Page and Sugden [3164], $D_0(\text{KF}) = 117$ kcal/mole. In the work of Page and Sugden, the value $D_0(\text{KF})$ was found as a result of an investigation on the radiowave absorption in flames containing small quantities of potassium and fluorine.*

The studies of the electron spectra of the KF molecule, carried out by Barrow and Caunt [648], enabled these authors to calculate the dissociation energy of potassium fluoride $D_0(\text{KF}) = 122.6$ kcal/mole, which is greatly in excess of the value adopted in the Handbook. The error in this value exceeds considerably the error in the value calculated by means of the thermochemical data, because the spectra of the molecules of the alkali halogenides are of a diffuse nature (see page 1869).

KCl (cryst.). The generally accepted heat of formation of potassium chloride, $\Delta H^\circ_f_{298.16} = -104.175$ kcal/mole, has been calculated by Rossini et al [813, 3508] on the basis of a critical analysis of the data on the heat of solution of potassium chloride, the heat of solution of metallic potassium in water and the heat of neutralization of caustic potash solutions with hydrochloric acid.

Rossini et al [3508] adopted the value 4201 cal/mole as the heat of solution of one mole of potassium chloride in 200 moles water. A careful investigation of the heat of this reaction has been carried out by Mishchenko and Kaganovich [301] with the aim of recommending this reaction as a calorimetric standard. The value 4194 ± 3 cal/mole was found in the work [301]. Sunner and Wadso [3901] found the slightly lower value 4181 ± 3 cal/mole. The difference between the values from the works [301, 3901] and the values adopted by Rossini et al [3508] is not important for the calculation of the heat of formation of potassium chloride.

The heat of solution of metallic potassium in water and the heat

of formation of caustic potash solution, connected with it, has been subjected to substantial reappraisal. Rossini et al [3508], based on relatively older data, adopted the heat of formation of caustic potash solution (infinite dilution) $\Delta H^{\circ}_f_{298.16} = -115.0$ kcal/mole. The slightly different value -115.22 kcal/mole was found in the work of Ketchen and Wallace [2378, 2377]. Messer, Fasolino and Thalmayer [2859] carried out a new, more accurate measurement of this value and found $\Delta H^{\circ}_f_{298.16} = -115.367 \pm 0.1$ kcal/mole.*

If allowance is made for the more accurate data of [2859] on the heat of solution of metallic potassium in water, the heat of formation of potassium chloride

$$\Delta H^{\circ}_f_{298.16}(\text{KCl, cryst.}) = -104.5 \pm 0.2 \text{ kcal/mole,}$$

is obtained, which has been adopted in the Handbook.

KCl (gas). The pressure of the saturated vapor of solid and molten potassium chloride has been investigated in numerous works (see the reviews of Kelley [2355] and Pugh and Barrow [3344]). On the basis of the most reliable data, obtained for solid potassium chloride, An. Nesmeyanov and Sazonov [313] (753-897°K), Niwa [3097] (853-955°K), Bradly and Volans [288] (673-873°K), Treadwell and Werner [4016] (859-1024°K) and Pugh and Barrow [3344] (819-945°K) calculated the heats of sublimation of potassium chloride at 0°K, the average of which is 53.2 ± 0.1 kcal/mole.

Kelley [2355] examined the results of eight works on the measurement of the vapor pressure of molten potassium chloride (1043-1773°K); his calculations give the mean value: $\Delta H_{s_0} = 52.8$ kcal/mole. Calculation on the basis of the data of Barton and Blum [669a] (1270-1540°K), obtained by the boiling method, gives the same value.

According to the data of Miller and Kusch [2914] on the velocity distribution of the molecules in a molecular beam, the dimer concen-

tration in potassium chloride vapor is about 10% within the temperature range 800-1000°K. An approximate calculation with allowance being made for the dimerization of the KCl vapor gives the values $\Delta H_{s0} = 53.4$ kcal/mole on the basis of the vapor pressure measurements over solid potassium chloride, and $\Delta H_{s0} = 53.1$ kcal/mole over molten potassium chloride.

The value adopted in the Handbook

$$\Delta H_{s0}(\text{KCl, cryst.}) = 53.4 \pm 0.3 \text{ kcal/mole}$$

is based on the data on the vapor pressure over solid KCl. The great accuracy of this value is due to the reliability of the data on the pressure of saturated potassium chloride vapor over the solid salt, the absence of large concentrations of associated molecules in the vapor and the relatively great accuracy of the thermodynamic functions of solid and gaseous potassium chloride at $T \leq 1000^\circ\text{K}$.*

To the adopted value of the heat of sublimation correspond

$$\begin{aligned} \Delta H^{\circ}f_0(\text{KCl, gas}) &= -51,018 \pm 0,36 \text{ kcal/mole,} \\ D_0(\text{KCl}) &= 101,158 \pm 0,38 \text{ kcal/mole.} \end{aligned}$$

Beutler and Levi [804] and Barrow and Caunt [648], as a result of an analysis of the electron spectrum of KCl, found the dissociation energy 101.2 and 108.1 kcal/mole, respectively. Taking into account the complexity of the electron spectra of the alkali halogenides, this difference cannot be considered to be unexpected; within the limits of possible error, these values agree satisfactorily with the thermochemical data.**

The standard states of rubidium and cesium are Rb (cryst.), and Cs (cryst.).

Rb (gas), Cs (gas). Stull and Sinke [3894] re-examined the results of the measurements of the pressure of saturated rubidium vapor [2398, 3555, 3665, 1918] and cesium vapor [1918, 1624, 3949, 2490] and calcu-

lated the heats of sublimation of these elements at 298.15°K, taking into account the formation of Rb₂ and Cs₂* molecules in the vapor

$$\Delta H_{298.15}^{\circ}(\text{Rb, cryst.}) = \Delta H_{298.15}^{\circ}(\text{Rb, gas}) = 19,6 \pm 0,3 \text{ kcal/g-atom,}$$

$$\Delta H_{298.15}^{\circ}(\text{Cs, cryst.}) = \Delta H_{298.15}^{\circ}(\text{Cs, gas}) = 18,67 \pm 0,3 \text{ kcal/g-atom,}$$

which have been adopted in the present Handbook.

TABLE 284

Adopted Values (in cal/mole) of the Thermochemical Values for the Alkali Metals and Their Compounds

Вещество A	Состояние B	D ₀ , ΔH _{298.15} ⁰ или I ₀ C	ΔH ⁰ ₀	ΔH ⁰ _{298.15}	ΔH ⁰ _{298.15}	H ⁰ _{298.15} - H ⁰ ₀	H ⁰ _{298.15} - H ⁰ ₀
Na	Крист. E	25 970 ^a	0	0	0	1498	1532
Na	Газ E	—	25 970	25 928	25 919	1456	1481
Na ⁺	»	118 515 ^b	144 485	145 899	145 915	1456	1481
NaF	Крист. D	68 300 ^a	-136 040	-136 604	-136 600	1971	2027
NaF	Газ E	114 210	-69 740	-70 110	-70 123	2165	2205
NaCl	Крист.	58 100 ^a	-98 445	-98 547	-98 540	2473	2534
NaCl	Газ	97 865	-43 345	-43 667	-43 678	2253	2296
K	Крист.	21 590 ^a	0	0	0	1659	1695
K	Газ	—	21 590	21 387	21 376	1456	1481
K ⁺	»	100 102 ^b	121 692	122 945	122 959	1456	1481
KF	Крист.	56 700 ^a	-134 441	-134 804	-134 800	2333	2391
KF	Газ	117 831	-77 741	-78 228	-78 242	2209	2250
KCl	Кр. ст.	63 400 ^a	-104 418	-104 505	-104 500	2649	2710
KCl	Газ	101 158	-51 018	-51 437	-51 449	2317	2361
Rb	Крист.	19 909 ^a	0	0	0	1753	1790
Rb	Газ	—	19 909	19 612	19 600	1456	1481
Rb ⁺	»	96 331 ^b	116 240	117 399	117 412	1456	1481
Cs	Крист.	19 048 ^a	0	0	0	1822	1859
Cs	Газ	—	19 048	18 682	18 670	1456	1481
Cs ⁺	»	89 800 ^b	108 848	109 938	109 951	1456	1481

- a) The heat of sublimation is given
 b) the ionization potentials of the corresponding neutral atoms are given
 A) Substance
 B) state
 C) or
 D) cryst.
 E) gas.

Rb⁺ (gas), Cs⁺ (gas). The ionization potentials of gaseous rubidium and cesium atoms have been adopted in accordance with the recom-

mendations of Moore [2941] as 33,691.32 and 31,406.71 cm^{-1} , respectively, or

$$I(\text{Rb}) = 96,331 \text{ kcal/g-atom,}$$
$$I(\text{Cs}) = 89,800 \text{ kcal/g-atom.}$$

The uncertainties in these values do not exceed $\pm 0.1 \text{ cm}^{-1}$ or $\pm 0.3 \text{ cal/g-atom}$. To the adopted ionization potentials correspond

$$\Delta H^{\circ}_f(\text{Rb}^+, \infty) = 116,24 \pm 0,3 \text{ kcal/g-atom,}$$
$$\Delta H^{\circ}_f(\text{Cs}^+, \infty) = 108,848 \pm 0,3 \text{ kcal/g-atom.}$$

Manu-
script
Page
No.

[Footnotes]

- 1875 Errors were obviously committed in the work [3429] in the processing of the experimental data in the calculation of the values J of the edge for NaCl (125 instead of 118 were found) and an error was made in the component $2B_e - 3\alpha_1$ in the formula for $\nu_{\text{kant}} - \nu_0$. Hence, the values of $\Delta G_{1/2}$ were recalculated on the basis of the experimental data of [3429] and the adopted values of the rotational constants and were found to be 363 ± 2 and $279 \pm 6 \text{ cm}^{-1}$ for NaCl.
- 1883 Neglecting to limit the number of rotational levels of NaCl and KCl has practically no effect on the accuracy of the calculated values of the thermodynamic functions.
- 1884 Martin [2986a] calculated for the entropy of sodium the value $S^{\circ}_{298.15} = 12.24 \pm 0.12 \text{ cal/g-atom}\cdot\text{degree}$.
- 1884 There are indications (see [1013]) that Grand and Walker have carried out measurements of the enthalpy of liquid sodium within the temperature range 373-873°K but these data have not yet been published.
- 1887 There are indications [1013] that Altman, Hallet and Johnston carried out measurements on the heat capacity of NaCl within the range of 13-300°K.
- 1887 Dworkin and Bredig [1431] have recently obtained the slightly smaller value for the heat of melting of NaCl $\Delta H_{1073} = 6.69 \pm 0.13 \text{ kcal/mole}$.
- 1890 Following completion of the work on this chapter, the results

of the measurements of the heat of melting of KCl, carried out by Dworkin and Bredig [1431] were published; according to the data of these authors $\Delta H_{1013} = 6.34 \pm 0.13$ kcal/mole. There are references in this work to the value $\Delta H_{1043} = 6.27$ kcal/mole, obtained in an unpublished work of Douglas.

- 1891 Dauphinee et al [1268] found for rubidium an anomaly of the specific heat in the 150-200°K range, connected with the thermal history of the sample and of hysteresis type. In the cesium sample investigated by the authors of the work [1268], which contained 0.3% oxygen, three specific heat anomalies were established: a) a slight specific heat anomaly (about 2%) in the 100-200°K range; b) a phase transformation of the second kind in the 262.5-272°K range with a heat of transformation of 75 cal/g-atom; c) an apparent "premelting" of cesium, beginning at 285°K.
- 1892 Kirillov and Grachev [229] carried out accurate measurements of the vapor pressure of sodium above its boiling point by the static method in the range 1153-1573°K (vapor pressure 1-15 atmospheres). Calculation of the heat of sublimation of sodium on the basis of these data, taking into account the dimerization of the vapor, gives a heat of sublimation of $\Delta H_{s_0} = 25.98 \pm 0.06$ kcal/g-atom, which is in excellent agreement with that adopted in the Handbook.
- 1893 A value of 200 ± 20 cal/mole has been taken in the works [130, 129] for the heat of solution of NaF in 400 moles H₂O. Recalculation to infinite dilution gives a value which coincides with that adopted by Rossini et al [3508].
- 1896 The value $D_0(\text{NaF}) = 121 \pm 7$ kcal/mole, which differs considerably from that adopted in the Handbook, has been found in the work [1010a] by a flame photometric method.
- 1896 The heat of neutralization of strongly dilute solutions of sulfuric and hydrochloric acid with caustic soda solution has been measured in the work [3182]. The value thus obtained, $\Delta H_{298.15} = -13.50 \pm 0.05$ kcal/mole (infinite dilution) differs greatly from that adopted in the work of Rossini et al [3508] $\Delta H_{298.15} = -13.36$ kcal/mole. The value from the work [3182] is in good agreement with the results of the electrochemical measurements and is more accurate than that adopted by Rossini. The authors of the work [3182] believe that the principal cause of the difference between these values is the inaccurate extrapolation to infinite dilution of the measurement results obtained at higher concentrations [3508]. Because the heat of neutralization of solutions with relatively

high concentration enters into the thermochemical cycle used for the calculation of the heat of formation of sodium chloride, there is no need for introducing a corresponding correction into the results of the calculations [3508].

- 1896 In the work of Gunn and Green [1887], published after completion of the work on the present chapter, the value -112.48 ± 0.006 kcal/mole was obtained which is apparently no less reliable than that adopted in the present Handbook on the basis of the measurements of Messer, Fasolino and Thalmayer [2859]. The difference between these two values is only one third of the uncertainty in the adopted value of the heat of formation of NaCl.
- 1898 This value agrees with that obtained by Eisenstadt, Rothberg and Kusch [1472] on the basis of the second law of thermodynamics and the thermodynamic functions adopted in the Handbook and recalculated to 0°K.
- 1898 A comparison between the results of the measurements of the saturation pressure over molten NaCl, obtained by the flow method [670] and the boiling method [669a] and the calculation of the molecular weight of saturated sodium chloride vapor has been carried out in the report by Barton and Blum [670]. These data correspond to a very high dimer concentration in the saturated vapor (from 49 to 43% in the range 1290-1416°K) and are in contradiction with the data of other works in which the composition of saturated sodium chloride vapor had been investigated. It should be pointed out that the method of calculation of the composition of the vapor used by the authors of [670] is an indirect one; small errors in the vapor pressure measurements may lead to considerable errors in the mean molecular weight of the vapor.
- 1899 The similar value $D_0(\text{NaCl}) = 97.5 \pm 2$ kcal/mole has been obtained by the method of flame photometry in the work [1010a].
- 1899 Grachev and Kirillov [166a] measured the pressure of saturated potassium vapor by the static method in the temperature range 823-1553°K (vapor pressure $4.6 \cdot 10^{-2}$ to 23 atm.). Calculation of the heat of sublimation of potassium on the basis of these data gives a heat of sublimation of potassium of 21.6 ± 0.15 kcal/mole, which is in satisfactory agreement with the value adopted in the Handbook.
- 1901 The absolute values of the saturation pressure of KF vapor, obtained in the work of Eisenstadt, Rothberg and Kusch [1472] are inaccurate as in the case of LiF and NaF (see above).
- 1902 The coincident value $D_0(\text{KF}) = 117 \pm 8$ kcal/mole has been obtained in the work [1010a] by the method of flame photometry.

Manu-
script
Page
No.

- 1903 In the work of Gunn and Green [1887] published after completion of the work on the materials of the present chapter, the value $\Delta H^\circ_f_{298.15} = 115.323 \pm 0.010$ kcal/mole was obtained which is evidently no less reliable than that adopted in the Handbook, obtained in the work [2859]. The difference between these two values (0.044 kcal/mole) is less than the uncertainty in the Handbook values.
- 1904 Barton and Blum [670] calculated the mean molecular weight of the saturated vapor over molten potassium chloride in the range 1250-1473°K. These data attest to the fact that the concentration of dimeric molecules in potassium chloride vapor is relatively high (32-27%). However, as in the case of sodium chloride the measurement accuracy in this work is apparently not very great.
- 1904 The value $D_0(\text{KCl}) = 99.6 \pm 2$ kcal/mole was obtained in the work [1010a] by the flame photometric method.
- 1905 The authors of the work [3894] took $D_0(\text{Rb}_2) = 11.30 \pm 0.3$ and $D_0(\text{Cs}_2) = 10.38 \pm 0.3$ kcal/mole for the dissociation energies of the diatomic rubidium and cesium molecules in accordance with the recommendations of [2020, 1513].

Chapter 30

ZIRCONIUM, LEAD, MERCURY AND THEIR COMPOUNDS

(Zr, ZrO, ZrO₂, Pb, PbO, PbF, PbF₂, Hg, HgF)

The data necessary for the calculation of the thermodynamic quantities of mercury, zirconium, lead and some of their compounds with oxygen and fluorine are examined in the present chapter. In addition to elemental mercury, one of its compounds, HgF, will also be considered. The absence of data on HgO in the Handbook is explained by the fact that this compound is almost completely decomposed into its elements in the gas phase. Mercuric oxide is not even very stable in the condensed state: at a temperature of only 750°K, the vapor pressure of the dissociation products is 1 atm. [917].

The data presented in the Handbook on Zr, ZrO and ZrO₂ provide a fairly accurate idea of the thermodynamic properties of the Zr - O system.

In addition to the PbO, the simplest lead oxide which is given in the Handbook, Pb₃O₄, Pb₂O₃ and PbO₂ are also known in the condensed state; however, heating causes these compounds to be decomposed into oxygen and PbO. Only the lead monoxide PbO is stable in the gas state. Thus, the thermodynamic properties of the Pb - O system at high temperatures can be represented sufficiently accurately by the data in the Handbook. At lower temperatures, however (approximately below 550°C, the temperature at which Pb₃O₄ is transformed into PbO), the formation of the higher oxides of lead in the condensed state must be taken into account.

The compounds PbF and PbF_2 which are treated in the Handbook, represent the properties of the Pb - F system sufficiently accurately because lead tetrafluoride PbF_4 is decomposed at higher temperatures to PbF_2 and fluorine.

§119. MOLECULAR CONSTANTS

Zr. In the ground state the zirconium atom has the electronic configuration $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 4d^2 5s^2$ to which correspond the terms $^3F, ^3P, ^1D, ^1G, ^1S$.

Excitation of one of the $5s^2$ electrons causes the appearance of the group of terms $\dots 4d^2 5s(^4F)nl$, whose series limit is situated $56,961 \text{ cm}^{-1}$ above the ground state. This group consists of triplet and quintet levels with the values $L = l - 3, l - 2, \dots, l + 3$.

Simultaneous excitation of two $5s$ electrons and also of other electrons of the Zr atom causes the appearance of a large number of term groups. Table 285 gives the groups of terms which have been used in the subsequent calculations, with indication of their series limits and multiplicity. As the Table shows, these groups appear upon excitation of one or two $5s$ electrons. The other groups of terms have not been taken into account because their levels have excitation energies of the order of $70,000 \text{ cm}^{-1}$ and over.

Table 286 gives the energy levels of the zirconium atom, corresponding to the above-listed term groups; their excitation energies have been chosen on the basis of the values recommended by Moore [2941]. The levels with similar excitation energies were combined into a single level with a summary statistical weight and mean excitation energy.

The summary [2941] gives data only for a small number of terms of these groups. These are mainly the terms with the electronic configurations $\dots 4d^2 5s 5p$ and $\dots 4d^3 5s$. All terms which have been observed experimentally in the spectra of the zirconium atom, were assigned in

accordance with the adopted rules of evaluation to the series limits of their groups. An exception has been made for the terms corresponding to the configuration $\dots 4d^2 5p^2$, to which the energy of the term $\dots 4d^2 5p^2 \ ^5G$ has been ascribed, and for the states with the electronic configurations $\dots 4d^4$ which were assigned to a series of levels in correspondence with the arrangement of the analogous states in ions which are isoelectronic with the zirconium atom. Simple assignment of the absent terms to a single known level or to an ionization limit in this case results in large errors because the states corresponding to the $\dots 4d^4$ configuration have relatively low excitation energies.

TABLE 285

Term Groups of the Zr Atom Considered in the Handbook

Группа A	B Терм	Мультиплетность C	D Серияльная граница, см ⁻¹
$\dots 4d^2 5s \ (^4F) nI$	$ L-3 , \dots, L+3 $	Триплет, квинтет	E 56 961
$\dots 4d^2 \ (^4F) nI^I$	$ L-3 , \dots, L+3 $	• •	F 59 350
$\dots 4d^2 5s \ (^3D) nI^{II}$	$ L-2 , \dots, L+2 $	Синглет, триплет	F 60 479
$\dots 4d^2 5s \ (^3P) nI^{III}$	$ L-1 , L, L+1 $	• •	F 62 060
$\dots 4d^2 5s \ (^3F) nI^{IV}$	$ L-3 , \dots, L+3 $	• •	F 62 187
$\dots 4d^2 5s \ (^3P) nI^{V}$	$ L-1 , L, L+1 $	Триплет, квинтет	E 63 937
$\dots 4d^2 \ (^3G) nI^{VI}$	$ L-4 , \dots, L+4 $	Синглет, триплет	F 64 000
$\dots 4d^2 \ (^3P) nI^{VII}$	$ L-1 , L, L+1 $	Триплет, квинтет	E 65 901
$\dots 4d^2 \ (^3H) nI^{VIII}$	$ L-5 , \dots, L+5 $	Синглет, триплет	F 68 266

A) Group; B) term; C) multiplicity; D) series limit, cm⁻¹; E) triplet, quintet; F) singlet, triplet.

TABLE 286

Energy Levels of the Zirconium Atom

Номер уровня А	Состояние		Статисти- ческий вес	Энергия, см ⁻¹
	электронная конфигура- ция	терм D		
0	...4d ⁵ 5s ²	² F _{7/2}	5	0
1	...4d ⁵ 5s ²	² F _{5/2}	7	570,41
2	...4d ⁵ 5s ²	² F _{3/2}	9	1240,84
3	...4d ⁵ 5s ²	² P _{3/2} , ² P _{1/2}	6	4187,90
4	...4d ⁵ 5s ²	² P _{1/2}	3	4376,28
5	...4d ⁵ (4F) 5s	² F _{7/2}	3	4870,53
6	...4d ⁵ (4F) 5s	² F _{5/2}	5	5023,41
7	...4d ⁵ 5s ²	¹ D	5	5101,68
8	...4d ⁵ (4F) 5s	² F _{3/2}	7	5249,07
9	...4d ⁵ (4F) 5s	² F _{1/2}	9	5540,54
10	...4d ⁵ (4F) 5s	² F _{3/2}	11	5888,93
11	...4d ⁵ 5s ²	¹ G ₄	9	8057,30
12	...4d ⁵ (4P) 5s	² P _{1/2}	3	10885
13	...4d ⁵ (4P) 5s	² P _{3/2}	5	11017
14	...4d ⁵ (4P) 5s	² P _{3/2}	7	11258
15	...4d ⁵ (4F) 5s	² F _{5/2}	5	11641
16	...4d ⁵ (4F) 5s	² F _{3/2}	7	11956
17	...4d ⁵ (4F) 5s	² F _{1/2}	9	12342
18	...4d ⁵ (4G) 5s	² G ₃	7	12503
19	...4d ⁵ (4G) 5s	² G ₄ , ² G ₆	20	12768
20	...4d ⁵ 5s ²	¹ S	1	13142
21	...4d ⁵ (4D) 5s	² D ₁	3	14123
22	...4d ⁵ (4D) 5s	² D ₃	5	14349
23	...4d ⁵ (4D) 5s	² D ₅	7	14697
24	...4d ⁵ 5s (4F) 5p ...4d ⁵ (4H) 5s	² G ₄ ² H ₄	14	14789
25	...4d ⁵ (4H) 5s	² H ₆	11	14999

Номер уровня A	Состояние B		Статисти- ческий вес E	Энергия, ед. ⁻¹ F
	C электронная конфигурация	D терм.		
26	...4d ⁹ (H) 5s ...4d ⁹ (F) 5s	³ H ₀ ³ F ₀	18	15127
27	...4d ⁹ 5s (F) 5p	³ G ₀	7	15201
28	...4d ⁹ (F) 5s	³ F ₀	7	15457
29	...4d ⁹ (F) 5s ...4d ⁹ 5s (F) 5p	³ F ₄ ³ G ₂	18	15710
30	...4d ⁹ (P) 5s	³ P	9	15932
31	...4d ⁹ 5s (F) 5p	³ G ₀ , ³ F ₂	16	16311
32	...4d ⁹ 5s (F) 5p	³ F ₁	3	16787
33	...4d ⁹ 5s (F) 5p	³ F ₀	7	16844
34	...4d ⁹ 5s (F) 5p ...4d ⁹ (P) 5s	³ G ₀ , ³ F ₂ ³ P ₁	21	17000
35	...4d ⁹ (P) 5s	³ P ₂	5	17143
36	...4d ⁹ (P) 5s ...4d ⁹ (D) 5s	³ P ₀ ³ D ₂	6	17244
37	...4d ⁹ 5s (F) 5p	³ F ₀ , ³ D ₁	10	17424
38	...4d ⁹ 5s (D) 5p ...4d ⁹ 5s (F) 5p	³ D ³ F ₄	14	17542
39	...4d ⁹ (G) 5s	³ G ₄	9	17753
40	...4d ⁹ 5s (F) 5p	³ F ₄ , ³ D ₂	14	17828
41	...4d ⁹ 5s (F) 5p	³ F ₂ , ³ D ₀	18	18284
42	...4d ⁹ (H) 5s	³ H	11	18730
43	...4d ⁹ 5s (F) 5p	³ D _{0,1}	4	19087
44	...4d ⁹ 5s (F) 5p	³ D ₁	5	19324
45	...4d ⁹ 5s (F) 5p	³ D ₂	7	19628
46	...4d ⁹ 5s (F) 5p	³ D ₃	9	19834
47	...4d ⁹ 5s (P) 5p	³ P	9	20458
48	...4d ⁹ ...4d ⁹ 5s (F) 5p ...4d ⁹ 5s (P) 5p	³ D _{0,1,2} ³ G ₀ ³ S ₁	19	21880
49	...4d ⁹ 5s (F) 5p ...4d ⁹	³ G ₄ ³ D ₀	16	22144
50	...4d ₀	³ D ₄	9	22398
51	...4d ⁹ 5s (F) 5p	³ G ₀	11	22564
52	...4d ⁹ 5s (P) 5p ...4d ⁹ 5s (D) 5p	³ D ³ F	12	22816

Номер уровня A	Состояние B		E Статисти- ческий вес	F Энергия, см ⁻¹
	электронная конфигура- ция C	терм D		
53	...4d ⁵ 5s (4P) 5p	² D ₁ , ¹ S ₁ , ⁴ D ₀	9	23067
54	...4d ⁵ 5s (4P) 5p	² D ₁ , ⁴ D ₁	8	23292
55	...4d ⁵ 5s (4P) 5p ...4d ⁵ (4F) 5p	¹ D ₂ , ¹ D ₃ ³ F _{2,3}	24	23585
56	...4d ⁵ 5s (4P) 5p	¹ D ₂	7	23889
57	...4d ⁵ (4F) 5p	³ F ₄	9	24006
58	...4d ⁵ 5s (4P) 5p ...4d ⁵ 5s (4F) 5p	¹ D ₂ ¹ F	16	24381
59	...4d ⁵ 5s (4P) 5p ...4d ⁵ (4F) 5p	³ P _{1,2} ³ G ₂ , ³ G ₃	20	25648
60	...4d ⁵ 5s (4P) 5p ...4d ⁵ (4F) 5p	¹ P ₁ ¹ G ₂	14	25935
61	...4d ⁵ (4F) 5p ...4d ⁵ 5s (3D) 5p ...4d ⁵ 5s (3P) 5p ...4d ⁵ (3D) 5p	¹ G ₂ ³ F ₂ ³ D ₁ ¹ F	24	26103
62	...4d ⁵ (4F) 5p ...4d ⁵ 5s (3P) 5p ...4d ⁵ 5s (3D) 5p	¹ G ₂ , ¹ G ₃ ³ D ₂ ³ F ₂	32	26430
63	...4d ⁵ (4F) 5p	¹ G ₂	11	26766
64	...4d ⁵ (4F) 5p ...4d ⁵ 5s (4F) 5p ...4d ⁵ 5s (3D) 5p	³ D ₁ ¹ G ₂ ³ F ₄	21	26930
65	...4d ⁵ (4F) 5p ...4d ⁵ 5s (3P) 5p	¹ G ₂ , ¹ D ₂ ¹ D ₃	25	27167
66	...4d ⁵ (4F) 5p ...4d ⁵ (3D) 5p ...4d ⁵ 5s (4P) 5p	³ D ₂ ³ D ₃ ³ P	21	27554
67	...4d ⁵ 5s (4F) 5p ...4d ⁵ (3G) 5p	³ F ₂ ³ H ₄	14	27897
68	...4d ⁵ 5s (4F) 5p ...4d ⁵ ...4d ⁵ (3G) 5p	³ F ₂ ³ P ³ H ₄	27	28191
69	...4d ⁵ 5s (4F) 5p ...4d ⁵ (3G) 5p ...4d ⁵ (4F) 5p ...4d ⁵ 5s (3D) 5p	¹ G ₂ , ³ F ₄ ³ H ₄ ³ F ₁ ³ P ₀	33	28538

Номер уровня A	Состояние B		Статисти- ческий вес	Энергия, см ⁻¹
	электронная конфигура- ция C	терм D		
70	...4d ⁵ 5s (³ D) 5p ...4d ⁵ 5s (³ F) 5p ...4d ⁵ (⁴ F) 5p	³ P _{1,3} , ³ D ₁ ³ G ₄ ³ F ₃	27	28798
71	...4d ⁵ (³ F) 5p ...4d ⁵ 5s (³ F) 5p ...4d ⁵ 5s (³ D) 5p	³ F ₄ ³ G ₃ ³ P, ³ D ₁	28	29050
72	...4d ⁵ 5s (³ D) 5p ...4d ⁵ (⁴ F) 5p	³ D ₃ ³ F ₆ , ³ D ₀	19	29442
73	...4d ⁵ (⁴ F) 5p	³ D _{1,3}	8	29784
74	...4d ⁵ (⁴ F) 5p	³ D _{3,4}	16	30255
75	...4d ⁵ (³ G) 5p ...4d ⁵ 5s (⁴ P) 5p ...4d ⁵ (³ F) 5p	³ G ³ S ₁ ³ G _{3,4}	28	31412
76	...4d ⁵ (³ F) 5p ...4d ⁵ ...4d ⁵ 5s (³ P) 5p	³ G ₃ , ³ F ₃ ³ H ^o ³ P	52	32458
77	...4d ⁵ (³ F) 5p ...4d ⁵ (⁴ P) 5p ...4d ⁵ ...4d ⁵ (³ D) 5p ...4d ⁵ 5s (³ F) 5p	³ F ₃ ³ S ₁ , ³ D _{0,1} ³ G ^o ³ F ³ D ₁	65	33350
78	...4d ⁵ 5s (³ F) 5p ...4d ⁵ (³ F) 5p ...4d ⁵ (⁴ P) 5p ...4d ⁵ ...4d ⁵ 5s (³ G) 5p	³ D ₃ ³ F ₄ ³ D _{3,3} ³ D ^o , ³ D ^o , ³ F ^o ³ H	78	33778
79	...4d ⁵ (⁴ P) 5p ...4d ⁵ 5s (³ F) 5p ...4d ⁵ (³ H) 5p	³ D ₄ ³ D ₂ ³ H ₄	25	34333
80	...4d ⁵ (³ H) 5p ...4d ⁵ (⁴ P) 5p ...4d ⁵ 5s (³ D) 5p	³ H ₃ ³ P _{1,3} ³ D	24	34737
81	...4d ⁵ (³ H) 5p ...4d ⁵ (⁴ P) 5p ...4d ⁵ 5s (⁴ F) 6s	³ H ₃ ³ P ₂ , ³ P _{1,3} ³ F _{1,3,3}	43	35228
82	...4d ⁵ 5s (⁴ F) 6s ...4d ⁵ 5s (³ G) 5p ...4d ⁵ (³ H) 5p ...4d ⁵ (⁴ P) 5p	³ F ₄ ³ F _{2,3} ³ J ₃ ³ S ₂	37	35798

Номер уровня A	Состояние B		E Статисти- ческий вес	F Энергия, см ⁻¹
	электронная конфигура- ция C	D терм		
83	...4d ⁹ (⁴ P) 5p ...4d ⁹ 5s (² G) 5p ...4d ⁹ (² H) 5p ...4d ⁹ (² D) 5p ...4d ⁹ 5s (² F) 5p	³ D ² F _{3,2} , ¹ G ₄ ² F _{3,2} ² F ₃ ² F ₃	73	36205
84	...4d ⁹ (² D) 5p ...4d ⁹ 5s (² D) 5p ...4d ⁹ 5s (² G) 5p ...4d ⁹ (² G) 5p	² P ₁ ² P _{0,1} , ¹ F ² H ² G ₃	54	36737
85	...4d ⁹ (² D) 5p ...4d ⁹ 5s (² D) 5p ...4d ⁹ ...4d ⁹ (² G) 5p ...4d ⁹ 5s (⁴ F) 6s	² P ₂ ² P ₂ ¹ G ^o , ¹ I ^o ² G _{1,3} , ² F ² F _{2,3}	85	37451
86	...4d ⁹ 5s (⁴ F) 6s ...4d ⁹ (² F) 5p ...4d ⁹ (² H) 5p ...4d ⁹ 5s (² D) 5p	² F ₄ ² D ¹ I ² F _{2,3}	49	38440
87	...4d ⁹ 5s (² D) 5p ...4d ⁹ 5s (² G) 5p ...4d ⁹ ...4d ⁹ (² D) 5p ...4d ⁹ (² G) 5p ...4d ⁹ 5s (⁴ F) 5d	² F ₄ ² G _{3,4} ¹ S ^o ¹ P, ² D _{1,3} ¹ F, ¹ H ² H _{3,4}	71	39728
88	...4d ⁹ 5s (² G) 5p ...4d ⁹ (² D) 5p ...4d ⁹ ...4d ⁹ 5s (⁴ F) 5d ...4d ⁹ 5s (² D) 5p ...4d ⁹ (² F) 5p ...4d ⁹ (⁴ F) 6s	² G ₃ ² D ₃ ¹ F ^o ² G _{2,3} , ² H ₃ ² P _{0,1} , ¹ P ¹ D ² F _{1,2,3,4}	84	40615
89	...4d ⁹ (⁴ F) 6s ...4d ⁹ 5s (⁴ F) 5d ...4d ⁹ 5s (² D) 5p ...4d ⁹ (² H) 5p	² F ₄ ² H ₃ , ² G _{3,4} ² P ₂ ¹ G	71	41486
90	...4d ⁹ (² H) 5p ...4d ⁹ ...4d ⁹ 5s (⁴ F) 5d ...4d ⁹ 5s (² D) 5p	² G, ¹ H ² F ^o ² H ₁ ² F ₂ , ² D	84	42401

Номер уровня A	Состояние B		E Статисти- ческий вес	F Энергия, см ⁻¹
	электронная конфигура- ция C	терм D		
91	...4d ⁵ s ² (³ D) 5p ...4d ⁶ (¹ P) 5p	³ F _{3,2} ³ S ₁ , ³ P, ³ D _{1,3}	36	44185
92	...4d ⁶ (¹ P) 5p ...4d ⁵ 5p ² ...4d ⁶ (¹ F) 5p ...4d ⁵ s ² (³ D) 5p	³ D ₃ ¹ G ¹ F ³ D ₁	62	46746
93	...4d ⁵ 5p ²	¹ S ^o (2), ¹ S ^o , ¹ P ^o (2) ³ P ^o (5), ³ P ^o , ¹ D ^o (6) ³ D ^o (3), ³ D ^o (3), ¹ F ^o (3) ³ F ^o (5), ³ F ^o , ¹ G ^o (4), ³ G ^o (2) ³ H ^o , ³ H ^o (2)	526	47700
94	...4d ⁵ s ² (³ D) 5p ...4d ⁶ ...4d ⁵ s ² (³ S) 5p	³ D _{2,3} ³ S ^o , ¹ D ^o , ¹ G ^o , ³ P ^o ¹ P	39	49157
95	...4d ⁵ s ² (⁴ F) nI	..	26668	56961
96	...4d ⁶ (⁴ F) nII	..	26624	59350
97	...4d ⁵ s ² (³ D) nIII	..	9590	60479
98	...4d ⁵ s ² (³ P) nIII	..	5748	62080
99	...4d ⁵ s ² (³ F) nI ^{IV}	..	13412	62187
100	...4d ⁵ s ² (⁴ P) nI ^V	..	11496	63937
101	...4d ⁶ (³ G) nI ^{VI}	..	17244	64090
102	...4d ⁶ (⁴ P) nI ^{VII}	..	11496	65901
103	...4d ⁶ (³ H) nI ^{VIII}	..	21076	68266

* This state has not been observed experimentally.

** All states with this electron configuration corresponding to $n \leq 11$ and not taken into account in the preceding levels.

A) Number of level; B) state; C) electron configuration; D) term; E) statistical weight; F) energy, cm⁻¹.

The terms of the Zr atom which are absent in the summary of Moore [2941] are marked by an asterisk in Table 286. It should be pointed out that the maximum uncertainty in the estimation of the energies of these groups varies from 4000 (for the first group) to 16,000 cm⁻¹ (for the last group) at excitation energies of these levels of 57,000 to 68,000 cm⁻¹. The maximum uncertainty in the estimation of the terms, corresponding to the ...4d²5p² electron configurations does not exceed

8000 cm^{-1} at a minimum energy of the term of about 48,000 cm^{-1} .

Most important from the point of view of the subsequent calculations is the accuracy of estimation of the energies of the states with the $\dots 4d^4$ electron configuration because these states have low excitation energies. The maximum uncertainty in this case evidently does not exceed 10,000 cm^{-1} at energies of the order of 22,500 cm^{-1} .

Pb. In the ground state the lead atom has the electron configuration $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 4d^{10} 4f^{14} 5s^2 5p^6 5d^{10} 6s^2 6p^2$, to which correspond the three terms 3P , 2D and 1S . Excitation of one 6p electron gives rise to the group of terms $\dots 6s^2 6p(2P)n\ell$, whose limit is situated 59,821.0 cm^{-1} above the lower 3P_0 state. This group consists of singlet and triplet terms with the values $L = \ell$ at $\ell = 0$ and $L = \ell; \ell \pm 1$ at $\ell > 0$. Owing to the strong splitting of the substates $6s^2 6p^2 P_{1/2}$ and $6s^2 6p^2 P_{3/2}$ of the Pb^+ ion (about 14,000 cm^{-1}), the substates of the triplet terms of the Pb atom with the greatest value of J (for example, the 3P_2 substate) are situated much higher than the other components of the same terms. Table 287 gives the levels of the Pb atom with excitation energies up to 50,000 cm^{-1} , adopted from Moore [2941]. It should be pointed out that most of the levels of lead, even those belonging to the $\dots 6s^2 6p(2P)n\ell$ group have not yet been experimentally observed. As regards the terms of the other series, in particular of other groups, connected with the excitation of a 6p electron and the states corresponding to the electron configuration $\dots 6s 6p^3$, these are quite unknown. Because transitions have been observed in the Pb spectra only between a small number of states, the estimation of the excitation energies of the unknown terms is difficult and has not been carried out in the present Handbook.

Hg. The atom of mercury in the ground state 1S has the electronic configuration $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 4d^{10} 4f^{14} 5s^2 5p^6 5d^{10} 6s^2$. Excitation

of an electron of the mercury atom causes the appearance of a series of $\dots 6s(2S)n\ell$ terms, whose limit is situated $84,184 \text{ cm}^{-1}$ above the ground state. This series consists of singlet and triplet terms with the values $L = \ell$. The terms of the series $\dots 6s(2S)n\ell$, with the exception of the $\dots 6s(2S)6p^3P$ term, have excitation energies over $50,000 \text{ cm}^{-1}$. Even larger excitation energies (over $68,000 \text{ cm}^{-1}$) have the terms connected with the excitation of a 5d electron. Hence Table 288 gives data only for two states of the Hg atom, the terms $\dots 6s^2 1S$ and $6s(2S)6p^3P$, adopted from Moore [2941].

TABLE 287

Energy Levels of the Lead Atom

Номер уровня A	Состояние B		Статисти- ческий вес E	Энергия, см ⁻¹ F
	электронная конфи- гурация C	терм D		
0	$\dots 6s^2 6p^3$	3P_0	1	0
1	$\dots 6s^2 6p^3$	3P_1	3	7819,35
2	$\dots 6s^2 6p^3$	3P_2	5	10850,47
3	$\dots 6s^2 6p^3$	1D	5	21457,9
4	$\dots 6s^2 6p^3$	1S	1	29466,8
5	$\dots 6s^2 6p(^3P_{1/2}) 7s$	$^3P_0, ^3P_1$	4	35205
6	$\dots 6s^2 6p(^3P_{1/2}) 7p$	3P_1	3	42918,7
7	$\dots 6s^2 6p(^3P_{1/2}) 7p$	3P_0	9	44719
	$\dots 6s^2 6p(^3P_{1/2}) 7f$	$^3D_1, ^3D_2$		
8	$\dots 6s^2 6p(^3P_{1/2}) 6d$	$^3F_2, ^3F_3$	20	48000
	$\dots 6s^2 6p(^3P_{1/2}) 6d$	$^3D_1, ^3D_1$		
9	$\dots 6s^2 6p(^3P_{1/2}) 7s$	$^3P_1, ^1P$	12	48671
	$\dots 6s^2 6p(^3P_{1/2}) 8s$	$^3P_0, ^3P_1$		

A) Number of level; B) state; C) electron configuration; D) term; E) statistical weight; F) energy, cm^{-1} .

ZrO. The spectrum of ZrO has been studied by many authors (see [649, 2020]). In recent years, the spectrum of ZrO has been investigated in the works of Afaf [493], Herbig [1995], Uhler [4035], Lager-

qvist, Barrow and Uhler [2536], Uhler and Akerlind [4036] and Akerlind [495] in emission as well as in absorption, obtained during studies of star spectra [1995]. However, in spite of the numerous works, the information on the molecular constants of ZrO is incomplete which is due to the complex structure of this molecule and the overlapping of bands which belong to different systems.

TABLE 288

Energy Levels of the Mercury Atom

Номер уровня A	Состояние B		Статисти- ческий вес E	Энергия, см ⁻¹ F
	электронная кон- фигурация C	терм D		
0	...6s ²	¹ S	1	0
1	...6s (¹ S) 6p	³ P ₀	1	37845,06
2	...6s (¹ S) 6p	³ P ₁	3	39412,3
3	...6s (¹ S) 6p	³ P ₂	5	44043

A) Number of level; B) state; C) electron configuration; D) term; E) statistical weight; F) energy, cm⁻¹.

Five band systems have been analyzed in the ZrO spectrum to date: three triplets (α , β and γ) and two singlets (A and B). Intercombination transitions have not been observed in the spectra of ZrO and the intensities of the singlet and triplet systems of bands in the absorption spectrum are of similar magnitude. Hence the experimental data do not permit determination of the type of ground state of the ZrO molecule and the relative position of its singlet and triplet states. The correlation rules does not provide an unequivocal answer to this question because the ground state of ZrO can be a singlet or triplet state depending on which electrons of the Zr atom participate in bond formation.

Afa [493], based on the fact that the bands of one of the triplet systems of ZrO (α system) have a slightly greater intensity in the

absorption spectrum than the bands of the singlet systems, arrived at the conclusion that the lower state of the α system is the electronic ground state of the ZrO molecule. The authors of later works [4035, 2536, 4036, 495] adopted this hypothesis of Afaf which appears to be confirmed by the fact that the dissociation energy of ZrO in the lower triplet state is much greater than in the lower singlet state. However, new investigations of the spectra of ZrO will possibly modify the current ideas on the systematics of the electronic states of this molecule.

Firstly, the molecular constants of ZrO in the four triplet states, connected with the α , β and γ band systems were determined by Lowater [2666, 2667] who showed that all three systems have a common ground state, but the analysis of the rotational structure of the bands in these works and also in the work of Tanaka and Horie [3922] was not carried out correctly.

Afaf [493] carried out the most complete investigation on the ZrO spectrum, obtaining a large number of band systems of this molecule in the region of 2500-9000 A on high-dispersion apparatus. On the basis of an analysis using the band edges of the vibrational structure of three triplet and two singlet systems, Afaf calculated the vibrational constants of ZrO in several states.

An analysis of the rotational structure of the 0-0 bands in the α , β and γ systems of ZrO was carried out in the works of Lagerqvist, Uhler and Barrow [2536] and Uhler [4035]. The authors of the works [2536, 4035] showed that these systems belong to the transitions $A^3\Phi - X^3\Delta$, $B^3\Pi - X^3\Delta$ and $C^3\Delta - X^3\Delta$ and determined the rotational constants B_0 and D_0 in these states, using the relations proposed in the work of Budo for the $^3\Pi$ states (see page 80) for the energies of the rotational levels of ZrO in the states $^3\Delta$ and $^3\Phi$.

Because the analysis of the rotational structure of the triplet systems of ZrO in the works [2536, 4035] was carried out only for the 0-0 bands, the authors of these works determined the values of the constant α_1 by means of the relation (1.38). The rotational constants of ZrO in the four singlet states were investigated in the works of Uhler and Akerlind [4036] and Akerlind [495]. The authors of the works [495, 4036] carried out an analysis of the 0-0, 0-1, 1-0 and 2-1 bands of the system A and the 0-0 and 0-1 bands of the system B. It was shown on the basis of the data thus obtained that these systems do not possess common states as proposed by Afaf; and the rotational constants of ZrO in the singlet states and also the vibrational constants in the upper $b^1\Sigma$ state of the system A were determined. Uhler and Akerlind, based on the nature of the fine structure of the bands and the absence of Q-branches arrived at the conclusion that both band systems belong to $^1\Sigma - ^1\Sigma$ or $^1\Delta - ^1\Delta$ transitions. Because the state $^1\Delta$ is the lower singlet state of the analogous TiO molecule, Akerlind [495] suggested that the lower state of the system A should also be a $^1\Delta$ state and that this system belongs to the $b^1\Delta - a^1\Delta$, transition.

The adopted values of the molecular constants of ZrO are given in Table 289. The vibrational constants in the States $X^3\Delta$, $A^3\Delta$, $B^3\Pi$, $C^3\Phi$, $a^1\Delta$ and $c^1\Sigma$ have been taken from the data of Afaf [493] and the rotation-vibration interaction constants in the states $X^3\Delta$, $A^3\Delta$ and $C^3\Phi$ were calculated by means of formula (1.38) on the basis of the adopted values of ω_e and $\omega_e x_e$ in these states and the value of B_0 , determined in the works [2536, 4035]. The rotational constants of ZrO in the states $a^1\Delta$, $b^1\Delta$, $c^1\Sigma$ and $d^1\Sigma$ were adopted from the data of Uhler and Akerlind [4036] and Akerlind [495], and the vibrational constants in the state $d^1\Sigma$ from the work of Akerlind [495]. It should be pointed out that the relative positions of the states $^1\Sigma$ and $^1\Delta$ are unknown be-

cause two singlet band systems are observed in the spectrum of ZrO which do not have common states. The excitation energy of the $a^1\Delta$ state of the ZrO molecule has been adopted on the basis of an estimate by Afaf.

PbO. The molecule of lead monoxide, like the diatomic molecules of carbon and silicon monoxide have the ground state $^1\Sigma$. Five band systems have been found so far in the emission and absorption spectra of PbO, situated at 2600 to 6000 Å and connected with transitions between the ground state and five excited states. Although the bands of all these systems have a simple form, the overlapping of the bands and the superposition of the spectra of the three isotope modifications of PbO make an analysis of their rotational structure and the determination of the types of higher states of the corresponding transitions difficult. The only investigation of the fine structure of the PbO bands is the work of Christy and Bloomenthal [1104] who carried out an analysis of the structure of three bands (0-2, 0-3 and 0-5) in the system $A \rightarrow X^1\Sigma$ and one band (0-2) of the system $D \rightarrow X^1\Sigma$, obtained in emission on an apparatus with a dispersion of 1.3 Å/mm. The rotational structure of the bands consisted of single lines of the P and R branches so that the authors of the work [1104] arrived at the conclusion that the two band systems are connected with a transition between singlet states having the same value of Λ , i.e., with the $^1\Sigma - ^1\Sigma$ transitions. On the basis of the data obtained by the authors of the work [1104], the rotational constants of PbO in the state $X^1\Sigma$ and that of the constant B_0 in the states $A^1\Sigma$ and $D^1\Sigma$ were found.

An analysis of the vibrational structure of the PbO spectrum was first carried out by Mecke [2829] by means of the band edges of the systems $A^1\Sigma - X^1\Sigma$ and $B^1\Sigma - X^1\Sigma$. Bloomenthal [844] improved the data of Mecke [2829] as a result of a study of the emission spectrum of

PbO over a wider wavelength region. Shawhan and Morgan [3696], Howell [2136] and Vago and Barrow [4045] investigated the absorption spectrum of lead monoxide, found two new band systems in the ultraviolet region of the spectrum and carried out an analysis of their vibrational structure. It should be pointed out that the vibrational constants of PbO in the states $X^1\Sigma$, $B^1\Sigma$ and $D^1\Sigma$, found in the works [844, 2136, 3696] agree to within 0.1 cm^{-1} . Table 289 gives the molecular constants of PbO, adopted in the present Handbook. The constants in the states $X^1\Sigma$, $B^1\Sigma$, $C^1\Sigma$ and $D^1\Sigma$ were taken from the data of Bloomenthal [844] and in the states $E^1\Sigma$ from the work of Vago and Barrow [4045]. It should be mentioned that the molecule PbO should have, in addition to the singlet states, triplet states which have not yet been observed in the spectra of lead monoxide and for which it can be assumed in analogy with CO, that the lower triplet state has a lower excitation energy than the known state $A^1\Sigma$.

PbF. The spectrum of lead monofluoride has first been studied by Morgan [2947] and Rochester [3463] who obtained the absorption and emission spectrum of this molecule in the region of 4100-5300 Å. The authors of the works [2947, 3463] interpreted the observed band system as being connected with transitions between one component of the ground state $X^2\Pi$ and the excited state of PbF and determined, on the basis of the band edges, the vibrational constants in both states. The constants found in these works are in good agreement.

In 1938, Rochester [3464] again investigated the absorption and emission spectrum of PbF in the region 2300-6000 Å on high-dispersion apparatus. One band system was observed in the emission spectrum which has been interpreted by the authors [3464] as corresponding to a transition from the B state of PbF to the upper component of the ground state $X^2\Pi$. Five bands, corresponding to transitions from the lower com-

ponent of the state $X^2\Pi$ to the excited states B, C, D, E and F, were obtained in the absorption spectrum. Three bands of the most intense system ($B - X^2\Pi_{1/2}$) were also observed in emission. By means of the band edges Rochester [3464] carried out an analysis of six systems and determined the vibrational constants of several states. The values found by him are in good agreement with those obtained in previous works and are given in Table 289.

TABLE 289

Adopted Values of the Molecular Constants of ZrO, PbO, PbF and HgF

Молекула A	Состояние B	T_e	ω_e	$\omega_e x_e$	B_e	$\alpha_1 \cdot 10^2$	$D_e \cdot 10^4$	r_e
		cm ⁻¹ C						
ZrO	$X^2\Delta^o$	0	937,20	3,35	0,4147 ^b	0,21 ^b	0,33	1,728
	$a^1\Delta^o$	1300 ^a	978,07	5,04	0,4241	0,23	0,33	1,711
	$A^2\Phi$	16741 ^a	856,97	3,37	0,4035 ^b	0,21 ^b	0,33	1,752
	$B^2\Pi$	17745 ^b	846,28	3,41	—	—	—	—
	$C^2\Delta$	21548	820,58	3,31	0,3927 ^b	0,21 ^b	0,28	1,775
	$D^1\Delta$	23444 ^a	843,27	3,04	0,3951	0,19	0,35	1,772
	$c^1\Sigma$	x	—	—	0,4167 ^b	0,12	0,35	1,725
	$d^1\Sigma$	x+19272	—	—	0,3988	0,21	—	1,764
PbO	$X^1\Sigma$	0	721,8	3,70	0,3073 ^a	0,19	0,22	1,922
	$A^1\Sigma$	19863,3	451,7	3,33	0,2579 ^b	—	0,27	2,098
	$B^1\Sigma$	22284,9	498,0	2,20	—	—	—	—
	$C^1\Sigma$	24864,0	518,0	3,90	—	—	—	—
	$D^1\Sigma$	30198,7	530,5	2,92	0,2707 ^b	—	0,28	2,048
	$E^1\Sigma$	34900	430	—	—	—	—	—
	$F^1\Sigma$	—	—	—	—	—	—	—
Pb ²⁺ F ⁻	$X^2\Pi_{1/2}$	0	507,2	2,30	2,23 ^a	—	—	2,05 ^a
	$X^2\Pi_{3/2}$	8286	531,1	1,50	—	—	—	—
	$A^1\Sigma$	22566,6	397,8	1,77	—	—	—	—
	$B^1\Sigma$	35843	612,8	3,42	—	—	—	—
	$C^1\Sigma$	38046	594,0	2,50	—	—	—	—
	$D^1\Pi$	43820	597	—	—	—	—	—
	E	45400	562	—	—	—	—	—
	$F^1\Pi$	47870	628	—	—	—	—	—
HgF	$X^2\Sigma$	0	490,8	4,05	0,23 ^a	—	—	2,0 ^a
	$A^2\Pi_{1/2}$	39060,0	506	—	—	—	—	—
	$A^2\Pi_{3/2}$	42999,6	469,4	10,05	—	—	—	—

a) $A \approx 175 \text{ cm}^{-1}$

b) the value of B_0 is given

c) calculated by means of the relation (1.38)

- d) this state is possibly the ground state of ZrO
 - e) adopted on the basis of an estimate by Afaf
 - f) the mean value for the multiplet is given
 - g) the constant of the $\text{Pb}^{206}\text{O}^{16}$ molecule is given.
 - h) estimate
- A) Molecule; B) state; C) cm^{-1} .

An analysis of the rotational structure of the bands of PbF was not carried out in the works [2947, 3463, 3464], and the type of excited states of this molecule has been determined by Rochester [3464] on the basis of the appearance of the bands and in analogy with the diatomic molecules of the halogen compounds of other elements of the 4th Group.* The splitting of the components of the state $X^2\Pi$ of the PbF molecule has been calculated by Rochester from the band edges of the systems $B^2\Sigma - X^2\Pi_{1/2}$ and $B^2\Sigma - X^2\Pi_{3/2}$. This is in good agreement with that found recently in the work of Barrow, Butler, Jones and Powell [647] from an analysis of the systems $A^2\Sigma \leftrightarrow X^2\Pi_{1/2}, X^2\Pi_{3/2}$.

The rotational constant of the PbF molecule, given in Table 289, has been calculated for $r_{\text{PbF}} = 2.05 \text{ \AA}$. This value of the interatomic distance in the PbF molecule has been adopted on the basis of the bond length Pb - F in the PbF_2 molecule, which, according to [70], is 2.13 \AA , and a comparison of the bond lengths M - X in the molecules MX, MX_2 and MX_4 , where M is an atom of the 4th Group and X is a halogen atom. The probable error in the adopted value of r_{PbF} is $\pm 0.07 \text{ \AA}$.

The first edition of the Handbook and the work of Varshni [4073] recommended a lower value (1.98 \AA) which agrees with the adopted value within the limits of accuracy of the estimations.

HgF. The only investigation of the spectrum of mercury monofluoride has been carried out by Howell [2137] who observed in the emission spectrum of HgF two band systems in the region 2300-2600 \AA . These band systems were interpreted as being due to transitions from two

components of the excited state to the ground state $X^2\Sigma$ of the HgF molecule. On the basis of an analysis of the bands, Howel [2137] determined the vibrational constants in the states $X^2\Sigma$ and $A^2\Pi$, adopted by Herzberg [2020] and in the Handbook [649] and given in Table 289. The rotational structure of the bands of HgF has not been resolved and experimental data on the magnitude of its rotational constant are absent from the literature for this reason.

It should be noted that the estimate of the interatomic distance in the molecule HgF is difficult, in contrast to the diatomic molecules formed by the other elements of the 2nd and 7th Groups because of the absence of data on the bond length of Hg - F in the HgF₂ molecule (see [388]). Based on an estimate of the interatomic distance Hg - F in the molecule HgF₂ ($r \approx 2.05$ A) and also by means of various semiempirical methods, Spiridonov [388] and the authors of the Handbook estimated the interatomic distance in the HgF molecule and adopted the values 2.03 and 2.00 ± 0.05 A, respectively. Varshni [4073], based on the relation of values inversely proportional to the square root of the force constants of the diatomic molecules, formed by the elements of the 2nd and 7th Groups, and their interatomic distances, found $r_{\text{HgF}} = 1.81$ A. This value is certainly erroneous and too low because it is close to the interatomic distance M - F in the molecules MgF₂ and ZnF₂ (1.77 and 1.81 A, respectively). The rotational constant given in Table 289 has been calculated on the assumption that $r_{\text{HgF}} = 2.00$ A.

ZrO₂. Indications with regard to any investigations of the spectra and structure of the molecule ZrO₂ are absent from the literature. A reliable theoretical estimate of the molecular constants of ZrO₂ is difficult because of the absence of data on the structure of this molecule. In analogy with CO₂ it can be assumed that the molecule of

zirconium dioxide ZrO_2 has a linear structure. However, between the molecules CO_2 and ZrO_2 there is an important difference. The carbon atom forms bonds in the CO_2 molecule in the valency state sp in consequence of which CO_2 has a linear structure. The two other $2p$ electrons of the C atom in CO_2 form π bonds. At the same time, the Zr atom can be present in the ZrO_2 molecule in four valency states (d^2 , sp , sd , or pd) and whereas a linear structure of the zirconium dioxide molecule should correspond to the two first states, in the two last states the angle between the bonds may be of the order of $90-100^\circ$. Even indirect data of any kind which could enable a choice to be made between the two possible models of the ZrO_2 molecule are not available at present. It has been assumed in the Handbook that it has a linear structure and the molecular constants have been estimated for this model.

The product of the inertial moments, given in Table 290, has been calculated for $r_{Zr-O} = 1.7 \text{ \AA}$ (as in ZrO): the uncertainty in this value is $\pm 0.05 \text{ \AA}$. The fundamental frequencies of ZrO_2 were calculated by means of Eqs. (P4.31), using $f_d(ZrO_2) = k_e(ZrO) = 7.03 \cdot 10^5$, $f_{dd} = 0.1$, $f_d = 0.7 \cdot 10^5$ and $f_\alpha/d^2 = 0.29 \cdot 10^5 \text{ dyne}\cdot\text{cm}^{-1}$. The values of f_{dd} and f_α/d^2 were estimated on the basis of the adopted value of f_d and the relations between f_d , f_{dd} and f_α/d^2 in the CO_2 molecule. The uncertainty in the adopted values of the frequencies is 10-20%.

An analogous estimate of the frequencies of ZrO_2 has been carried out in the work of Chupka et al [1108] and $\nu_1 = 865$, $\nu_2 = 271$ and $\nu_3 = 1003 \text{ cm}^{-1}$ were obtained.

TABLE 290

Adopted Values of the Molecular Constants of ZrO_2

ν_1	$\nu_2(2)$	ν_3	I	σ
1 cm^{-1}			$10^{-2} g \cdot cm^2$	
900	300	1000	15,9	2

1) cm^{-1} ; 2) $g \cdot cm^2$.

PbF_2 . The spectrum of lead difluoride has not been studied by anyone to date. The structure of the PbF_2 molecule has been investigated simultaneously with the other compounds of Pb and Sn in the work of Akishin, Spiridonov and Khodchenkov [70] by the method of electron diffraction. Because only one maximum is observed on the radial distribution curves, plotted for the investigated molecules (for PbF_2 at $r = 2.13 \pm 0.2$ A) and because coincidence of the theoretical and experimental curves for the dependence of the intensity of the scattered electrons on the scattering angles could not be achieved at any values of the angle F - Pb - F, the authors of the work [70] arrived at the conclusion that it is impossible to determine the structure of these molecules without additional data on the composition of the vapor of the investigated substances. A mass spectrometric investigation of the composition of lead diiodide vapor was carried out later at the same laboratory and it was shown that it consists mainly of PbI molecules. This permits the assumption that the non-coincidence of the theoretical and experimental intensity curves is not due to a change in the composition of the PbF_2 vapor but to the large vibration amplitude of the atoms in this molecule which makes determination of the angle F - Pb - F by the electron diffraction method impossible.

The product of the moments of inertia of PbF_2 given in Table 291 has been calculated for $r_{Pb-F} = 2.13$ A and $\angle F - Pb - F = 95^\circ$. The uncertainty in this value may be as much as 10-15%.

The fundamental frequencies of the PbF_2 molecule, given in Table 291, were calculated by means of Eqs. (P4.30) and the following force

constants: $f_d(\text{PbF}_2) = k_e(\text{PbF}) = 2.64 \cdot 10^5$, $f_d/d^2 = 0.1$, $f_d = 0.26 \cdot 10^5$ dyne·cm⁻¹. The uncertainty in the calculated frequencies is 10-15%.

TABLE 291

Adopted Values of the Molecular Constants of PbF₂

ν_1	ν_2	ν_3	$I_A I_B I_C$	σ
1 cm ⁻¹			10 ⁻³⁰ (g·cm ²) ³	
			2	
500	230	510	4,57·10 ⁸	2

1) cm⁻¹; 2) g·cm².

§120. THERMODYNAMIC FUNCTIONS OF GASES

The thermodynamic functions of zirconium, lead and mercury, and also of the above considered compounds of these metals in the ideal gas state were calculated for temperatures of 293.15-6000°K and are given in the Tables 253, 255, 256, 258-260, 262, 263 and 265 of 2nd volume of the Handbook. The differences between the constants of the individual isotope modifications of the molecules were not taken into account in the calculation of the thermodynamic functions of these gases. Appendices 5 and 6 give information on the molecular interaction constants and the critical constants of mercury; for the other gases, considered in this chapter, analogous data are lacking.

Zr. The thermodynamic functions of gaseous monatomic zirconium, given in Table 262 (II), have been calculated by means of Eqs. (II.22) and (II.23). The translational components were calculated by means of the relations (II.8)-(II.9) (the corresponding values of A_ϕ and A_S are given in Table 292), and the electron components, on the basis of the excitation energies of the levels, given in Table 286. The partition functions of the electron states and its derivative with respect to the temperature were calculated by direct summation of the energy

levels on a high-speed electronic computer. The calculation was carried out, taking into account a limited number of electronic states of the zirconium atom in accordance with the method described earlier (see page 133). In order to simplify the calculation, the maximum value of the principal quantum number was assumed to be 11 and constant for all temperatures. The uncertainties in the thermodynamic functions of zirconium calculated in this way at temperatures up to 3000°K do not exceed ± 0.005 cal/g-atom·degree and are mainly due to the inaccuracy of the physical constants adopted in the Handbook.

At a temperature of 6000°K, the main error in the functions is caused by the estimate of the energy of the states with the electron configuration $\dots 4d^4$; the corresponding error in the value of Φ_{6000}^* is about ± 0.02 cal/g-atom·degree. The errors due to the inclusion of excess energy levels, the approximate estimate of the energies of a large number of terms, the neglect of the following groups of terms and the inaccuracy of the determination of n_{\max} is 0.01 cal/g-atom·degree. The total error in the calculated value of Φ_{6000}^* is 0.03-0.04 cal/g-atom·degree. The relatively small total error in the value of Φ_T^* in spite of the fact that the excitation energies of many levels were estimated with low accuracy, is explained by the fact that the lower levels of the zirconium atom have a high multiplicity and make a very large contribution to the sum of the states. Because of this, the sum of the states at 6000°K is of the order of 43 compared with a few units for the other atoms considered in the Handbook.

The thermodynamic functions of monatomic zirconium were calculated by Katz and Margrave [2334] (Φ_T^* for $\leq 2000^\circ\text{K}$), Stull and Sinke [3894] ($T \leq 3000^\circ\text{K}$) and by Kolsky et al [2462] ($T \leq 8000^\circ\text{K}$). Up to 6000°K the results of these calculations and the data of the present Handbook agree within the limits of 0.01 cal/g-atom·degree. This coin-

cides, in spite of the fact that the Handbook authors included a large number of additional levels (in the work [2462] only the levels given by Moore [2941] were taken into account in the calculations), is due to the relatively small contribution of these levels in the sum of states because of its large absolute value.*

TABLE 292

Numerical Values (in cal/g-atom·degree) of the Constants for the Calculation of the Thermodynamic Functions of Gaseous Zr, Pb and Hg.

Вещество A	A_{ϕ}	A_S
Zr	6,1700	11,1382
Pb	8,6161	13,5843
Hg	8,5196	13,4878

A) Substance.

Pb and Hg. The thermodynamic functions of gaseous monatomic lead and mercury, given in Tables 253 (II) and 259 (II), were calculated by means of Eqs. (II.22) and (II.23). The translational components were calculated by means of the relations (II.8)-(II.9) (the corresponding values of A_{ϕ} and A_S are given in Table 292). The electron components were calculated by direct summation of the energy levels of the atoms Pb and Hg, given in Tables 287 and 288.

The accuracy of the numerical values of the thermodynamic functions of monatomic lead and mercury at temperatures to 4000-5000°K calculated in this manner depends only on the accuracy of the physical constants used for the calculation and the errors thus do not exceed 0.005 cal/g-atom·degree. The fact that the levels with principal quantum number $n \leq 11$ and the excitation energies over 50,000 cm^{-1} and also the levels of the Pb atom, for which experimental data on their excitation energies are lacking (states with the configuration $\dots 6s6p^3$ had not been allowed for in the calculations begins to make itself felt at higher temperatures. However, the corresponding errors are small and do not exceed ± 0.01 and ± 0.003 cal/g-atom·degree in the values of Φ_{6000}^* for lead and mercury.

The thermodynamic functions of monatomic lead have been calculated earlier by Brewer [1093] (S_T° for $T \leq 2000^\circ\text{K}$), Stull and Sinke [3894] ($T \leq 3000^\circ\text{K}$) and Kolsky et al [2462] ($T \leq 8000^\circ\text{K}$), and for mercury, in the works of Wieland and Herzog [4259a] ($T \leq 1500^\circ\text{K}$), Kelley [2364a] (S_T° for $T \leq 3000^\circ\text{K}$), Busey and Giauque [1035] ($T \leq 700^\circ\text{K}$) and Stull and Sinke [3894] ($T \leq 3000^\circ\text{K}$). The results of all these calculations, with the exception of Kelley's data [2364a] are in good agreement. The data of Kelley differ from those given in the present Handbook by 0.03-0.05 cal/g-atom-degree.

The differences between the thermodynamic functions of lead and mercury, given in the first and present Handbook editions, are due to a slight improvement in the data on the electron levels of Pb and the difference in the values of the physical constants.

ZrO. The thermodynamic functions of gaseous zirconium monoxide, given in Table 263 (II), were calculated by means of Eqs. (II.161) and (II.162). The values of $\ln \Sigma$ and $T \partial/\partial T \ln \Sigma$ in these equations were calculated by the method of Gordon and Barnes [Eqs. (II.137) and (II.138)] without application of corrections for the limited summation of the rotational energy levels; the values of $\ln \Delta$ and $T \partial/\partial T \ln \Delta$ in Eqs. (II.161) and (II.162) were calculated by means of Eqs. (II.157) (II.158). Table 293 gives the values of the constants C_ϕ and C_S for the calculation of the components of the translational motion and the rigid rotator, the values of θ and x for the interpolation by means of the anharmonic oscillator Tables, and also the values of the coefficients in the Eqs. (II.137) and (II.138).

The components of the excited states $a^1\Delta$, $A^3\Phi$, $B^3\Pi$ and $C^3\Delta$ were calculated by the simplest method [Eqs. (II.120)-(II.121)], i.e., without allowance being made for the difference in the molecular constants in the ground and excited states of this molecule. The use of more ac-

curate methods for calculating the components of the excited states is not justified because of the absence of experimental data on the excitation energy of the lowest state $a^1\Delta$. For the same reason, the other electron states of ZrO, given in Table 289, have not been taken into account in the calculations.

TABLE 293

Numerical Values of the Constants for the Calculation of the Thermodynamic Functions of Gaseous ZrO, PbO, PbF and HgF

Вещество	θ	$\alpha \cdot 10^3$	$\beta_1 \cdot 10^3$	$\beta_2 \cdot 10^3$	$\frac{q_0}{T}$	$\frac{d_0 \cdot 10^3}{T}$	C_0	C_5
A	град B				град ⁻¹ B		кал/моль·град C	
ZrO	1348,2	3,5745	0,5065	0,26	1,67631	2,67	11,2392	18,1945
PbO	1038,3	5,1261	0,6202	0,30	2,26922	6,52	10,4662	17,4215
PbF	729,75	4,5317	—	—	—	—	12,4524	19,4079
HgF	706,16	8,2518	—	—	—	—	12,3645	10,3200

A) Substance; B) degree; C) cal/mole·degree.

The main uncertainties in the calculated values of the thermodynamic functions of zirconium monoxide are due to the absence of experimental data on the excitation energy of the state $a^1\Delta$, the energy of the higher vibrational levels of ZrO in the state $X^3\Delta$, the interaction constant α_1 and the bond constant A in the state $X^3\Delta$ and also the approximate calculation of the components of the excited states. Moreover, reliable data on the type of electron ground state of this molecule are lacking.

At low temperatures, the error in the calculated values of the thermodynamic functions are practically determined only by the absence of reliable data on the bond constant A of the ZrO molecule in the state $X^3\Delta$. It can be assumed that the corresponding error in the value of $\Phi_{298.15}^*$ does not exceed ± 0.1 cal/mole·degree and that it decreases with increase in the temperature. At higher temperatures the errors

to the other causes pointed out above, become important. The total error in the values of Φ_{3000}^* and Φ_{6000}^* should be of the order of 0.5 and 1.5 cal/mole·degree, respectively, if the ground state of ZrO is the $^3\Delta$ state.

The table of the thermodynamic functions of ZrO is being published for the first time; in the work [1108], the thermodynamic functions of zirconium oxide were calculated for the processing of the experimental data on the basis of the molecular constants given in the book of Herzberg [2020], including the incorrect value B_0 obtained by Lowater [2666].

PbO. The thermodynamic functions of gaseous lead monoxide, given in Table 255 (II), were calculated by means of Eqs. (II.161) and (II.162). The values of $\ln \Sigma$ and $T \partial/\partial T \ln \Sigma$ were calculated by the method of Gordon and Barnes on the basis of the molecular constants, adopted in Table 289.

Table 293 gives the values of C_ϕ and C_S for the calculation of the components of the translational motion and the rigid rotator, the values of θ and x for interpolation by means of the anharmonic oscillator Tables and also the values of the coefficients in Eqs. (II.137) and (II.138).

The components of the excited state $A^1\Sigma$ of the PbO molecule in the values of the thermodynamic functions of this gas have been calculated by means of Eqs. (II.126)-(II.127) and the components of the higher states, by means of Eqs. (II.120)-(II.121), i.e., without taking into account the difference between the constants of the ground and excited state.

The main uncertainties in the calculated values of the thermodynamic functions of lead monoxide are due to the lack of data on the energy of the higher vibrational and rotational levels of the PbO mo-

lecule in the state $X^1\Sigma$ and also the difference between the magnitude of the dissociation energy, as adopted in the Handbook, and that calculated by linear extrapolation. However, owing to the fact that the PbO molecule has a relatively high dissociation energy, the corresponding errors and also the errors due to neglecting the need for limiting the number of rotational levels of the PbO molecule are slight and apparently do not exceed 0.05 cal/mole·degree in the values of Φ_T^* at $T \leq 3000^\circ\text{K}$ and 0.1 cal/mole·degree in the values of Φ_{6000}^* .

The entropy of lead oxide has been calculated earlier by Kelley [2363] at $T \leq 2000^\circ\text{K}$. The calculation was carried out by means of the rigid rotator-harmonic oscillator model approximation on the basis of an erroneous vibration frequency. The discrepancy between the results of this calculation and the values given in Table 255 (II) of the present Handbook attains 0.1 cal/mole·degree.

The thermodynamic functions of PbO, given in the first and present editions of the Handbook, are identical.

PbF and HgF. The thermodynamic functions of lead and mercury monofluoride, given in Tables 256 (II) and 260 (II), were calculated by means of Eqs. (II.161) and (II.162) on the basis of the rigid rotator-harmonic oscillator model approximation. The calculation was carried out on the basis of the constants which were adopted in the preceding section. Table 239 gives the values of C_ϕ and C_S for the calculation of the components of the translational motion and the rigid rotator and also the values of θ and x for the calculation of the components of the anharmonic oscillator by the method of Gordon and Barnes.

The multiplicity of the ground state of HgF has been taken into account in the calculation of the thermodynamic functions of this gas by including the addend $R \ln 2$ in the values of C_ϕ and C_S , given in Table 293. In the case of PbF, in view of the large splitting of the

components of the $X^2\Pi$ ground state of this molecule, the multiplicity of the ground state was allowed for by adding the component $R \ln 2$ to the values of C_p and C_s (component of the Λ doubling of the $^2\Pi_{1/2}$ component).

The components of the second component of $^2\Pi_{3/2}$ of the ground state was calculated as a component of the excited state together with the components of the states $A^2\Sigma$ and $B^2\Sigma$. Because experimental data on the rotational constants of PbF are lacking, the components of the excited states $X^2\Pi_{3/2}$, $A^2\Sigma$ and $B^2\Sigma$ were calculated by the simplest method [Eqs. (II.120)-(II.121)].

The basic uncertainty in the calculated values of the thermodynamic functions of PbF and HgF at low temperatures is connected with the absence of experimental data on the rotational states of these molecules (of the order of 0.15 and 0.1 cal/mole·degree, respectively). At high temperatures, errors due to the absence of data on the centrifugal stretch constants and rotation-vibration interaction are added to these uncertainties (up to 0.15 cal/mole·degree in the values of Φ_{6000}^*). The total errors in the values $\Phi_{298.15}^*$, Φ_{3000}^* and Φ_{6000}^* of lead and mercury monofluoride are of the order of 0.15; 0.2 and 0.3 cal/mole·degree.

The thermodynamic functions of HgF, given in the first and present Handbook editions, are identical; the differences between the values of the functions of PbF are due to the more accurate estimate of the interatomic distance of this molecule and amount to about 0.13 cal/mole·degree.

Other calculations of the thermodynamic functions of PbF and HgF are not known in the literature.

ZrO₂. The thermodynamic functions of gaseous zirconium dioxide, given in Table 265 (II), were calculated by means of Eqs. (II.241) and

(II.242) using the rigid rotator-harmonic oscillator model approximation. The calculation was carried out on the basis of the data given in Table 290. Table 294 gives the values of C_ϕ and C_S for the calculation of the components of the translational motion and the rigid rotator, and the values of θ_n for the calculation of the components of the harmonic oscillators.

The uncertainties in the calculated values of the thermodynamic functions of zirconium dioxide due to the inaccuracy of the interatomic distance and the fundamental frequencies of this molecule amount to ± 0.5 , ± 1.5 and ± 1.8 cal/mole·degree in the values of $\Phi_{298.15}^*$, Φ_{3000}^* , and Φ_{6000}^* , respectively. The errors in these values due to the fact that the calculation was based on the rigid rotator-harmonic oscillator model approximation, are about 0.2; 1.2 and 1.8 cal/mole·degree. It must be remembered that the difference in the values of Φ_T^* , calculated for the linear and angular model of the ZrO_2 molecule, are about 6; 4.5 and 4 cal/mole·degree, respectively, at 198.15; 3000 and 6000°.

The errors in the values of $\Phi_{298.15}^*$, Φ_{3000}^* and Φ_{6000}^* , given in Table 265 (II), are assumed to be ± 0.7 , ± 2.5 and ± 3.5 cal/mole·degree.

TABLE 294

Numerical Values of the Constants for the Calculation of the Thermodynamic Functions of ZrO_2 and PbF_2

Вещество A	θ_1	θ_2	θ_3	C_ϕ	C_S
	град ⁻¹ B				
ZrO_2	1294,9	431,64	1438,8	8,4191	15,3742
PbF_2	719,40	330,92	733,78	13,0000 ^a	21,0477 ^a

a) The values C_ϕ^1 and C_S^1 are given. A) Substance; B) degree⁻¹; C) cal/mole·degree.

Other calculations of tables of the thermodynamic functions of zirconium dioxide are not known in the literature. In the work [1108], the thermodynamic functions of zirconium dioxide for several temperatures were calculated during the processing of the experimental data (see page 1954) by means of the nearest values of the fundamental frequencies, but with an inaccurate value of $r_{\text{Zr-O}}$, found by Lowater for ZrO (see page 1922).

PbF₂. The thermodynamic functions of gaseous lead difluoride, given in Table 258 (II), were calculated by means of Eqs. (II.243) and (II.244) and the molecular constants, given in Table 291, using the rigid rotator-harmonic oscillator model approximation. The corresponding values of C_p , C_v and θ_n are given in Table 294.

The error in the calculated values of the thermodynamic functions of lead difluoride, owing to the absence of experimental data on the fundamental frequencies and the angle between the bonds in the PbF₂ molecule, and also because of basing the calculation on the rigid rotator-harmonic oscillator model approximation, is of the order of 0.5; 2 and 3 cal/mole·degree in the values of $\Phi_{298.15}^*$, Φ_{3000}^* and Φ_{6000}^* , respectively.

The thermodynamic functions of lead difluoride, given in the first and present Handbook editions, differ by 0.436 cal/mole·degree because of the more accurate value found for the interatomic distance Pb - F in this molecule. Other calculations of the thermodynamic functions of lead difluoride are not known in the literature.

§121. THERMODYNAMIC PROPERTIES OF Zr, ZrO₂, Pb, PbO AND PbF₂ IN THE SOLID AND LIQUID STATE

The thermodynamic functions of Zr, ZrO₂, Pb, PbO and PbF₂ in the solid and liquid state (Tables 261, 264, 252, 254 and 257 of the 2nd volume) were calculated by means of Eqs. (III.9)-(III.11) on the basis

of the thermodynamic values given in Table 295. The uncertainties in the calculated values of Φ_T^* at the temperatures 298, 1000, 1500, 2000 and 3000°K, estimated on the basis of an analysis of the possible errors in the original data, are given in Table 296.

TABLE 295

Adopted Values of the Thermodynamic Quantities for Zirconium, Lead and Their Compounds in the Solid and Liquid State

Вещество A	Состояние B	$H_{298,15}^\circ - H_0^\circ$	$S_{298,15}^\circ$	$C_{p,298,15}^\circ$	Коэффициенты в уравнении для C_p°			Интервал температур °K	T_{tr} или °K	ΔH_{tr} или кал/моль
		кал/моль	кал/моль-град	кал/моль-град	a	b·10 ³	c·10 ⁻⁵			
Zr	Крист. H	1313	9,29	6,01	6,65	1,11	0,864	298,15—1136	1136	900
Zr	°	—	—	—	7,37	—	—	1136—2128	2128	4000
Zr	Жидк. I	—	—	—	8,0	—	—	2128—6000	—	—
ZrO ₂	Крист.	2100	12,12	13,40	16,64	1,80	3,36	298,15—1478	1478	1420
ZrO ₂	°	—	—	—	17,8	—	—	1478—2950	2950	20800
ZrO ₂	Жидк.	—	—	—	24,0	—	—	2950—6000	—	—
Pb	Крист.	1644	15,49	6,32	5,629	2,327	—	298,15—600,6	600,6	1140
Pb	Жидк.	—	—	—	7,765	-0,74	—	600,6—1200	—	—
Pb	°	—	—	—	6,88	—	—	1200—3600	—	—
PbO	Крист. K	2177	15,8	10,95	8,64	7,76	—	298,15—762	762	120
PbO	красн. K	—	—	—	11,08	2,71	0,85	762—1170	1170	6100
PbO	Жидк. L	—	—	—	15,50	—	—	1170—2900	—	—
PbF ₂	Крист.	3800	27,0	17,7	12,58	17,17	—	298,15—720	720	500
PbF ₂	°	—	—	—	24,5	—	—	720—1095	1095	2850
PbF ₂	Жидк.	—	—	—	24,0	—	—	1095—2900	—	—

a) $C_p^\circ = a + bT - cT^{-2}$ (cal/mole·degree)

A) Substance

B) state

C) cal/mole

D) cal/mole·degree

E) coefficients in the equation for C_p°

F) temperature range

G) or

H) crystalline

I) liquid

K) red

L) yellow

Two crystalline modifications of zirconium are known, a low-temperature, hexagonal closepacked modification (structure type of Mg) which is stable up to 1136°K, and a cubic body centered one (structure

type of Na) existing at higher temperatures.

The heat capacity of zirconium at low temperatures has been investigated by Todd [3994] (53-297°K), Skinner and Johnston [3757] (14-298°K), Burk, Estermann and Friedberg [1024] (20-200°K) and Wolcott [4308] (1.2-20°K). The most accurate measurements are those of Skinner and Johnston, according to whom $S_{298.15}^{\circ} = 9.29 \pm 0.04$ cal/g-atom·degree and $H_{298.15}^{\circ} - H_0^{\circ} = 1313$ cal/g-atom. The correctness of the extrapolation of the heat capacity of Zr below 14°K ($S_{14}^{\circ} = 0.03$ cal/g-atom·degree) [3757] is confirmed by the data of Wolcott [4308] ($S_{14}^{\circ} = 0.029$ cal/g-atom·degree).

The enthalpy of solid zirconium at high temperatures was measured by Jaeger and Veenstra [2202] (294-1074°K), Coughlin and King [1197] (390-1371°K), Skinner [3755] (up to 1800°K) and Douglas and Victor [1390] (273-1173°K). For the low-temperature modification of zirconium, which is stable to 1136°K, the most reliable data are those of Douglas and Victor [1390], who carried out 58 enthalpy measurements on a specimen of pure zirconium (99.91% Zr) by means of a copper calorimeter, suitable for high-precision investigations. On the basis of the values $H_{673.15}^{\circ} - H_{298.15}^{\circ} = 2535$ cal/g-atom and $H_{1073.15}^{\circ} - H_{298.15}^{\circ} = 5560$ cal/g-atom, adopted from the data [1390], and the value $C_{p298.15}^{\circ} = 6.01$ cal/g-atom·degree [3757], the authors of the Handbook derived an equation for the heat capacity of the low-temperature modification of zirconium (see Table 295). The heat of the polymorphous transformation, $\Delta H_{1136} = 0.90$ kcal/g-atom, has also been taken from the data of Douglas and Victor [1390]. The measurements of the enthalpy of the high-temperature modification of zirconium, carried out by the authors of the work [1390] within a narrow temperature range (1136-1173°K) gives an excessive value for the heat capacity of Zr ($C_p^{\circ} = 8.85$ cal/g-atom·degree). Skinner [3755] on the basis of his own measurements and those

of Coughlin and King [1197] concluded that the heat capacity of zirconium increases linearly and amounts to 6.79 at 1200°K and 7.75 cal/g-atom·degree at 1800°K. The constant value $C_p^\circ = 7.37$ cal/g-atom·degree, derived on the basis of the values $H_{1136}^\circ - H_{298}^\circ = 6925$ cal/g-atom [1390] and $H_{1800}^\circ - H_{298}^\circ = 11,820$ cal/g-atom [3755], has been adopted in the Handbook for the heat capacity of the hightemperature modification of zirconium.

The melting point of zirconium has been measured by MacPherson (2118° ± 25°K, see the work of Adenstedt [492]), Oriani and Jones [3417] (2141° ± 10°K, maximum deviations of individual measurements from the mean value up to 19°) and Deardorff and Hayes [1279] (2128 ± 15°K, reproducibility ±3°K). The last value has been adopted in the Handbook as the most reliable. The heat of melting of zirconium has not been determined experimentally. Kubaschewski and Evans [2494] estimated the heat of melting of zirconium as 4.6 ± 0.7 kcal/g-atom ($\Delta S_m = 2.2$ cal/g-atom·degree). According to Stull and Sinke [3894], the entropy of melting of elements with cubic body-centered lattice is approximately 1.9 cal/g-atom·degree, which corresponds to the value $\Delta H_m = 4.0$ kcal/g-atom, adopted in the Handbook. The accuracy of this value is 0.5-0.7 kcal/g-atom. The heat capacity of molten zirconium, according to the estimate [3894] is 8.0 cal/g-atom·degree.

The thermodynamic functions of zirconium in the solid and molten state, calculated for the temperatures 293.15-6000°K, are given in Table 261 (II). The uncertainties in the calculated values of Φ_T^* are given in Table 296.

The calculated values of the thermodynamic functions differ from those given in the Handbook of Stull and Sinke [3894] by not more than 0.07 and 0.15 cal/g-atom·degree in the values of Φ_T^* and S_T° in consequence of the improvement in the heat capacity of solid zirconium as a

result of the work of Douglas and Victor [1390] which has been taken into account in the present Handbook.

ZrO₂. Zirconium dioxide is known in two modifications, the low-temperature monoclinic modification (distorted structure of the type CaF₂), stable up to 1478°K, and the high-temperature cubic modification which is stable at higher temperatures.

Kelley [2362] investigated the heat capacity of zirconium dioxide within the range of 54-295°K and calculated $S_{298.15}^{\circ} = 12.03 \pm 0.08$ cal/mole·degree. Later on, Kelley [2364] corrected an error in this calculation and recommended the value $S_{298.15}^{\circ} = 12.12 \pm 0.08$ cal/mole·degree. Extrapolation of the heat capacity of ZrO₂ to 0°K gives $S_{50.12} = 0.445$ cal/mole·degree. The value $H_{298.15}^{\circ} - H_0^{\circ} = 2100 \pm 10$ cal/mole, given in the Handbook, has been calculated on the basis of the data [2362].

The enthalpy of ZrO₂ within the temperature range 298-1850°K has been measured by Coughlin and King [1197] who recommended a trinomial equation for the specific heat of the low-temperature modification of ZrO₂ (see Table 295), the heat of transformation $\Delta H_{1478} = 1.42 \pm 0.1$ kcal/mole and the specific heat of the high-temperature modification of ZrO₂ is $C_p^{\circ} = 17.8$ cal/mole·degree (1478-1850°K). These values have been adopted in the Handbook. The heat capacity of zirconium dioxide at higher temperatures up to the melting point 2950° [3508] has also been taken as 17.8 cal/mole·degree. The heat of melting of ZrO₂ $\Delta H_m = 20.8$ kcal/mole has been adopted on the basis of the recommendation of Kelley [2356] who calculated it from the state diagrams of the two-component systems. The heat capacity of molten ZrO₂ is estimated as 24.0 cal/mole·degree.

The thermodynamic functions of zirconium dioxide in the solid and liquid state, calculated for the temperatures 293.15-6000°K, are

given in Table 264 (II). The magnitude of the possible errors in the calculated values of Φ_T^* is indicated in Table 296.

Pb. Lead is known in the form of the cubic face-centered modification (structure type of Ag). Indications with regard to any polymorphism of lead are absent in the literature.

Kelley [2364] based on the data of different authors on the specific heat of lead within the range 2-298°K recommended the value $S_{298.15}^a = 15.49 \pm 0.05$ cal/g-atom·degree. The value $H_{298.15}^o - H_0^o = 1644 \pm 5$ cal/g-atom has been adopted in the Handbook on the basis of the data of Meads, Forsythe and Giaque [2824]. The results of the specific heat measurements on lead, published in 1952, carried out by Horowitz, Silvidi, Malaker and Daunt [2130] within the range of 1-75°K do not introduce any important corrections into the above-given values.

The most reliable data on the enthalpy of solid and molten lead up to 1200°K have been obtained by Douglas and Dever [1387]. The linear equations for the specific heat of solid and molten lead, derived by the authors [1387] and also the melting point (600.6°K) and the heat of melting of lead ($\Delta H_{600.6} = 1140 \pm 20$ cal/g-atom) are given in Table 295. The heat of melting of lead $\Delta H_{600.6} = 1158 \pm 20$ cal/g-atom, obtained recently by Oelsen [3111], agrees within the limit of possible error with the above value.

Stull and Sinke [3894] used in their calculation of the thermodynamic functions of molten lead above 1200°K the extrapolation of the equation of Douglas and Dever [1387], according to which the specific heat decreases from $C_{P1200}^o = 6.88$ to $C_{P200}^o = 6.29$ cal/g-atom·degree. The last value is evidently too low. The experimental data show that the heat capacity of metals is usually within the range of 7 to 8 cal/g-atom·degree. Hence, the constant value $C_{P1200}^o = 6.88$ cal/g-atom·

degree has been adopted in the Handbook for the heat capacity of molten lead above 1200°K.

The numerical values of the thermodynamic functions of lead in the solid and molten state, calculated for the temperatures 293.15-3600°K, are given in Table 252 (II). The magnitude of the possible error in the calculated values of Φ_T^* is indicated in Table 296.

The calculated thermodynamic functions of lead [Table 252 (II)] are identical with those given in the first edition of the Handbook. The differences between these and the thermodynamic functions of molten lead, calculated by Stull and Sinke [3894] are only slight and amount to 0.02 and 0.14 cal/g-atom·degree in the values of Φ_{2000}^* and S_{2000}° , respectively.

PbO. Lead monoxide exists in two crystalline modifications, the red tetragonal modification which is stable at temperatures under 762°K, and the yellow rhombic modification, which is thermodynamically stable at higher temperatures. When the yellow modification of PbO is cooled rapidly, the transformation to the red modification does not take place which has made it possible to investigate the heat capacity and enthalpy of yellow PbO to very low temperatures. At the present time, the thermodynamic quantities for yellow PbO are known with greater accuracy than for the red modification.

The most reliable measurements of the specific heat of the yellow modification of PbO at low temperatures were carried out by Strelkov, Kostryukov and Morozova [50, 251a] (12-303°K). The authors of the work [50] calculated the values $H_{298.15}^\circ - H_0^\circ = 2208 \pm 4$ cal/mole and $S_{298.15}^\circ = 16.42 \pm 0.03$ cal/mole·degree. Extrapolation of the heat capacity to below 12°K gives the value $S_{12}^\circ = 0.15 \pm 0.01$ cal/mole·degree. The data of Nernst and Schwes [3046] on the specific heat of PbO (21-93°K) to 70°K are lower and at higher temperatures higher than the

data of [50] (maximum deviation 2%). The value $S_{298.15}^{\circ} = 16.6 \pm 0.5$ cal/mole·degree, calculated by the authors [3046] is too high because of considerable errors (up to 6%) in the extrapolation of the experimental data to room temperature. The results of the heat capacity measurements on PbO, carried out by King [2403] within the range 53-296°K, are in agreement with the data of Strelkov, Kostryukov and Morozova [50] with an accuracy of 0.5%. However, King obtained for the standard entropy of PbO the inaccurate value 16.1 ± 0.2 cal/mole·degree because of the unreliability of the extrapolation of the specific heat from 53 to 0°K ($S_{51}^{\circ} = 2.78$ cal/mole·degree [2403] instead of $S_{51}^{\circ} = 3.10$ cal/mole·degree on the basis of the data [50]).

The enthalpy of yellow lead monoxide at high temperatures was measured by Spencer and Spicer [3823] (673-923°K) and Rodigina, Gorn'skiy and Luginina [33] (298-1170°K).* The data of Spencer and Spicer [3823] at a temperature of 675°K are 1.6%, and within the range of 773-925°K, 3-4% higher than the data of [33]. The causes of these discrepancies are not clear. The larger values in [3823] are possibly due to oxidation of the PbO samples which has been noted by the authors of [3823] and also the presence of an admixture of the red modification of PbO with its larger specific heat. The Handbook adopted the data of Rodigina et al [33] with presumably greater accuracy ($\pm 0.3\%$) than the data of Spencer and Spicer [3823] ($\pm 1\%$).

The specific heat of the red modification of PbO within the range of 53-296°K has been investigated by King [2403]. The uncertainty in the value of $S_{298.15}^{\circ} = 15.6$ cal/mole·degree, calculated by him, apparently exceeds the value ± 0.2 cal/mole·degree, indicated by the author of the work [2403] in consequence of the unreliability of the extrapolation of the specific heat from 53 to 0°K ($S_{51}^{\circ} = 2.48$ cal/mole·degree). The error in the analogous extrapolation of the data for yellow

PbO, carried out by King [2403], was 0.32 cal/mole·degree.

In order to bring the entropies of red and yellow lead monoxide at 762°K into agreement with the entropy of the equilibrium transformation of PbO at this temperature (see further on) and also taking into account the inaccuracy of the value $S_{298.15}^{\circ}$ for red PbO due to the extrapolation of the specific heat to 0°K, the Handbook authors have chosen the value $S_{298.15}^{\circ} = 15.8 \pm 0.2$ cal/mole·degree, which is more probable than that recommended by King [2403] ($S_{298.15}^{\circ} = 15.6 \pm 0.2$ cal/mole·degree) for the entropy of the red modification of PbO at 298.15°K. For the specific heat of red PbO at temperatures over 298°K the authors of the Handbook established a binomial equation (see Table 295) on the basis of the value $C_{p,298.15}^{\circ} = 10.95$ cal/mole·degree [2403] and the enthalpy change $H_{723}^{\circ} - H_{298}^{\circ} = 5355$ cal/mole, based on the data of Spencer and Spicer [3823] (198-823°K). This equation has been used for the calculation of the thermodynamic functions of red PbO up to the temperature of the equilibrium transformation into yellow PbO of 762°K [3508]. The heat of transformation $\Delta H_{762} = 0.12$ kcal/mole was calculated from the heat of transformation of PbO at 298°K (see page 1958) and the enthalpy changes adopted in the Handbook for red and yellow lead monoxide. The trinomial equation, derived by Rodigina et al [33] has been adopted for the heat capacity of yellow PbO within the temperature range of 762-1170°K.

The melting point of yellow lead monoxide, $1170 \pm 4^{\circ}\text{K}$, the heat of melting $\Delta H_{1170} = 6.10 \pm 0.2$ kcal/mole, and the specific heat of molten PbO, $C_p = 15.5$ cal/mole·degree, were adopted on the basis of the measurements, carried out by Rodigina, Gomel'skiy and Luginina [33] up to a temperature of 1200°K. The last value has also been used in the calculations of the thermodynamic functions at higher temperatures.

The thermodynamic functions of lead monoxide in the solid and

molten state, calculated for the temperatures 293.15-2900°K, are given in Table 254 (II). The magnitude of the possible error in the calculated values of Φ_T^* is indicated in Table 296.

The calculated thermodynamic functions of crystalline and molten PbO [Table 254 (II)] differ from those given in the first Handbook edition by approximately 1 cal/mole·degree due to the improvement of the specific heat and enthalpy of lead monoxide as a result of the works [50, 33].

PbF₂. Lead difluoride is known in two modifications, the low-temperature rhombic (structural type of PbCl₂) and the high-temperature cubic modification (structural type of CaF₂). The transformation temperature is 720°K [3508].

Experimental data on the specific heat of PbF₂ at low temperatures are lacking. The value $S_{298.15}^\circ = 27.0 \pm 2.0$ cal/mole·degree has been adopted in the Handbook on the basis of an estimate by Brewer [1093], and Kubaschewski and Evans [2495]. This value has also been confirmed by calculation of the specific heat of PbF₂ at low temperatures by the method of Karapet'yants [222] using the data on the specific heat of BaF₂ [3263] and the value $C_{P,298.15}^\circ = 17.7$ cal/mole·degree for PbF₂, found by Schottky [3652].* The value $H_{298.15}^\circ - H_0^\circ = 38^\circ \pm 200$ cal/mole has been calculated by means of the above-described calculation by the method of Karapet'yants [222].

Measurements of the enthalpy of solid PbF₂ within the range 664-1087°K have been carried out by Banashek, Patsukova and Rassonskaya [330, 85]. Based on the value $H_{700}^\circ - H_{298}^\circ = 8500$ cal/mole, obtained from the data [85] and the value $C_{P,298.15}^\circ = 17.7$ cal/mole·degree, a linear equation has been derived for the specific heat of the low-temperature rhombic modification of PbF₂. As the authors of the work [85] established, the polymorphic transformation during rapid cooling

of the high-temperature modification of PbF_2 during the calorimetric experiments was not complete, so that the data on the enthalpy of PbF_2 above the transformation temperature (720°K [3508]) and also the value $\Delta H_{\text{tr}} = 0.5$ kcal/mole, calculated from these data, are evidently too low. The authors of the work [85], based on their own data, calculated for the specific heat of the high-temperature modification of PbF_2 the trinomial equation

$$C_p = 25,26 + 0,814 \cdot 10^{-3} T - 7,356 \cdot 10^{-6} T^2.$$

In view of the low accuracy of the data of [85] it is advisable to use for the specific heat of PbF_2 at temperatures over 720°K the value $C_p^\circ = 24.5$ cal/mole·degree, calculated on the basis of the values $H_{750}^\circ - H_{298}^\circ = 10,230$ and $H_{1150}^\circ - H_{298}^\circ = 17,580$ cal/mole. The enthalpy of molten PbF_2 has been measured by Banashek, Patsukova and Rassonskaya [85] within the very narrow temperature range of 1104 - 1164°K , so that the calculation of the specific heat of molten PbF_2 becomes inaccurate. The Handbook adopted for the specific heat of molten PbF_2 the value $C_p^\circ = 24.0$ cal/mole, calculated on the basis of the value $H_{1100}^\circ - H_{298}^\circ = 21,650$ and $H_{1150}^\circ - H_{298}^\circ = 22,850$ cal/mole [85] and for the heat of fusion the value $\Delta H_{1095} = 2.85 \pm 0.3$ kcal/mole.*

The thermodynamic functions of lead difluoride in the solid and molten state, calculated for the temperatures 293.15 - 2900°K , are given in Table 257 (II). The magnitude of the possible errors in the calculated values of Φ_T^* are indicated in Table 296.

The differences between the values of the thermodynamic functions of PbF_2 , given in the first and present Handbook editions, amounts to 2 cal/mole·degree in the values of Φ_T^* and S_T° and are due to the improvement in the estimation of $S_{298.15}^\circ$ in the present edition.

TABLE 296

Uncertainties (in cal/mole·degree) in the Values of Φ_T^* of Zirconium, Lead and Their Compounds in the Solid and Molten State

T, °K	Zr	ZrO ₂	Pb	PbO	PbF ₂
298	±0.04	±0.08	±0.05	±0.2	±2.0
1000	±0.08	±0.2	±0.10	±0.3	±2.2
1500	±0.12	±0.3	±0.15	±0.4	±2.5
2000	±0.15	±0.4	±0.3	±0.8	±3.0
3000	±0.4	±1.0	±0.6	—	—

§122. THERMOCHEMICAL QUANTITIES

The standard state of Zirconium is Zr (cryst.).

Zr (gas). Skinner, Edwards and Johnston [3756] carried out six measurements on the pressure of saturated zirconium vapor by the method of evaporation from a surface within the temperature range 1949-2054°K. The rate of evaporation was determined by differential (on the basis of the weight loss) as well as integral methods (on the basis of the quantity of condensate). The difference between the results obtained with these methods, did not exceed 2.5%. The authors of the work [3756] calculated the heat of sublimation of zirconium, $\Delta H_{s,0} = 142.15 \pm 0.35$ kcal/g-atom, under the assumption that its evaporation coefficient is equal to unity. The thermodynamic functions of solid zirconium used in the calculations in the work [3765] were insufficiently accurate. Calculation of the heat of sublimation of zirconium by means of the more accurate thermodynamic functions, given in the present work, gives the value

$$\Delta H_{s,0}(\text{Zr, } ^{\text{cryst.}}) = \Delta H_f^0(\text{Zr, } \dots) = 145 \pm 5 \text{ kcal/g-atom,}$$

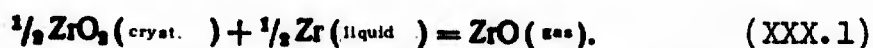
which has been adopted in the Handbook. The uncertainty in the adopted value is much greater than in that recommended by Skinner et al [3756],

in particular, because the hypothesis that the evaporation coefficient of zirconium during evaporation from a surface is equal to unit, is open to doubt (see, for example, the work of An. Nesmeyanov and De Dyk Man [310]).

ZrO (gas). Linear extrapolation of the vibrational energy levels of the state $X^3\Delta$, $A^3\Phi$, and $B^3\Pi$ of the ZrO molecule gives dissociation energies of 189, 200 and 197 kcal/mole, respectively. Taking into account, however, that the state $X^3\Delta$ is possibly not the ground state, these values cannot even be considered as approximate dissociation energies.

The review of Brewer [917] gives for the dissociation energy of ZrO the value $D_0 = 170 \pm 20$ kcal/mole, calculated on the basis of the work of Skinner, Edwards and Johnston [3756] in which it had been found that the pressure of ZrO over the system $Zr + ZrO_2$ is less than the vapor pressure of zirconium or zirconium dioxide. However, the results of the mass spectrometric study [1108] contradict these values.

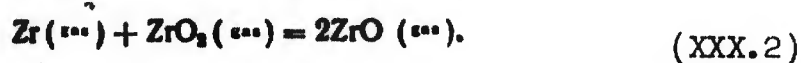
Chupka, Berkowitz and Inghram [1108] carried out a mass spectrometric investigation on the composition of the vapor over the system $Zr - ZrO_2$ and found that ZrO is the main evaporation product of this system at temperatures of 2100 - 2400°K. Using the results of the calibration of the mass spectrometer, based on the evaporation of a known quantity of silver, the authors of this work were able to calculate the absolute partial pressure of ZrO over this system and the heat effect $\Delta H_{2190} = 144.3$ kcal/mole of the reaction



The recalculation of the heat effect of this reaction to absolute zero carried out by the authors of [1108] gives $\Delta H_0 = 151 \pm 6$ kcal/mole. Recalculation of the data of Chupka, Berkowitz and Inghram [1108],

based on the thermodynamic properties of the zirconium compounds, adopted in the present Handbook, led to $\Delta H_0 = 153$ kcal/mole. To this value corresponds the heat of formation of zirconium monozide $\Delta H^\circ f_0$ (ZrO, gas) = 23 kcal/mole.

Chupka, Berkowitz and Inghram [1108], using the measured ionic currents of Zr^+ , Zr^{2+} and ZrO_2^+ , calculated the equilibrium constant $K_{P_{2408}} = 1000$ for the reaction



To this value of the equilibrium constant corresponds the heat of the reaction $\Delta H_0 = -7.7$ kcal/mole* and the heat of formation of zirconium monozide $\Delta H^\circ f_0 = 27$ kcal/mole. The last value is less reliable because the calculation was based on the heat of sublimation of zirconium dioxide which is not known with sufficient accuracy.

The Handbook adopted the heat of formation of zirconium monoxide

$$\Delta H^\circ_f(ZrO, \dots) = 23 \pm 6 \text{ kcal/mole,}$$

calculated by means of the results of Chupka, Berkowitz and Inghram [1108]. The selected value is confirmed by the results obtained by processing the experiments of Ackerman and Thorn [488, 1013] in which the vapor pressure of zirconium dioxide was measured (see page 1953). The high degree of dissociation of zirconium oxide under the experimental conditions of these authors enables the heat of formation of zirconium monoxide to be determined as $\Delta H^\circ f_0$ (ZrO, gas) = 20.5 kcal/mole, if the value 177 kcal/mole is accepted for the heat of sublimation of ZrO_2 .

To the adopted value of the heat of formation of zirconium monoxide corresponds

$$D_0(ZrO) = 180,987 \pm 8 \text{ kcal/mole.}$$

ZrO₂ (cryst.). The heat of formation of zirconium dioxide, found in earlier works [4197, 3528, 3722, 3519, 3054, 3524] by the method

of combustion of metallic zirconium is inaccurate because the specimens were insufficiently pure. Moreover, the accuracy of the results was reduced in consequence of the fact that the combustion was carried out in presence of substances which relieve the combustion (paraffin oil, cellophane). The works of Roth and Becker [3519], Neumann, Kroger and Kunz [3054] and Roth, Borger and Siemonsen [3524], in which the values -258.1, -258.2 and -258.8 kcal/mole respectively, had been found, received most recognition in the thermochemical literature.

Humphrey [2156] determined the heat of combustion of a fairly pure (99.67%) zirconium sample which burned completely without addition of substances to facilitate combustion. The heat of formation of zirconium dioxide found in this work

$$\Delta H_{298.15}^{\circ}(\text{ZrO}_2, \text{cryst.}) = -261,5 \pm 0,2 \text{ kcal/mole}$$

is the best of those in the literature and has been adopted in the Handbook.

ZrO₂ (gas). Hoch, Nakata and Johnston [2090] measured the vapor pressure over ZrO₂ and over a Zr + ZrO₂ mixture (2014-2290°K) by the effusion method. Based on the fact that the vapor pressure was the same in both cases, the authors arrived at the conclusion that the main evaporation product is ZrO₂. To the vapor pressures of zirconium dioxide found in the work [2090] corresponds $\Delta H_{s_0} = 146$ kcal/mole. However, the measurements carried out by the authors of [2090] proved to be wrong because the main reaction in both cases was the reduction of zirconium dioxide by the metallic tantalum, of which the effusion cell was made [932, 1108].

Chupka, Berkowitz and Inghram [1108] carried out a mass spectrometric investigation of the composition of zirconium dioxide vapor. The investigation showed that ZrO is the main constituent of the vapor during evaporation. Using the relation of the logarithm of the

ZrO₂⁺ ion current and the temperature (2200-2500°K), the authors found for the heat of sublimation of zirconium dioxide the value $\Delta H_{s_{2350}} = 171 \pm 4$ kcal/mole, as the average of several test series carried out with effusion chambers of thoria and zirconia. Recalculation of this value to 0°K on the basis of the data in the present Handbook gave the value $\Delta H_{s_0} = 179$ kcal/mole.*

Chupka et al [1108] calibrated the apparatus by evaporating a known quantity of silver and were thus enabled to calculate the absolute partial pressures of ZrO₂ in different test series (seven points within the range 2331-2480°K). By means of these data they calculated the heat of sublimation of zirconium dioxide $\Delta H_{s_{2405}} = 169$ kcal/mole. A calculation based on the thermodynamic functions adopted in the Handbook, gave the value $\Delta H_{s_0} = 177$ kcal/mole.

According to the data obtained by the effusion method by Ackerman and Thorn [488], the vapor pressure over solid zirconia is described by the equation $\lg p_{(atm)} = 8.30 - 3.57 \cdot 10^4 T^{-1}$, and the heat of sublimation of ZrO₂ is $\Delta H_{s_{2584}} = 163$ kcal/mole (to this corresponds $\Delta H_{s_0} = 172$ kcal/mole). The mass spectrometric measurements [1108] and the thermodynamic calculations show that under the experimental conditions of the authors [488] zirconium dioxide vapor should contain a considerable quantity (50-70%) ZrO (gas). Hence the calculations of the experiments of Ackermann and Thorn [488] cannot give independent values of the heat of sublimation of ZrO₂ or the heat of formation of ZrO. These calculations showed the following relation between these magnitudes (in kcal/mole):

$\Delta H_f(\text{ZrO}_2 \text{ cryst.})$	$\Delta H_f(\text{ZrO gas})$
174	24,6
175	23,1
177	20,5
179	19,1

Thus the results of the calculations on the experiments of Ackermann and Thorn [488] are in good agreement with the results obtained on the basis of the measurements of Chupka, Berkowitz and Inghram [1108]: $\Delta H^{\circ}f_0(\text{ZrO}, \text{ gas}) = 23 \text{ kcal/mole}$ and $\Delta Hs_0(\text{ZrO}_2, \text{ cryst.}) = 177$ and 179 kcal/mole.

The heat of sublimation of zirconium dioxide which is the average of the values calculated from the measurement results [1108, 488]

$$\Delta Hs_0(\text{ZrO}_2, \text{ cryst.}) = 177 \pm 6 \text{ kcal/g-atom,}$$

has been adopted in the Handbook. The uncertainty in this value has been chosen on the basis of the estimate of Chupka, Berkowitz and Inghram [1108]. To the adopted value of $\Delta Hs_0(\text{ZrO}, \text{ cryst.})$ correspond

$$\begin{aligned} \Delta H^{\circ}f_0(\text{ZrO}_2, \text{ ...}) &= -83,212 \pm 6 \text{ kcal/mole,} \\ D_0(\text{ZrO}_2) &= 346,186 \pm 8 \text{ kcal/mole.} \end{aligned}$$

The standard state of lead is Pb (cryst.).

Pb (gas). Table 297 gives a summary of the literature data on the vapor pressure of lead. The heat of sublimation of lead calculated on the basis of these data is given in the last column of the Table.

The most reliable data are those of Rodebush and Dixon [3466, 3467] obtained by two different methods (method of boiling point determination and static method) which give the value $\Delta Hs_0 = 47.0 \text{ kcal/g-atom}$. The same value is obtained by calculation based on the boiling point of lead, found by Fischer [1563]. Similar values for the heat of sublimation of lead were calculated on the basis of the data of Egerton [1468] (47.4 kcal/g-atom*), Harteck [1965] (46.6 kcal/g-atom). The Handbook adopted the heat of sublimation of lead

$$\Delta Hs_0(\text{Pb}, \text{ 1 cryst.}) = \Delta H^{\circ}f_0(\text{Pb}, \text{ ...}) = 47,0 \pm 0,3 \text{ kcal/g-atom.}$$

PbO (cryst., red). Bichowsky and Rossini [813], based on the measurements of Thomsen [3981] of the heat of solution of lead oxide in nitric acid and the heat of precipitation of lead chloride by potas-

sium chloride, calculated -52.91 kcal/mole for the heat of formation of lead monoxide.

TABLE 297

Results of the Calculation of the Heat of Sublimation of Lead

Авторы A	Год B	Метод C	Интервал температур, °K D	Число измерений E	$\Delta H_{\text{с}}$ ккал/г-атом F
Гринвуд [1852] . . . G . .	1911	Кипения R	1588—1770	3	$43,2 \pm 0,5$
Вартенберг [4152] H . .	1913	Протока S	1081—1451	3	$46,1 \pm 0,2$
Инголд [2172] . . . I . .	1922	Кипения R	1196—1611	57	$45,8 \pm 0,5$
Эджертон [1468] . . . K . .	1923	Эффузионный T	800—1048	23	$47,4 \pm 0,2$
Родешуш, Диксон [3466, 3467] L . .	{ 1925 1925	{ Кипения R Статический U	{ 1433—1601 1391—1512	{ 21 8	{ $47,0 \pm 0,02$ $46,97 \pm 0,03$
Хартек [1965] . . . M . .	1928	Эффузионный T	1353—1477	4	$46,6 \pm 0,1$
Руфф, Бергдал [3551] N	1929	Кипения R	1548—1828	6	$43,6 \pm 0,5$
Лейтгебель [2588] . . . O	1931	•	2013	1	46,7
Баур, Бруннер [691] P	1934	•	1329—1673	4	$45,8 \pm 1,2$
Фишер [1563] Q	1934	•	2028	1	47,0

- A) Authors
- B) year
- C) method
- D) temperature range
- E) number of measurements
- F) kcal/g-atom
- G) Greenwood
- H) Wartenberg
- I) Ingold
- K) Egerton
- L) Rodebush, Dixon
- M) Harteck
- N) Ruff, Bergdahl
- O) Leitgebel
- P) Baur, Brunner
- Q) Fischer
- R) boiling
- S) flow
- T) effusion
- U) static

A more accurate value for the heat of formation of lead monoxide can be calculated on the basis of measurements carried out by several authors [3779, 2184, 3622, 286] of the e.d.s. of voltaic cells in which reactions involving lead oxide are taking place.*

Smith and Woods [3779], Ischikawa and Shibata [2184] and Makolkin

[286] measured the e.d.s. of an element in which the reaction



took place and found values of $\Delta G_{298.15}^\circ$ of -11.51, -11.64 and -11.16 kcal/mole, respectively. On the basis of these data and the standard entropies of the reaction components the heat of formation of lead oxide in the red crystalline modification, equal to -52.40, -52.27 and -52.75 kcal/mole, respectively, was calculated.

Measurements of the e.d.s. of elements were carried out at different temperatures in the works [3779, 286] which made it possible to calculate the heat of the reaction (XXX.3) and the corresponding heat of formation of lead monoxide -52.40 and -51.72 kcal/mole by means of the Gibbs-Helmholtz equation.

Spencer and Mote [3822] measured the e.d.s. of an element in which reduction of mercuric oxide (red modification) by metallic lead took place with formation of the red or yellow modification of lead monoxide. Recalculation of the values obtained by the authors of the work [3822] using the more accurate thermodynamic properties of mercuric oxide [1193, 917, 3508] gives values for the heat of formation of lead monoxide of -51.41 kcal/mole (red modification) and -51.08 kcal/mole (yellow modification). On the basis of the results of the investigation [562] which had been carried out somewhat earlier by the same method, the values -51.84 kcal/mole (red modification) and -51.31 kcal/mole (yellow modification) were obtained.

The most reliable results are those of the measurements [3779, * 286, 2184], on the basis of which the value

$$\Delta H_{\text{f, m. u.}}^\circ (\text{PbO}, \text{cryst.}, \text{red}) = -52.4 \pm 0.5 \text{ kcal/mole.}$$

has been adopted in the Handbook for the heat of formation of the red modification of lead monoxide.

The heat of transformation of the red modification of PbO into

the yellow modification is used in the selection of the numerical values of the thermodynamic functions of lead monoxide in the condensed state (see preceding Section). This value has been calculated not only on the basis of the results obtained in the measurements of the e.d.s. of the corresponding elements: $\Delta H_{298.15} = 327$ [3822] and 527 cal/mole [562], but also on the basis of the determination of the solubility of this modification in caustic solution, Randall and Spencer [3368a] found by this method* the value 348 cal/mole and Garrett et al [1654] 465 cal/mole. On the basis of these measurements, the value $\Delta H_{298.15} = 0.4 \pm 0.1$ kcal/mole has been adopted for the heat of transformation of the red modification of lead monoxide into the yellow modification. The same value has been recommended in the work of Millar [2912] and in the Handbook of the U. S. Bureau of Standards [3508].

To the adopted values of the heat of transformation and heat of formation of the red modification corresponds

$$\Delta H_{298.15}^{\circ}(\text{PbO, cryst., yellow}) = -52.0 \pm 0.51 \text{ kcal/mole.}$$

PbO (gas). Table 298 gives the heat of sublimation of PbO, calculated by using the thermodynamic functions adopted in the present Handbook based on the results of measurements of the saturation pressure of lead monoxide vapor.** The data of Feiser [1545] are not reliable because of the imperfection of the method used for the measurement of the vapor pressure. The results of the effusion measurements, carried out by An. Nesmeyanov et al [316, 315] have a large scatter in consequence of the reaction of the PbO with the effusion chamber materials. The results of the measurements by the flow method, obtained by An. Nesmeyanov et al [316, 315] and by Horbe and Knacke [2122] and also the results of the measurements by the method of evaporation from a surface [316, 315] are in satisfactory agreement and give the mean heat of sublimation of the red modification of lead monoxide

$$\Delta H_s(\text{PbO, } \text{cryst. , red}) = 64,0 \pm 1,0 \text{ kcal/mole.}$$

This value has been adopted in the Handbook. To it correspond

$$\Delta H_f^0(\text{PbO, } \text{cryst.}) = 12,104 \pm 1,1 \text{ kcal/mole,}$$

$$D_0(\text{PbO}) = 93,883 \pm 1,2 \text{ kcal/mole.}$$

TABLE 298

Results of the Calculation of the Heat of Sublimation of the Red Modification of Lead Monoxide

Авторы A	Год B	Метод C	Интервал температуры, °K D	Число измерений E	ΔH_s , ккал/моль F
Фейзер [1545] G. . . .	1929	a	1023—1473	10	62,0±0,5
Хёрбе, Кнакке [2122] H.	1959	К Протока	1198—1500	10	63,3±0,7
	1959	»	1055—1153	15	64,0±0,5
Ан. Н. Несмеянов, Фирсова, Исакова [316, 315] I	1959	L Испарения с поверхности	887—1043	14	64,6±0,5
	1959	M Эффузионный	1029—1151	6	64,1±1,6

a) Feiser measured the pressure of saturated lead monoxide vapor by the weight loss of a platinum boat containing PbO, heated in an inert gas atmosphere.

- A) Authors
- B) year
- C) method
- D) temperature range
- E) number of measurements
- F) kcal/mole
- G) Feiser
- H) Horbe, Knacke
- I) An. N. Nesmeyanov, Firsova, Isakova
- K) flow
- L) evaporation from a surface
- M) effusion

Linear extrapolation of the vibrational energy levels of the molecule PbO in the ground state $X^1\Sigma$ gives $D_0(\text{PbO}) = 100 \text{ kcal/mole}$. Erhard and Norrish [1489a] pointed out that Sugden and Bulewicz had found approximately the same value on the basis of preliminary data. These values are considerably less accurate than those calculated on the basis of the thermodynamic data.

PbF (gas). Linear extrapolation of the vibrational energy levels

of the molecule PbF for the ground state $X_1^2\Pi_{1/2}$ on the basis of the constants given in Table 289, gives $D_0(\text{PbF}) = 79$ kcal/mole. By studying the predissociation of the molecule PbF in the state $B^2\Sigma$, Wieland and Newburgh [4260] found the limit of the possible values of the dissociation energy $D_0 \leq 82$ kcal/mole. In a later work [4261] the same authors improved the dissociation energy, calculating $D_0 = 74$ kcal/mole. On the basis of these works, Gaydon [1668] recommended the value

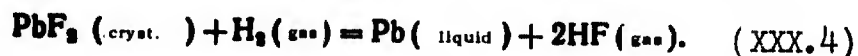
$$D_0(\text{PbF}) = 74 \text{ kcal/mole,}$$

which has been adopted in the Handbook. The uncertainty in this value, as given by Wieland and Newburgh [4260], is ± 4 kcal/mole. To this value of the dissociation energy corresponds

$$\Delta H_f^\circ(\text{PbF, gas}) = -8.5 \pm 4.1 \text{ kcal/mole.}$$

PbF_2 (cryst.)_ Guntz [1898, 1899] measured the heat of precipitation of lead fluoride from a solution of lead nitrate. Based on these data, Bichowsky and Rossini [813] calculated the heat of formation of lead difluoride -159.5 kcal/mole.

Jellinek and Rudat [2220] investigated the equilibrium



By means of the equilibrium constants found in this work and using the thermodynamic potentials adopted in the present Handbook, the heat of formation of lead difluoride was calculated as -158.6 kcal/mole.

The data obtained by Domange [1361] in the study of the equilibrium



are not entirely accurate because the calculation of the heat of this reaction by means of the temperature dependence of the equilibrium constants and using the thermodynamic potentials gives values which differ by ± 10 kcal/mole.

Wartenberg [4155] measured the heat of solution of the yellow mod-

ification of PbO in hydrofluoric acid and found for the heat of formation of lead difluoride -159.3 ± 0.3 kcal/mole. Application of a correction by taking into account the more precise value of the heat of formation of PbO (see page 1958) gives -159 kcal/mole.

Brewer [1093], based on the data [3678] on the solubility of lead difluoride in water, calculated the value $\Delta H_{298.15}^{\circ}(\text{PbF}_2, \text{cryst.}) = -158.1$ kcal/mole.

Ivett and DeVries [2195] measured the e.d.s. of an element in which the reaction



took place, at 15, 25 and 35°C and found by means of the Gibbs-Helmholtz equation for the heat of this reaction values of -112.39 to -112.09 , depending on the concentration of the NaF solution. It follows from these data that the heat of formation of lead difluoride is -160.4 ± 1.0 kcal/mole. The results of the work of Ivett and De Vries [2195] are the most reliable. The Handbook adopted

$$\Delta H_{298.15}^{\circ}(\text{PbF}_2, \text{cryst.}) = -160.4 \pm 1.0 \text{ kcal/mole.}$$

PbF₂ (gas). The saturation pressure of PbF₂ vapor has been measured by Wartenberg and Bosse [4161] by the method of boiling point determination (1351-1562°K) and by An. N. Nesmeyanov and Iofa [311] by the effusion method (792-988°K). Calculation of the heat of sublimation of PbF₂ at 0°K on the basis of these data gives the values 54.4 ± 0.5 kcal/mole, respectively. The heat of sublimation calculated on the basis of $\lg p$ as a function of $1/T$, found in the work [4161], with subsequent recalculation to 0°K gives the value $\Delta H_{s_0} = 54.8$ kcal/mole. An analogous calculation on the basis of the data of the work [311] cannot be carried out with sufficient reliability owing to the strong scatter of the experimental points. The mean of the three preceding values has been adopted in the Handbook for the heat of sublimation of

PbF₂:

$$\Delta H_s(\text{PbF}_2, \text{cryst.}) = 54,8 \pm 2 \text{ kcal/mole,}$$

the uncertainty of which is mainly determined by the inaccuracy of the thermodynamic functions of gaseous and condensed PbF₂.

To the adopted heat of sublimation correspond

$$\begin{aligned} \Delta H^{\circ} f_0(\text{PbF}_2, \text{gas}) &= -105,645 \pm 2,2 \text{ kcal/mole,} \\ D_0(\text{PbF}_2) &= 189,645 \pm 2,5 \text{ kcal/mole.} \end{aligned}$$

The standard state of mercury is Hg (liq.).

Below the melting point 243.29°K, the standard state is mercury in the crystalline state.

Hg (gas). Busey and Giauque [1035], based on the data of Menzies [2853] and Beattie and co-workers [702] on the saturation pressure of mercury vapor calculated the heat of evaporation of mercury at the boiling point, equal to $\Delta H_{629,88} = 14.127 \text{ kcal/g-atom}$. The thermodynamic functions of liquid mercury, calculated by the authors of [1035] on the basis of the results of their own measurements of the specific heat of mercury (15-330°K) and the data of Douglas, Ball and Ginnings [1386] on the enthalpy of liquid mercury above 273°K were used in these calculations. Recalculation of the heat of evaporation of mercury to 0°K gives the value

$$\Delta H_s(\text{Hg, cryst.}) = \Delta H^{\circ} f_0(\text{Hg, gas}) = 15,40 \pm 0,03 \text{ kcal/g-atom,}$$

which has been adopted in the Handbook.

The data of Spedding and Dye [3816] on the vapor pressure of mercury (250-360°C) and the data of Smith and Wolcott [3794] on the specific heat* of mercury at very low temperatures (1.3-20°K) were published in the years 1955-1956. Making allowance for the results of these works does not have any marked effect on the value adopted in the Handbook.

HgF (gas). Linear extrapolation of the vibrational energy levels of

the molecule HgF, carried out on the basis of the adopted molecular constants (Table 289), gives the dissociation energy

$$D_0(\text{HgF}) = 40 \pm 7 \text{ kcal/mole.}$$

This value, which is in agreement with that recommended by Herzberg [2020] and Rosen [649] has been adopted in the Handbook. To the adopted dissociation energy corresponds

$$\Delta H_f^\circ(\text{HgF, ...}) = -6.1 \pm 7 \text{ kcal/mole.}$$

TABLE 299

Adopted Values (in cal/mole) of the Thermochemical Properties of Zirconium, Mercury and Their Compounds

Вещество A	Состояние B	D, или ΔH_a , C	ΔH_f°	$\Delta H_f^\circ /_{298.15}$	$\Delta H_f^\circ /_{298.15}$	$H_{298.15}^\circ - H_0^\circ$	$H_{298.15}^\circ - H_0^\circ$
Zr	Крист. D	145 000 ^a	0	0	0	1283	1313
Zr	Газ	—	145 000	145 314	145 316	1597	1629
ZrO	» E	180 987	23 000	22 627	22 636	1930	1986
ZrO ₂	Крист. D	177 000 ^a	-280 212	-281 502	-281 500	2033	2100
ZrO ₂	Газ	346 186	-83 212	-83 933	-83 942	2602	2658
Pb	Крист. D	47 000 ^a	0	0	0	1613	1644
Pb	Газ	—	47 000	46 843	46 837	1456	1481
PbO	Крист., Красная F	64 000 ^a	-51 896	-52 407	-52 400	2122	2177
PbO	Газ	93 883	12 104	11 574	11 565	2103	2142
PbF	»	74 000	-8 500	-8 977	-8 986	2173	2214
PbF ₂	Крист.	54 800 ^a	-160 445	-160 419	-160 400	3712	3800
PbF ₂	Газ	189 645	-105 645	-106 422	-106 430	2909	2970
Hg	Жидк. G	18 400 ^a	0	0	0	2199	2232
Hg	Газ	—	18 400	14 657	14 649	1456	1481
HgF	»	40 000	-8 100	-7 153	-7 164	2183	2224

- a) The heat of sublimation is given
 b) solid up to the melting point 234.29°K.
 A) Substance
 B) state
 C) or
 D) cryst.
 E) gas
 F) red
 G) liq.

- 1927 In the work of Rochester, the first excited state of PbF has been regarded as the state $^2\Delta$. Barrow and co-workers [647] have shown recently that the first excited state of the molecules SiF, GeF, SnF and PbF is the state $^2\Sigma$.
- 1931 This relation has been used on the basis of the data given in Table 40 of the monograph by Herzberg [52].
- 1933 It should be pointed out that at $T > 6000^\circ\text{K}$ the corresponding differences begin to increase rapidly.
- 1947 For references to the work on the measurement on the enthalpy of PbO at temperatures to 550°K , see Kelley [2363].
- 1949 The value $S_{298.15}^\circ = 29$ cal/mole-degree, recommended by Rossini et al [3508] is obviously slightly too high.
- 1950 The authors of the work [85] recommend for the heat of melting of PbF_2 the value 3.0 kcal/mole, while calculation of this value on the basis of the equations for the enthalpy of solid and molten PbF_2 , carried out in the same work, gives 2.7 kcal/mole. The heat capacity of molten PbF_2 , $C_p^\circ = 26.97$ cal/mole-degree, calculated by the authors of [85] is obviously a little too high.
- 1953 The authors of the work [1108] calculated the considerably larger value $\Delta H_0 = -14.4$ kcal/mole. The reasons for this discrepancy are not clear (see footnote page 1955).
- 1955 Chupka, Berkowitz and Inghram [1108] recalculated this value to absolute zero and obtained the much larger value $\Delta H_{s,0} = 188.2$ kcal/mole. Approximately the same differences were found in the recalculations of the other thermoal effects, measured by Chupka et al [1108]. The reason for these differences is not clear because practically the same initial data were used for the calculation of the thermodynamic functions of ZrO_2 (gas) and ZrO_2 (cryst.) in the Handbook and in the work [1108], and the entropy of sublimation of ZrO_2 calculated by the authors of [1108] and the variation of the entropy in the reaction (XXX.3) coincided with that calculated on the basis of the Handbook data.
- 1956 The vapor pressures obtained by Egerton [1468] are somewhat low because of the large dimensions of the effusion orifice.
- 1957 Fried [1613] carried out the measurements only at 0°C , and the value found by him was not used for this reason for the calculation of the heat of formation of lead monoxide.

- 1958 The good agreement between the values calculated by means of the Gibbs-Helmholtz equation and the entropy of the modification of PbO, clearly attests to the fact that the results of the work of Smith and Woods [3779] are the most accurate.
- 1959 The original works give the variations of the isobaric potential ($\Delta G_{298.15}^{\circ}$) upon transition of the red modification of lead monoxide into the yellow one. The heats of transformation given in the text were calculated on the basis of these values and the standard entropies of the modifications of lead monoxide as adopted in the Handbook.
- 1959 Under neutral conditions, lead monoxide evaporates without dissociation into its elements, which has been confirmed by the thermodynamic calculations as well as experimentally. An. Nesmeyanov, Firsova and Isakova [316] found that the results of the measurements of the vapor pressure of lead monoxide by the flow method are independent of the nature of the gas used as carrier: nitrogen, air or oxygen.
- 1963 Kostryukov and Strelkov [253] measured the heat capacity of mercury within the range 210-240°K and demonstrated the absence of any marked effect of heterophase fluctuations on the heat capacity of pure mercury even within 0.01° of the melting point of mercury (234.29°K) [1035].

[Transliterated Symbols]

- 1957 э.д.с. = e.d.s. = elektrodvishushchaya sila = e.m.f. (electromotoric force).

Chapter 31

INERT GASES (He, Ne, Ar, Kr, Xe) AND THE ELECTRON GAS (e^-)

§123. THERMODYNAMIC FUNCTIONS OF THE NOBLE GASES

Because the noble gases (He, Ne, Ar, Kr and Xe) have completed electron shells, the ground state of these atoms is the 1S state, and the first excited states are characterized by a high excitation energy (159, 850.318; 134,043.790; 93143.800; 79,972.535 and 67,068.047 cm^{-1} for He, Ne, Ar, Kr and Xe, respectively [2941]). Hence, in the calculation of the thermodynamic functions of the noble gases within the temperature range 293.15-6000°K, the electronic component was assumed to be zero and the calculation was carried out by means of the equations for the translational components in the values of Φ_T^* and S_T° [Eqs. (II.8)-(II.9)]; the corresponding constants A_Φ and A_S are given in Table 300. The uncertainty in the thermodynamic functions calculated in this manner for the noble gases is determined only by the inaccuracy of the fundamental physical constants and does not exceed ± 0.005 cal/g-atom·degree.

The thermodynamic functions of the noble gases, given in different Handbooks (see, for example [2142, 3680]), agree with those calculated in the present Handbook within the limits of the above-indicated uncertainty. The values of the thermodynamic functions of the noble gases, calculated by Kolsky, Gilmer and Gilles [2462], exceed those calculated by the Handbook authors by 0.005-0.01 cal/g-atom·degree in the values of Φ_T^* which is obviously explained by the fact that the atomic weights used in the work [2462] for the calculation were taken from the

physical scale.

TABLE 300

Values (in cal/g-atom·degree) of the Constants in Eqs. (II.8)-(II.9) for the Noble Gases

Вещество A	A_{Φ}	A_S
He	-3,14892	-1,81928
Ne	1,67356	6,64176
Ar	3,70843	8,67663
Kr	5,91714	10,88534
Xe	7,25571	12,22391

A) Substance

ro.

The thermodynamic functions of He, Ne, Ar, Kr, and Xe are given in the Tables 34, 35, 36, 37 and 38 of Volume II of the Handbook.

§124. THERMODYNAMIC FUNCTIONS OF THE ELECTRON GAS

The electron gas, as Sommerfeld [3810] was the first to show, is a degenerate Fermi - Dirac gas, and its properties differ from the properties of a gas which obeys the classic Boltzmann statistics the more, the lower its absolute temperature or the higher the density. In his work, Sommerfeld derived formulas for the calculation of the thermodynamic functions of the electron gas (for greater details, see the monograph of Bethe and Sommerfeld [91] and Mayer and Goppert-Mayer [285]). In contrast to the classical equations, these equations contain integrals which are not calculated in the final form. Tables of the values of these integrals have been compiled by McDougall and Stoner [2701] and also by Gordon [1806].

It should be pointed out that the formulas obtained by Sommerfeld, were derived for the case of the electronic gas in a metal, where the density of the gas is so high that its degeneration is considerable up

The calculation of the equilibrium constants of the ionization reaction of the noble gases has not been carried out in the present Handbook because the atoms of the noble gases have high ionization potentials.

As the standard state of the noble gases, the corresponding monatomic gases at a pressure of 1 atm has been taken. Hence the heats of formation of the noble gases are correspondingly zero.

to temperatures of $\sim 1,000,000^\circ\text{K}$. However, if we consider the free electron gas at a pressure of 1 atm, its density is fairly small and at temperatures above $1000\text{-}1500^\circ\text{K}$ such a gas can be considered with a high degree of accuracy as a non-degenerate classical Boltzmann gas. At lower temperatures the thermodynamic functions of the electron gas, calculated with allowance being made for state of degeneracy, differ considerably from those calculated by means of the classical formulas (the difference in the enthalpy value at 293.15°K amounts to approximately 800 cal/mole). However, when concrete problems connected with the calculation of the ionization equilibrium are considered, these differences in the numerical values of the thermodynamic functions are not of any practical importance, because the partial pressure of the electrons at moderate temperatures (up to 1500°K) is very low.*

In view of this, the thermodynamic properties of the electron gas were calculated in the present Handbook within the temperature range $293.15\text{-}20,000^\circ\text{K}$ for the nondegenerate ideal gas at a pressure of 1 atm. The calculation was carried out by means of Eqs. (II.3)-(II.4), where it has been assumed in correspondence with the two possible orientations of the spin of the electron that $\Phi_{\text{vn}}^* = S_{\text{vn}} = R \ln 2$, and the translational components of Φ_{ppst}^* and S_{fost}° were calculated by means of the relations (II.6)-(II.7) using the "atomic weight" of the electron (on the chemical scale) of $5.4861 \cdot 10^{-4}$ (see Appendix 1). Finally:

$$\begin{aligned}\Phi_r^\circ &= 11,43960 \lg T - 28,2870, \\ S_r^\circ &= 11,43960 \lg T - 23,3188.\end{aligned}$$

The thermodynamic functions of the electron gas have been calculated earlier by Huff, Gordon and Morrell [2142] within the temperature range $298.15\text{-}6000^\circ\text{K}$. As in the present Handbook, the calculation was carried out for the nondegenerate gas; hence the values of thermodynamic functions of the electron gas obtained by the authors of the work

[2142] agree completely with those given in Table 38 (II).

Manu-
script
Page
No.

[Footnotes]

1969 If necessary one can easily recalculate the partial pressure of the electrons, calculated by means of the Boltzmann statistics, to the pressure of the degenerate gas. The details of such a calculation have been described in the work of Gordon [1806].

[Transliterated Symbols]

1969 ВН = vn = vnutrenniy = internal

1969 ПОСТ = post = postupatel'nyy = translational

Part 3
APPENDIX

Appendix 1

ATOMIC WEIGHTS- ISOTOPE COMPOSITION AND NUCLEAR SPINS OF
THE CHEMICAL ELEMENTS TREATED IN THE HANDBOOK

The present Handbook treats the thermodynamic properties of 31 chemical elements and their compounds. The following values, characteristic for every chemical element, are used in the calculation of the Tables of the thermodynamic properties of these substances on the basis of the methods of statistical mechanics and thermodynamics:

- 1) the atomic weight of the element (average);
- 2) the concentration of the stable isotopes in the natural element;
- 3) the atomic weights of the isotopes;
- 4) the nuclear spins of the isotopes;

Table 301 gives these values for the chemical elements as adopted in the present Handbook.

The changes in the adopted values of the atomic weights of the elements, which have taken place during the years 1894-1949, are indicated in the Handbook [319]. During the years 1950-1958, the changes in the atomic weights of the elements were published in the reports of the Commission on Atomic Weights of the International Union for Pure and Applied Chemistry [4245-4249].

The atomic weights, recommended by the Commission in 1956 [4248] and left unchanged in 1958 [4249] have been adopted in the present Handbook. The same atomic weights were also adopted in the first Handbook edition. The atomic weights of the elements used in the Handbook are

given in column 4 of Table 301.

Column 5-8 of Table 301 gives data for the stable longlived isotopes of the elements treated in the Handbook. An exception is only hydrogen for which data are presented not only for the stable isotopes protium (H^1) and deuterium (H^2 or D) but also for the radioactive isotope tritium (H^3 or T) whose half life according to [3889] is 12.262 years. Of the radioactive but longlived isotopes Table 301 gives data for K^{40} , Rb^{87} and Zr^{96} , whose half lives according to [3889] are $1.25 \cdot 10^9$, $5.0 \cdot 10^{10}$ and $1.6 \cdot 10^{17}$ years, respectively.

Columns 6 and 8 of Table 301 give the data adopted in the Handbook on the percentage concentrations of the isotopes in the natural element in accordance with the data recommended in 1958 by Strominger, Hollander and Seaborg [3889]. Slight differences are due to rounding off of the figures so that the sum of the isotopes in the natural mixture in atomic percent should add up to 100%. Data on the nuclear spin are absent in the summary [3889] for some isotopes because they had not yet been determined on the basis of experimental studies. Such data are mostly lacking for the isotopes with even atomic number and mass number. According to the empirical rule [92] the spins of such nuclei are zero. The nuclear spins, marked with an asterisk in Table 301, have been determined not on the basis of experimental investigations, but have been taken as zero in accordance with this rule.

It should be pointed out that the experimental data on the isotope concentrations in different natural samples of the same element are not quite identical. Thus, for example, according to [3889] the concentration of the isotope H^1 in natural hydrogen varies within the limits of 99.9849 to 99.9861% and correspondingly the concentration of the isotope H^2 (D) varies within the limits of 0.0151 to 0.0139%. In different samples of natural boron the concentration of the isotope B^{10} varies

within the limits of 18.45 to 18.98% and, consequently, the concentration of the isotope B^{11} varies within the limits of 81.55 to 81.02%. The data on the concentration of the oxygen isotopes given in Table 301 correspond to their concentration in atmospheric oxygen according to the results of the mass spectrometric investigations of Nier [3094]. The summary [3889] indicates that the variation of the O^{16}/O^{18} ratio in nature does not exceed 4%.

Column 7 of Table 301 gives the atomic weights of the isotopes, recommended by Kravtsov [256-258] on the basis of a careful selection of the most accurate results, obtained in mass-spectrometric measurements and in measurements of the energies of nuclear reactions. Earlier reviews of the experimental results of determinations of the mass of the atoms of individual isotopes have been compiled by Dzhelepov and Zyryanova [179], Bainbridge [467] and others.

The atomic weights of the elements and their isotopes are normally expressed in slightly different units. The unit of atomic weight used in chemistry for the elements is 1/16th of the mean atomic weight of natural oxygen, while the unit of the atomic weight for isotopes is 1/16th of the atomic weight of the oxygen isotope O^{16} . Hence the terms chemical and physical scale of atomic weights. Since 1940 the International Commission on Atomic Weights has used the divisor 1.000275 to transform the data from the physical to the chemical scale (see [4248]). In the present Handbook, natural oxygen means atmospheric oxygen. It follows from the data given in Table 301 for oxygen and its isotopes that the mean atomic weight of oxygen on the physical scale is 1.000279 times greater than on the chemical scale. This ratio between the physical and chemical scales of the atomic weights has been used in the present Handbook.

TABLE 301

The Chemical Elements and Their Isotopes

Атомный номер A	Элемент B			F Стабильные изотопы			
	название C	символ D	атомный вес (химич. шкала) E	символ G	процентное содержание H	атомный вес (физич. шкала) I	спин ядра (в единицах $\frac{h}{2\pi}$) K
1	Водород	H	1,008	H ¹ L H ² или D H ³ или T	99,9851 0,0149 ~ 10 ⁻⁹	1,0081451 2,0147425 3,0170013	1/2 1 1/2
2	Гелий	He	4,003	He ³ He ⁴	0,00013 99,99987	3,0169807 4,0038761	1/2 0
3	Литий	Li	6,940	Li ⁶ Li ⁷	7,42 92,58	6,0170404 7,0182389	1 3/2
4	Бериллий	Be	9,013	Be ⁹	100	9,0150566	3/2
5	Бор	B	10,82	B ¹⁰ B ¹¹	18,45 81,55	10,0161236 11,0128153	3 3/2
6	Углерод	C	12,011	C ¹² C ¹³	98,892 1,108	12,0038156 13,0074900	0 1/2
7	Азот	N	14,008	N ¹⁴ N ¹⁵	99,635 0,365	14,0075256 15,0048783	1 1/2
8	Кислород	O	16,000	O ¹⁶ O ¹⁷ O ¹⁸	99,7587 0,0374 0,2039	16,0000000 17,0045374 18,0048850	0 5/2 0
9	Фтор	F	19,00	F ¹⁹	100	19,0044441	1/2
10	Неон	Ne	20,183	Ne ²⁰ Ne ²¹ Ne ²²	90,92 0,26 8,82	19,9987980 21,0005244 21,9983771	0 3/2 0
11	Натрий	Na	22,991	Na ²³	100	22,9970911	3/2
12	Магний	Mg	24,32	Mg ²⁴ Mg ²⁵ Mg ²⁶	78,60 10,11 11,29	23,9926891 24,9937819 25,9908538	0 5/2 0
13	Алюминий	Al	26,980	Al ²⁷	100	26,9901117	5/2
14	Кремний	Si	28,090	Si ²⁸ Si ²⁹ Si ³⁰	92,18 4,70 3,12	27,9858226 28,9857037 29,9832868	0 1/2 0
15	Фосфор	P	30,975	P ³¹	100	30,9836125	1/2
16	Сера	S	32,066	S ³² S ³³ S ³⁴ S ³⁶	95,018 0,750 4,215 0,017	31,9822388 32,9819473 33,9786635 35,9785253	0 3/2 0 [*] 0 [*]

Атомный номер A	Элемент B			F Стабильные изотопы			
	название C	символ D	атомный вес (химич. шкала) E	символ G	процентное содержание H	атомный вес (физич. шкала) I	спин ядра (в единицах $\frac{h}{2\pi}$) K
17	Хлор	Cl	35,457	Cl ³⁵ Cl ³⁷	75,4 24,6	34,979713 36,9776573	3/2 3/2
18	Аргон	Ar	39,944	Ar ³⁶ Ar ³⁸ Ar ⁴⁰	0,337 0,063 99,600	35,9789823 37,9748023 39,9750925	0 0* 0
19	Калий	K	39,100	K ³⁹ K ⁴⁰ K ⁴¹	93,08 0,01 6,91	38,9761000 39,9767119 40,9748565	3/2 4 3/2
20	Кальций	Ca	40,08	Ca ⁴⁰ Ca ⁴² Ca ⁴³ Ca ⁴⁴ Ca ⁴⁵ Ca ⁴⁶	96,97 0,64 0,144 2,06 0,003 0,183	39,9752931 41,9719681 42,9724429 43,9694714 45,9682984 47,967766	0 0* 7/2 0* 0* 0*
35	Бром	Br	79,916	Br ⁷⁹ Br ⁸¹	50,52 49,48	78,9434 80,94271	3/2 3/2
36	Криптон	Kr	83,80	Kr ⁷⁸ Kr ⁸⁰ Kr ⁸² Kr ⁸³ Kr ⁸⁴ Kr ⁸⁶	0,35 2,27 11,56 11,55 56,90 17,37	77,94517 79,94231 81,93946 82,9418 83,93827 85,93805	0* 0* 0 9/2 0 0
37	Рубидий	Rb	85,48	Rb ⁸⁵ Rb ⁸⁷	72,15 27,85	84,94007 86,93722	5/2 3/2
38	Стронций	Sr	87,63	Sr ⁸⁴ Sr ⁸⁶ Sr ⁸⁷ Sr ⁸⁸	0,56 9,86 7,02 82,56	83,9411 85,93697 86,93688 87,93389	0* 0 9/2 0
40	Цирконий	Zr	91,22	Zr ⁹⁰ Zr ⁹¹ Zr ⁹² Zr ⁹³ Zr ⁹⁴	51,46 11,23 17,11 17,40 2,80	89,93344 90,93470 91,93438 93,9364 95,9393	0* 5/2 0* 0* 0
53	Йод	I	126,91	I ¹²⁷	100	126,94503	5/2
54	Ксенон	Xe	131,3	Xe ¹²⁴ Xe ¹²⁶ Xe ¹²⁸	0,09 0,09 1,92	123,94553 125,94473 127,94433	0* 0* 0*

Атомный номер	Элемент			Стабильные изотопы			
	название	символ	атомный вес (химич. шкала)	символ	процентное содержание	атомный вес (физич. шкала)	спин ядра (в единицах $\frac{h}{2\pi}$)
54	Ксенон	Xe	131,3	Xe ¹²⁹	26,44	128,94574	1/2
				Xe ¹³⁰	4,08	129,94466	0*
				Xe ¹³¹	21,18	130,9465	3/2
				Xe ¹³²	26,89	131,94606	0
				Xe ¹³⁴	10,44	133,94778	0
				Xe ¹³⁶	8,87	135,95021	0
55	Цезий	Cs	132,91	Cs ¹³³	100	—	7/2
56	Барий	Ba	137,36	Ba ¹³⁰	0,10	129,94810	0*
				Ba ¹³²	0,10	131,94706	0*
				Ba ¹³⁴	2,42	133,94683	0
				Ba ¹³⁵	6,59	134,94845	3/2
				Ba ¹³⁶	7,81	135,94753	0
				Ba ¹³⁷	11,32	136,94906	3/2
				Ba ¹³⁸	71,66	137,94873	0
80	Ртуть	Hg	200,61	Hg ¹⁹⁶	0,14	196,0276	0
				Hg ¹⁹⁸	10,02	198,0290	0
				Hg ¹⁹⁹	16,84	199,0305	1/2
				Hg ²⁰⁰	23,13	200,0309	0
				Hg ²⁰¹	13,22	201,0330	3/2
				Hg ²⁰²	29,80	202,0336	0
				Hg ²⁰⁴	6,85	204,0369	0
82	Свинец	Pb	207,21	Pb ²⁰⁴	1,4	204,0363	0
				Pb ²⁰⁶	25,2	206,03872	0
				Pb ²⁰⁷	21,7	207,04050	1/2
				Pb ²⁰⁸	51,7	208,04160	0

- A) Atomic number
 B) element
 C) name
 D) symbol
 E) atomic weight (chemical scale)
 F) stable isotopes
 G) symbol
 H) percentage concentration
 I) atomic weight (physical scale)
 K) nuclear spin (in units of ...; L) or.
 1) Hydrogen
 2) helium
 3) lithium
 4) beryllium
 5) boron
 6) carbon
 7) nitrogen
 8) oxygen
 9) fluorine
 10) neon
 11) sodium

12) magnesium
 13) aluminum
 14) silicon
 15) phosphorus
 16) sulfur
 17) chlorine
 18) argon
 19) potassium
 20) calcium
 35) bromine
 36) krypton
 37) rubidium
 38) strontium
 40) zirconium
 53) iodine
 54) xenon
 55) cesium
 56) barium
 80) mercury
 82) lead

Attempts which have been made to unify the scale of atomic weights have not led to general agreement (on this question, see [4248, 4249, 2811]).*

Wapstra [4148] calculated the atomic weights of the chemical elements on the basis of the results of measurements of the isotope concentrations and masses of their atoms. The results of these calculations for most chemical elements practically coincide with the atomic weights recommended in the reports on the atomic Weight Commission [4248, 4249] and those adopted in the present Handbook. However, for certain elements, considerable differences were found. The greatest divergence from the recommendations of the Atomic Weight Commission was found by Wapstra for the following elements (of those treated in the Handbook): Ne(20.1722 ± 0.0008), Si(28.0874 ± 0.0010), Ar(39.94920 ± 0.00007), Br(79.9103 ± 0.0015), Ba(137.332 ± 0.005). The calculated atomic weights (on the chemical scale) are given in brackets.

Manu-
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Page
No.

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In 1960, the Council of the International Unions for Pure and Applied Chemistry [4233] adopted a resolution concerning the introduction of a new atomic weight unit, based on the integer 12 for the carbon isotope C^{12} which would be the same for physics and chemistry. The atomic weights on the chemical scale, based on the integer 16 for natural oxygen, must be divided by 1.000043 in order to transform them into the new atomic weight scale. This correction is practically of no importance in most cases. In view of this it has not been used in the present Handbook, whose numerical data have been prepared long before the introduction of the new scale of atomic weights.

Appendix 2

FUNDAMENTAL PHYSICAL CONSTANTS AND TRANSFORMATION FACTORS FOR THE UNITS OF ENERGY, VOLUME AND PRESSURE AS USED IN THE HANDBOOK

When relations are used which connect the thermodynamic functions with the molecular constants, it is necessary to know the numerical values of the following physical constants: h , the Planck constant; c , the velocity of light in vacuo; k , the Boltzmann constant; N , the Avogadro number; R , the universal gas constant. In addition, to calculate the thermodynamic functions on the basis of molecular data and in particular to determine the numerical values of the thermodynamic quantities on the basis of the results of experimental measurements one must know the numerical values of the transformation factors for the required energy units together with the adopted values of the fundamental physical constants.

All the calculations carried out in the preparation of the present Handbook, were based on a single system of physical constants (see Table 303) and on the transformation factors for the energy units (see Table 304).

1. Physical constants. For the physical constants, the present Handbook adopted the values recommended by Cohen, Du Mond et al in the works [1141-1143] on the basis of painstaking calculations in which the most accurate results of the measurement of different physical magnitudes and the theoretical relations, connecting them with the fundamental physical constants, were taken into account. At the same time the

analogous works of Bearden and Thomsen [700, 701] were published, in which values were obtained for the physical constants which agree within the limits of determination error with the values of the respective constants in the above-mentioned works of Cohen and DuMond.

The constants h , c , k , N , R recommended by Cohen and DuMond [1141-1143] and Bearden and Thomsen [700, 701] are compared in Table 302 with the numerical values used previously for these constants.

The most accurately determined physical constants are: the fine structure constant α , the electric charge e of the electron and the speed of light in vacuo c . Hence, Planck's constant h has been determined in the works [1141-1143, 700, 701] via the constants α , e and c by means of the relation

$$h = \frac{2\pi e^2}{\alpha c}.$$

The following numerical values have been used for the constants α and e in the works [1141-1143]:

$$\begin{aligned} \alpha &= (7,29729 \pm 0,00003) \cdot 10^{-3}, \\ e &= (4,80286 \pm 0,00009) \cdot 10^{-10} \text{ CGSE.} \end{aligned}$$

The constant α has been determined on the basis of the most accurate results of the measurements of the hyperfine structure of the spectral lines of hydrogen and deuterium, and the constant e on the basis of the most accurate determinations of the Faraday number $F = N \cdot e$ and the Avogadro number N .

For the velocity of light c the value

$$c = 299\,793,0 \pm 0,3 \text{ km/sec,}$$

has been recommended in the works [1141-1143], obtained in 1954 by Froome [1620] on the basis of the results of precision measurements by means of a microwave interferometer designed by this researcher.

The Avogadro number N has been determined in the works [1141-1143, 700, 701] on the basis of the results of the most accurate x-ray dif-

fraction measurements of the parameters of the crystal lattices with allowance being made for the results of other physical magnitudes.

TABLE 302

Values of the Fundamental Physical Constants, Used for the Calculation of the Thermodynamic Properties of Substances on the Basis of the Molecular Constants

Постоян- ная A	Единица измерения B	Интернац. критичес- кие табли- цы 1926 г. C [2179]	Бердз 1941 г. D [1171]	Дю-Мон и Кохн 1950 г. E [1415, 1416]	Россини и др. 1952 г. F [3563]	Дю-Монд и Коэн 1953 г. E [1417]	Бердз и Томсен 1955— —1957 гг. G [700, 701]	Кохн, Дю- Мон и др. 1955— —1957 гг. H [1141—1143]
h	10^{-7} эрг·сек I	6,551	6,6242	6,62377	6,62377	6,6252	6,6254	6,62517
c	10^{10} см/сек K	2,99840	2,99776	2,997902	2,997902	2,997929	2,997928	2,997930
N	10^{23} молекул/моль L	6,061	6,02283	6,02544 ^a	6,02380	6,02472 ^a	6,0247 ^a	6,02486 ^a
k	10^{-16} эрг/град M	1,372	1,380474	1,38026	1,380257	1,38042	1,38049	1,38044
R	дж/моль·град N	8,315	8,31436	—	8,31439	8,31662 ^a	8,3170 ^a	8,31696 ^a
	кал/моль·град O	1,9369	1,986467	1,98719	1,98719	—	—	1,98726

a) Related to one mole on the physical scale of atomic weights.

A) Constant

B) measurement unit

C) international critical Tables of 1926

D) Berdzh

E) DuMond and Cohen

F) Rossini et al.

G) Bearden and Thomsen

H) Cohen, DuMond et al

I) erg·sec

K) cm/sec

L) molecules/mole

M) erg/degree

N) joule/mole·degree

O) cal/mole·degree

One of the least accurately known physical constants is the universal gas constant R . It is determined on the basis of the equation for the ideal gas state by means of the following relation:

$$R = \frac{1}{T_0} \lim_{p \rightarrow 0} (pV)_0,$$

where $(pV)_0$ is the product of the pressure and the volume of one mole of any real gas at the temperature T_0 , regarded as a function of the pressure p . At a temperature of 0°C , the magnitude of $(pV)_0$ has been

measured at low pressures by numerous investigators for different gases. An analysis of the measurements, carried out before 1940 for He, Ne, Ar, H₂, N₂ and O₂ was carried out by Cragoe [1209]. The most accurate experimental measurements were carried out on oxygen. An analysis of the results of these measurements enabled Cragoe to determine the following limit value of pV at 0°C*:

$$\lim_{p \rightarrow 0} (pV)_0 = 22,4140 \pm 0,0003 \text{ l} \cdot \text{atm/mole} = 22414,6 \pm 0,3 \text{ cm}^3 \cdot \text{atm/mole} = \\ = 2271,16 \pm 0,03 \text{ joules/mole.}$$

The limit value of $(pV)_0$ determined in the work [1209] has been adopted in all subsequently published reviews of the physical constants. However, Berdzh [117] proposed twice the uncertainty than that given by Cragoe [1209] for the value of $\lim_{p \rightarrow 0} (pV)_0$ found in the work [1209] which has also been used in the works of Cohen and DuMond [1141-1143].

Until recently, the magnitude of T_0 , the temperature 0°C on the thermodynamic (absolute) scale, has been determined on the basis of experimental measurements of the thermal coefficients of the isobaric volume variation and the isochronic pressure variation of gases. On the basis of the results of experimental investigations of this kind, carried out by Berdzh [117], the value $T_0 = 273.160 \pm 0.010^\circ\text{K}$ has been recommended. In 1954, the 10th International Conference on Weights and Measures [1159] adopted a new determination of the thermodynamic temperature scale by means of a single fixed point, the ternary point of water, whose temperature is assumed to be exactly 273.16°K . As the melting point of ice at atmospheric pressure is 0.01° lower than the ternary point, $T_0 = 273.15^\circ\text{K}$ corresponds to the thermodynamic temperature scale established by the 10th International Conference.

The universal gas constant given in Table 303, as adopted in the present Handbook, is based on the corresponding value of $\lim_{p \rightarrow 0} (pV)_0$, recommended in the work of Cragoe [1209] and the value of T_0 , determined

by the 10th International Conference on Weights and Measures.

TABLE 303

Physical Constants Adopted in the Handbook

Постоянная A	Единица измерения B	Значение постоянной C
c	см/сек D	$(2,997930 \pm 0,000003) \cdot 10^{10}$
h	эрг·сек E	$(6,62517 \pm 0,00023) \cdot 10^{-27}$
N	молекула/моль F	$(6,02318 \pm 0,00021) \cdot 10^{23}$
k	эрг/град G	$(1,38044 \pm 0,00007) \cdot 10^{-20}$
R	кал/моль·град H	$1,98726 \pm 0,00005$
$R \ln 10$	кал/моль·град H	$4,57584 \pm 0,00011$
$\frac{R}{T^2} \frac{dT}{dT}$	моль·град/кал I	$0,218539 \pm 0,000006$
hc/h	см·град K	$1,43879 \pm 0,00011$

- A) Constant
- B) measurement unit
- C) numerical value of the constant
- D) cm/sec
- E) erg·sec
- F) molecule/mole
- G) erg·degree
- H) cal/mole·degree
- I) mole·degree/cal
- K) cm·degree

Table 303 gives also the values of several derivative constants which have been adopted in the present Handbook.

2. Relations between the energy units. When the thermodynamic properties of substances are calculated on the basis of the experimental results of spectroscopic, calorimetric, mass spectrometric and other studies, one has to deal with quantities of energy, expressed in different units. The traditional unit of energy measurement in calorimetric studies is the calorie, while the traditional energy units in spectrometric investigations is the reciproke centimeter and in mass spectrometric investigations, the electron volt. The erg and joule = 10^7 erg have long been traditional in mechanics as basic energy units. Whereas, the relations between the reciproke centimeter and erg, and electron volt and erg, are determined only by the values of the fundamental

physical constants, because

$$1 \text{ cm}^{-1} = h \cdot c \text{ ergs}, 1 \text{ ev} = e/c \cdot 10^6 \text{ ergs},$$

the relation between the units of thermal and mechanical energy have until recently been determined as a result of special experimental investigations. This situation obtained because the calorie was determined quite independently of the mechanical energy units as the quantity of heat, required for heating 1 g water by 1°C. However, depending on the chosen initial temperature, different calories were used in practice (see [1641]). Moreover, the accuracy of determination of the calorie was limited by the attainable accuracy of the experimental measurement of the specific heat of water. After the measurement of heat had been reduced to the corresponding electrical measurements, it was no longer necessary to determine the calorie by the heat of water. The 9th International Conference on Weights and Measures [1158] passed a resolution to consider the joule as the fundamental units for the measurement of heat, which is reflected in the currently valid Government Standards of the USSR for thermal units [166]. After the joule had been adopted as heat unit, the calorie was defined by a numerical relation via the joule (see [1158, 166]). However, two definitions of the calorie via the joule have arisen historically. The definition which has long been used in heat technology in connection with the use of the International Tables for the thermodynamic properties of steam reads as follows: 1 cal = 4.1868 joule [351]. Another definition of the calorie has become traditional in thermochemistry and chemical thermodynamics: 1 cal = 4.1840 joule [147, 381, 3501, 111a]. The relation between calorie and joule which is generally used in thermochemistry and chemical thermodynamics has been adopted in the present Handbook.

The thermodynamic properties of substances are expressed in energy units per a certain quantity of substance. The thermodynamic proper-

ties are usually expressed in energy units per mole (grammolecule) of substance. However, in some cases these magnitudes are also expressed in units of energy per molecule or per gram.

During the preparation of the present Handbook all recalculations involving transformation of certain energy units into others were carried out on the basis of Table 304, compiled in correspondence with the numerical values of the physical constants given in Table 303.

TABLE 304

Transformation Factors for Energy Units Adopted in the Handbook

	эрг/молекула A	дж/моль B	кал/моль C	эв/молекула D	см ⁻¹ E
эрг/молекула A	1	$(6,02318 \pm \pm 0,00021) \cdot 10^{23}$	$(1,43958 \pm \pm 0,00003) \cdot 10^{10}$	$(6,24196 \pm \pm 0,00012) \cdot 10^{11}$	$(5,03479 \pm \pm 0,00017) \cdot 10^{16}$
дж/моль B	$(1,66025 \pm \pm 0,00003) \cdot 10^{-17}$	1	0,239006	$(1,03632 \pm \pm 0,00006) \cdot 10^{-8}$	$(8,3590 \pm \pm 0,0005) \cdot 10^{-3}$
кал/моль C	$(6,94649 \pm \pm 0,00013) \cdot 10^{-17}$	4,184	1	$(4,3360 \pm \pm 0,00025) \cdot 10^{-8}$	$0,34974 \pm \pm 0,00002$
эв/молекула D	$(1,60206 \pm \pm 0,00003) \cdot 10^{-18}$	98 495 ± 6	23 063 ± 1	1	8066,03 ± 0,14
см ⁻¹ E	$(1,98618 \pm \pm 0,00007) \cdot 10^{-16}$	11,9631 ± 0,0008	2,85926 ± 0,0002	$(1,239766 \pm \pm 0,000022) \cdot 10^{-6}$	1

A) erg/molecule; B) joule/mole; C) cal/mole; D) ev/molecule; E) cm⁻¹.

3. Relations between units of volume and pressure. Of great importance in thermodynamic calculations is also the definition of the units of volume and pressure and a knowledge of the relations between these units.

The most widely used volume units are the liter and cubic centimeter. The liter is defined as the volume occupied by 1 kg of distilled water at 4°C. The present Handbook adopted the following relations bet-

ween liter and cubic centimeter, recommended in the works [3501, 3503, 117]:

$$1 \text{ liter} = 1000.028 \pm 0.004 \text{ cm}^3.$$

The thermodynamic functions of the ideal gases are given in the present Handbook reduced to standard pressure, i.e., the pressure of one normal physical atmosphere. This magnitude is defined [326] as the pressure of the air in equilibrium with a column of mercury with a height of 76 cm at 0°C* and at normal gravitational acceleration of $g = 980.665 \text{ cm/sec}^2$.

The relation between the basic units of pressure, the normal physical atmosphere (atm) and dyne/cm^2 has been defined [326] in the following manner:

$$1 \text{ atm} = 1013250 \text{ dynes} \cdot \text{cm}^{-2}.$$

The total pressure is measured in engineering in absolute technical atmospheres (ata). The technical atmosphere is the pressure exerted by 1 kg of force on an area of 1 cm^2 . The relation between the physical and technical atmosphere, according to [3501] is determined by the following equalities:

$$1 \text{ atm} = 760 \text{ mm Hg} = 1.0332275 \text{ atm.} \quad 1 \text{ atm} = 735.5592 \text{ mm Hg} = 0.9678411 \text{ atm.}$$

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[Footnotes]

- 1982 The magnitude $\lim_{p \rightarrow 0} (pV)_0$ is also known as the standard volume of 1 mole of an ideal gas because it is equal to the volume of 1 mole of an ideal gas at $p = 1 \text{ atm}$ and $T = 273.15^\circ\text{K}$.
- 1986 The density of mercury is assumed to be 13.5951 g/cm^3 .

Appendix 3

THE FORMULAS FOR THE PRINCIPAL MOMENTS OF INERTIA OF MOLECULES

The rotational energy of a molecule is determined by its principal moments of inertia. In a system of n point masses, the moment of inertia, I , about any axis is given by

$$I = \sum_{i=1}^n m_i \cdot r_i^2 \quad (\text{P3.1})$$

where m_i is the mass and r_i the radial distance of the i th point; this can be found in any textbook on theoretical mechanics (e.g., [318]).

The moments of inertia about the three principal axes of inertia, which stand at right angles to each other, are called principal moments of inertia. The principal axes of inertia go through the center of mass (gravity) of the molecule and are directed so that the products of inertia are zero:

$$\sum_{i=1}^n m_i y_i z_i = 0; \quad \sum_{i=1}^n m_i z_i x_i = 0; \quad \sum_{i=1}^n m_i x_i y_i = 0, \quad (\text{P3.2})$$

where x_i, y_i, z_i are the coordinates of the i th atom. The coordinates x_c, y_c, z_c of the center of mass of a system of n point masses are

$$x_c = \frac{\sum_{i=1}^n m_i x_i}{M}; \quad y_c = \frac{\sum_{i=1}^n m_i y_i}{M}; \quad z_c = \frac{\sum_{i=1}^n m_i z_i}{M}, \quad (\text{P3.3})$$

where $M = \sum_{i=1}^n m_i$ is the mass of the system (of the molecule).

The principal moments of inertia of a molecule whose structure is known can be calculated straight from the formulas (P3.1)-(P3.3) if the coordinates of all atoms are given in a rectangular system. For all

practical purposes, however, the following method of calculating the principal moments of inertia of molecules is more convenient. We lay down a rectangular system in which the i th atom (mass m_i) has the coordinates x_i, y_i, z_i , and then calculate the quantities A, B, C, D, E, F as given by the following expressions:

$$\begin{aligned}
 A &= \sum_{i=1}^n m_i (y_i^2 + z_i^2) - \frac{1}{M} \left[\left(\sum_{i=1}^n m_i y_i \right)^2 + \left(\sum_{i=1}^n m_i z_i \right)^2 \right] \\
 B &= \sum_{i=1}^n m_i (x_i^2 + z_i^2) - \frac{1}{M} \left[\left(\sum_{i=1}^n m_i x_i \right)^2 + \left(\sum_{i=1}^n m_i z_i \right)^2 \right] \\
 C &= \sum_{i=1}^n m_i (x_i^2 + y_i^2) - \frac{1}{M} \left[\left(\sum_{i=1}^n m_i x_i \right)^2 + \left(\sum_{i=1}^n m_i y_i \right)^2 \right]
 \end{aligned} \tag{P3.4}$$

$$\begin{aligned}
 D &= \frac{1}{M} \left(\sum_{i=1}^n m_i x_i \right) \left(\sum_{i=1}^n m_i y_i \right) - \sum_{i=1}^n m_i x_i y_i \\
 E &= \frac{1}{M} \left(\sum_{i=1}^n m_i x_i \right) \left(\sum_{i=1}^n m_i z_i \right) - \sum_{i=1}^n m_i x_i z_i \\
 F &= \frac{1}{M} \left(\sum_{i=1}^n m_i y_i \right) \left(\sum_{i=1}^n m_i z_i \right) - \sum_{i=1}^n m_i y_i z_i
 \end{aligned}$$

$$M = \sum_{i=1}^n m_i.$$

The principal moments of inertia of the molecule, $I_A, I_B,$ and I_C , can then be found as the roots of the third-degree equation

$$\begin{vmatrix} A - I_A & -D & -E \\ -D & B - I_B & -F \\ -E & -F & C - I_C \end{vmatrix} = 0. \tag{P3.5}$$

The roots of Eq. (P3.5) satisfy the relation

$$I_A I_B I_C = \begin{vmatrix} A & -D & -E \\ -D & B & -F \\ -E & -F & C \end{vmatrix}. \tag{P3.6}$$

From this relation we can at once find the product among the principal moments of inertia when calculating the rotational components of the thermodynamic functions of polyatomic gases (see Eqs. (II.181)-(II.184)). In publications, Eq. (P3.6) has been known as the Hirschfelder equation since it was first deduced by this author in his treatise

tise [2080] for the purpose of calculating the thermodynamic functions of gases.

When the molecules are plane, E and F will be zero and the expression for the product of the principal moments, (P3.6), reduces to

$$I_A I_B I_C = (AB - D^2) C, \quad (\text{P3.7})$$

with

$$\begin{aligned} A &= \sum_{i=1}^n m_i y_i^2 - \frac{1}{M} \left(\sum_{i=1}^n m_i y_i \right)^2, \\ B &= \sum_{i=1}^n m_i x_i^2 - \frac{1}{M} \left(\sum_{i=1}^n m_i x_i \right)^2, \\ C &= A + B, \\ D &= \sum_{i=1}^n m_i x_i y_i - \frac{1}{M} \left(\sum_{i=1}^n m_i x_i \right) \left(\sum_{i=1}^n m_i y_i \right). \end{aligned} \quad (\text{P3.8})$$

The structure of polyatomic molecules is usually given by the bond lengths and by the angles formed by the bonds (structural parameters). In practical calculations it is therefore more convenient to express the principal moments of inertia and their product in terms of the structural parameters of the molecule instead of the coordinates of the atoms.

Similar expressions for the simplest polyatomic molecules will be given below. The symbols used in the formulas are explained in the figures. The number of symmetries of all types of molecules to be considered is denoted by σ .

The angles between the bonds in hydrocarbon and halocarbon molecules are either near or equal to the tetrahedral value of $\theta = 109^{\circ}28'$. The moments of inertia of such molecules, as expressed in terms of the structural parameters, can be simplified since

$$\begin{aligned} \sin \theta &= \frac{2\sqrt{2}}{3}, & \sin \frac{\theta}{2} &= \frac{\sqrt{6}}{3}, \\ \cos \theta &= -\frac{1}{3}, & \cos \frac{\theta}{2} &= \frac{1}{\sqrt{3}}. \end{aligned} \quad (\text{P3.9})$$

X_2 Molecules ($\sigma = 2$)



Fig. 22

$$I = \frac{1}{2} m_X \cdot r_{XX}^2 \quad (P3.10)$$

XY Molecules ($\sigma = 1$)



Fig. 23

$$I = \frac{m_X \cdot m_Y}{m_X + m_Y} \cdot r_{XY}^2 \quad (P3.11)$$

X_3 Molecules, Linear, Symmetric ($\sigma = 2$)



Fig. 24

$$I = 2m_X \cdot r_{XX}^2 \quad (P3.12)$$

X_3 Molecules, Not Linear, Symmetric ($\sigma = 2$)

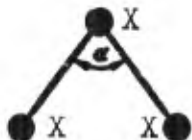


Fig. 25

$$\begin{aligned} I_A &= \frac{1}{3} m_X \cdot r_{XX}^2 \cdot \cos^2 \alpha, \\ I_B &= 2m_X \cdot r_{XX}^2 \cdot \sin^2 \alpha, \\ I_C &= I_A + I_B \end{aligned} \quad (P3.13)$$

XY_2 Molecules, Linear, Symmetric ($\sigma = 2$)



Fig. 26

$$I = 2m_Y \cdot r_{XY}^2 \quad (P3.14)$$

XY_2 Molecules, Not Linear, Symmetric ($\sigma = 2$)

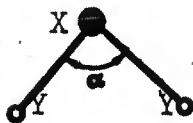


Fig. 27

$$\begin{aligned} I_A &= \frac{2m_Y \cdot m_X}{2m_Y + m_X} \cdot r_{XY}^2 \cdot \cos^2 \frac{\alpha}{2}, \\ I_B &= 2m_Y \cdot r_{XY}^2 \cdot \sin^2 \frac{\alpha}{2}, \\ I_C &= I_A + I_B. \end{aligned} \quad (P3.15)$$

$$I_A \cdot I_B \cdot I_C = \frac{2m_X \cdot m_Y^2}{(2m_Y + m_X)^3} \cdot (m_X + m_Y(1 - \cos \alpha)) \cdot \sin^2 \alpha \cdot r_{XY}^6 \quad (P3.16)$$

XY_2 Molecules, Not Linear, Asymmetric ($\sigma = 1$)

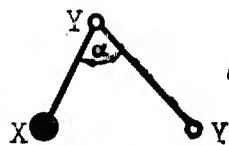


Fig. 28

$$\begin{aligned} I_A \cdot I_B \cdot I_C &= \frac{m_X \cdot m_Y^3}{M^3} \cdot r_{XY}^2 \cdot r_{YY}^2 \cdot \sin^2 \alpha \cdot [m_Y(m_X + m_Y) \cdot r_{YY}^2 + 3m_Y \cdot m_X \cdot r_{XY}^2 - \\ &\quad - 2m_X \cdot m_Y \cdot r_{XY} \cdot r_{YY} \cdot \cos \alpha]. \end{aligned} \quad (P3.17)$$

XYX' Molecules, Linear ($\sigma = 1$)



Fig. 29

$$r_{XY} = r_{XY'} = r.$$

$$I = \left[\frac{m_{Y'}(m_X + 2m_Y)^2}{(m_X + m_Y + m_{Y'})^3} + \frac{m_X(m_{Y'} - m_Y)^2}{(m_X + m_Y + m_{Y'})^3} + \frac{m_Y(m_X + 2m_{Y'})^2}{(m_X + m_Y + m_{Y'})^3} \right] \cdot r^2, \quad (P3.18)$$

XYX' Molecules, Not Linear ($\sigma = 1$)



Fig. 30

$$r_{XY} = r_{XY'} = r,$$

$$I_A = m_X \cdot y_1^2 + m_Y \cdot y_2^2 + m_{Y'} \cdot y_3^2,$$

$$I_B = m_X \cdot x_1^2 + m_Y \cdot x_2^2 + m_{Y'} \cdot x_3^2,$$

$$I_C = I_A + I_B.$$

$$(P3.19)$$

$$x_1 = u_1 \cdot \cos \beta + v_1 \cdot \sin \beta, \quad y_1 = -u_1 \cdot \sin \beta + v_1 \cdot \cos \beta,$$

$$x_2 = u_2 \cdot \cos \beta + v_2 \cdot \sin \beta, \quad y_2 = -u_2 \cdot \sin \beta + v_2 \cdot \cos \beta,$$

$$x_3 = u_3 \cdot \cos \beta + v_3 \cdot \sin \beta, \quad y_3 = -u_3 \cdot \sin \beta + v_3 \cdot \cos \beta,$$

$$u_1 = \mu_X \cdot \sin \frac{\alpha}{2}, \quad v_1 = (1 - \mu_X) \cdot r \cdot \cos \frac{\alpha}{2},$$

$$u_2 = (1 + \mu_2) \cdot r \cdot \sin \frac{\alpha}{2}, \quad v_2 = \mu_X \cdot r \cdot \cos \frac{\alpha}{2},$$

$$u_3 = -(1 - \mu_2) \cdot r \cdot \sin \frac{\alpha}{2}, \quad v_3 = -\mu_X \cdot r \cdot \cos \frac{\alpha}{2},$$

$$\mu_X = \frac{m_X}{m_X + m_Y + m_{Y'}}, \quad \mu_2 = \frac{m_{Y'} - m_Y}{m_X + m_Y + m_{Y'}};$$

the angle β is given by the equation

$$\operatorname{tg}^2 \beta + \left[\frac{m_X (v_1^2 - v_2^2) + m_Y (u_2^2 - u_3^2) + m_{Y'} (u_3^2 - u_1^2)}{m_X u_1 v_1 + m_Y u_2 v_2 + m_{Y'} u_3 v_3} \right] \operatorname{tg} \beta - 1 = 0; \quad (P3.20)$$

$$I_A \cdot I_B \cdot I_C = m_X \cdot m_Y [m_X + m_Y (1 - \cos \alpha)] \cdot \mu_1 (1 - \mu_2 \cdot \mu_X) \cdot r^6 \cdot \sin^2 \alpha,$$

where

$$\mu_1 = \frac{m_Y + m_{Y'}}{2M}, \quad \mu_2 = \frac{m_X + 2m_Y}{M}, \quad \mu_X = \frac{m_X}{M}.$$

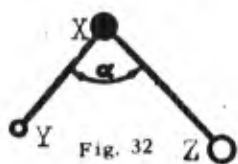
XYZ Molecules, Linear ($\sigma = 1$)



Fig. 31

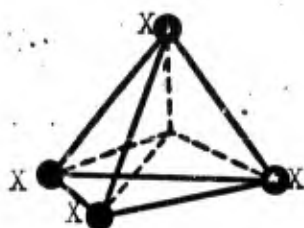
$$I = \frac{1}{M^3} [m_X (m_Y \cdot r_{XY} + m_Z \cdot r_{XZ})^2 + m_Y (m_Z \cdot r_{YZ} - m_X \cdot r_{XY})^2 + m_Z (m_X \cdot r_{XZ} + m_Y \cdot r_{YZ})^2]. \quad (P3.21)$$

X₃YZ Molecules, Not Linear ($\sigma = 1$)



$$I_A \cdot I_B \cdot I_C = \frac{m_X \cdot m_Y \cdot m_Z}{M^3} \cdot r_{XY}^2 \cdot r_{XZ}^2 \sin^2 \alpha [m_Z(m_X + m_Y) \cdot r_{XZ}^2 + m_Y(m_X + m_Z) \cdot r_{XY}^2 - 2m_Y \cdot m_Z \cdot r_{XY} \cdot r_{XZ} \cdot \cos \alpha]. \quad (P3.22)$$

X₄ Molecules, Tetrahedral ($\sigma = 12$)



$$I_A = I_B = I_C = m_X \cdot r_{XX}^2. \quad (P3.23)$$

X₂Y₂ Molecules, Linear, Symmetric ($\sigma = 2$)



$$I = \frac{1}{2} m_X \cdot r_{XX}^2 + 2m_Y \left(r_{XY} + \frac{1}{2} r_{XX} \right)^2. \quad (P3.24)$$

X₂YZ Molecules, Linear, Asymmetric ($\sigma = 1$)

$$I = \frac{1}{M^3} [m_X \cdot m_Z^2 (r_{XX}^2 + 2r_{XZ} \cdot r_{XX} + 2r_{XZ}^2) + 2m_X^2 \cdot r_{XX}^2 + m_X^2 \cdot m_Z (4r_{XZ}^2 + 3r_{XX}^2 + 4r_{XZ} \cdot r_{XX}) + m_X \cdot m_Y^2 (r_{XX}^2 + 2r_{XX} \cdot r_{XY} + 2r_{XY}^2) + m_X^2 \cdot m_Y (4r_{XY}^2 + 3r_{XX}^2 + 4r_{XX} \cdot r_{XY}) - m_X \cdot m_Y \cdot m_Z (2r_{XZ} \cdot r_{XX} + 2r_{XX} \cdot r_{XY} + 4r_{XZ} \cdot r_{XY}) + (m_Z \cdot m_Y^2 + 4m_X \cdot m_Y \cdot m_Z + m_X \cdot m_Z^2) (r_{XZ}^2 + r_{XX}^2 + r_{XY}^2)]. \quad (R3.25)$$

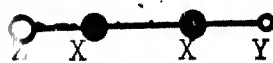


Fig. 35

X₂Y₂ Molecules, Spatial ($\sigma = 2$)

$$I_A \cdot I_B \cdot I_C = (AB - D^2) \cdot C, \quad (P3.26)$$

with

$$\begin{aligned} A &= 2m_Y (c^2 + d^2) - \frac{1}{M} (2m_Y \cdot d)^2, \\ B &= 2m_Y (b^2 + d^2) + 2m_X a^2 - \frac{1}{M} (2m_Y \cdot d)^2, \\ C &= 2m_Y (b^2 + c^2) + 2m_X \cdot a^2, \\ D &= 2m_Y \cdot b \cdot c; \end{aligned} \quad (P3.27)$$

$$a = \frac{1}{2} r_{XX}.$$

$$b = \frac{1}{2} r_{XX} - r_{XY} \cdot \cos \alpha.$$

$$c = r_{XY} \cdot \sin \alpha \cdot \sin \frac{\varphi}{2}.$$

$$d = r_{XY} \cdot \sin \alpha \cdot \cos \frac{\varphi}{2}.$$

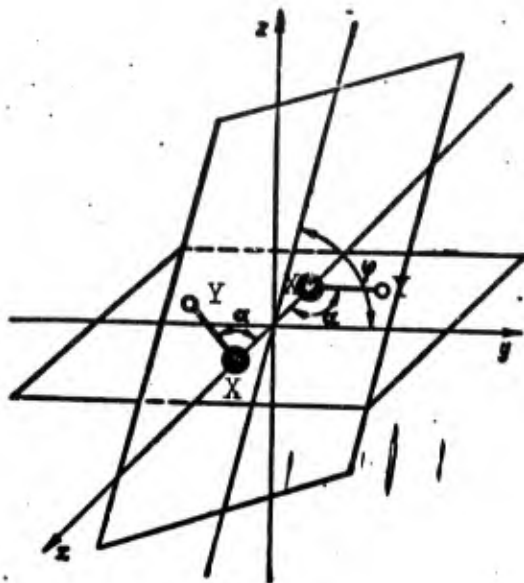


Fig. 36

XY_3 Molecules, Plane, Symmetric ($\sigma = 6$)

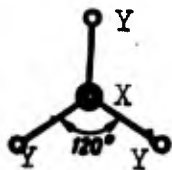


Fig. 37

$$I_A \cdot I_B \cdot I_C = \frac{27}{4} m_Y^2 \cdot r_{XY}^4. \quad (P3.28)$$

XY_3 Molecules, Pyramidal ($\sigma = 3$)

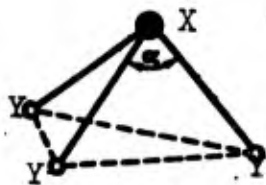


Fig. 38

$$I_A = 3m_Y \cdot r_{XY}^2 \cdot \sin^2 \beta,$$

$$I_B = I_C = \frac{3m_Y \cdot r_{XY}^2}{2 \left(1 + \frac{2m_Y}{m_X}\right)} \left[2 - \left(1 - \frac{3m_Y}{m_X}\right) \cdot \sin^2 \beta \right]; \quad (P3.29)$$

$$I_A \cdot I_B \cdot I_C = 2m_Y^2 (1 - \cos \alpha)^2 \left[1 + \mu \frac{1 + 2\cos \alpha}{1 - \cos \alpha} \right]^2 \cdot r_{XY}^4; \quad (P3.30)$$

$$\mu = \frac{m_X}{m_X + 3m_Y};$$

β is the angle between the X-Y bond and the axis of symmetry.

XY_2Z Molecules, Pyramidal ($\sigma = 1$)

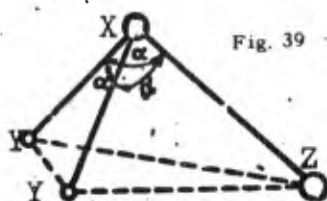


Fig. 39

$$I_A \cdot I_B \cdot I_C = ABC - BE^2, \quad (P3.31)$$

$$A = 2m_Y \cdot y_1^2 + m_X \cdot z_1^2 + m_Z \cdot z_2^2 - \frac{1}{M} (m_X \cdot z_1 - m_Z \cdot z_2)^2,$$

$$B = 2m_Y \cdot x_1^2 + m_Z \cdot x_2^2 + m_X \cdot z_1^2 + m_Z \cdot z_2^2 - \frac{1}{M} (2m_Y \cdot x_1 - m_Z \cdot x_2)^2 - \frac{1}{M} (m_X \cdot z_1 - m_Z \cdot z_2)^2,$$

(P3.32)

$$C = 2m_Y \cdot x_1^2 + m_Z \cdot x_2^2 + 2m_Y \cdot y_1^2 - \frac{1}{M} (2m_Y \cdot x_1 - m_Z \cdot x_2)^2,$$

$$E = m_Z \cdot x_2 \cdot z_2 - \frac{1}{M} (2m_Y \cdot x_1 - m_Z \cdot x_2) (m_X \cdot z_1 - m_Z \cdot z_2);$$

$$x_1 = \frac{\sqrt{3}}{3} r_{XY} \cdot \sin \frac{\alpha}{2}, \quad x_2 = \frac{2\sqrt{3}}{3} r_{XZ} \cdot \sin \frac{\alpha}{2}, \quad y_1 = r_{XY} \cdot \sin \frac{\alpha}{2},$$

$$z_1 = \sqrt{r_{XY}^2 \cdot \cos^2 \frac{\alpha}{2} - \frac{2}{3} r_{XY}^2 \cdot \sin^2 \frac{\alpha}{2}},$$

$$z_2 = \sqrt{r_{XZ}^2 \cdot \cos^2 \frac{\alpha}{2} - \frac{2}{3} r_{XZ}^2 \cdot \sin^2 \frac{\alpha}{2}} - z_1.$$

XY_4 Molecules, Tetrahedral ($\sigma = 12$)



Fig. 40

$$I_A \cdot I_B \cdot I_C = \frac{8}{3} m_Y \cdot r_{XY}^2 \quad (P3.33)$$

XY_3Z Molecules, Pyramidal ($\sigma = 3$)

$$I_A = I_B = m_Z \cdot r_{XZ}^2 + m_Y (2 + \cos \alpha) \cdot r_{XY}^2 - \frac{1}{M} (m_Z \cdot r_{XZ} - m_Y \sqrt{3} (1 + 2 \cos \alpha) \cdot r_{XY})^2, \quad (P3.34)$$

$$I_C = 2m_Y (1 - \cos \alpha) \cdot r_{XY}^2$$

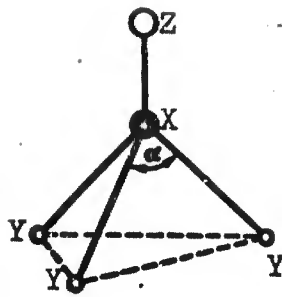


Fig. 41

In the event that $\alpha = \theta = 109^\circ 28'$ the equations of (P3.34), according to (P3.9), will assume the form

$$I_A = I_B = \frac{5}{3} m_Y \cdot r_{XY}^2 + m_Z \cdot r_{XZ}^2 - \frac{1}{M} (m_Y \cdot r_{XY} - m_Z \cdot r_{XZ})^2,$$

$$I_C = \frac{8}{3} m_Y \cdot r_{XY}^2. \quad (P3.35)$$

XY_2Z_2 Molecules, Symmetric ($\sigma = 2$)

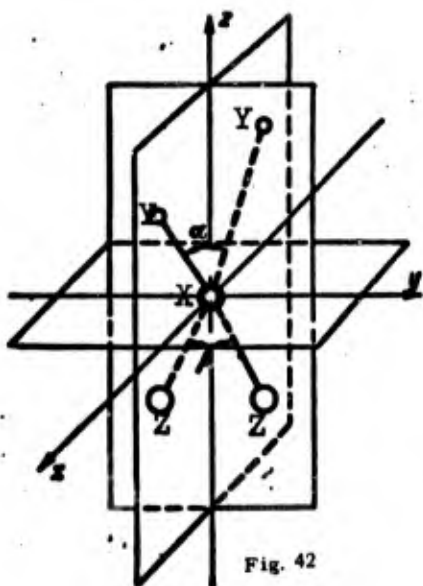


Fig. 42

$$I_A = 2m_Y \cdot r_{XY}^2 \cdot \cos^2 \frac{\alpha}{2} + 2m_Z \cdot r_{XZ}^2 -$$

$$- \frac{1}{M} (2m_Y \cdot r_{XY} \cdot \cos \frac{\alpha}{2} - 2m_Z \cdot r_{XZ} \cdot \cos \frac{\beta}{2})^2, \quad (P3.36)$$

$$I_B = 2m_Y \cdot r_{XY}^2 + 2m_Z \cdot r_{XZ}^2 \cdot \cos^2 \frac{\beta}{2} -$$

$$- \frac{1}{M} (2m_Y \cdot r_{XY} \cdot \cos \frac{\alpha}{2} - 2m_Z \cdot r_{XZ} \cdot \cos \frac{\beta}{2})^2,$$

$$I_C = 2m_Y \cdot r_{XY}^2 \cdot \sin^2 \frac{\alpha}{2} + 2m_Z \cdot r_{XZ}^2 \cdot \sin^2 \frac{\beta}{2}.$$

where $\alpha = \angle Y - X - Y$, $\beta = \angle Z - X - Z$.

When $\alpha = \beta = \theta = 109^\circ 28'$, the equations (P3.36), according to (P3.9) will assume the form

$$I_A = 2m_Y \cdot r_{XY}^2 + \frac{2}{3} m_Z \cdot r_{XZ}^2 - \frac{4}{3M} (m_Y \cdot r_{XY} - m_Z \cdot r_{XZ})^2,$$

$$I_B = \frac{2}{3} m_Y \cdot r_{XY}^2 + 2m_Z \cdot r_{XZ}^2 - \frac{4}{3M} (m_Y \cdot r_{XY} - m_Z \cdot r_{XZ})^2, \quad (P3.37)$$

$$I_C = \frac{4}{3} (m_Y \cdot r_{XY}^2 + m_Z \cdot r_{XZ}^2).$$

XY_2ZV Molecules ($\sigma = 1$)

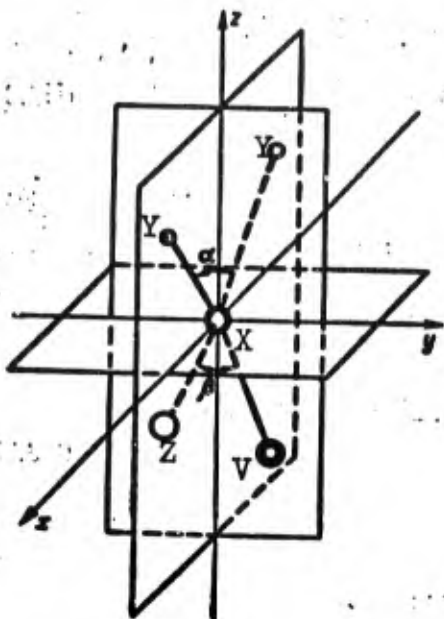


Fig. 43

$$I_A \cdot I_B \cdot I_C = A(BC - F^2), \quad (P3.38)$$

$$A = \left[(my^2) - \frac{(my)^2}{M} \right] \cdot \sin^2 \frac{\beta}{2} + (mx^2) \cdot \cos^2 \frac{\alpha}{2} + (my^2) \cdot \cos^2 \frac{\beta}{2} - \frac{1}{M} \left[(mz_1) \cdot \cos \frac{\alpha}{2} - (mz_2) \cdot \cos \frac{\beta}{2} \right]^2,$$

$$B = (mx^2) + (my^2) \cdot \cos^2 \frac{\beta}{2} - \frac{1}{M} \left[(mz_1) \cdot \cos \frac{\alpha}{2} - (mz_2) \cdot \cos \frac{\beta}{2} \right]^2, \quad (P3.39)$$

$$C = (mx^2) \cdot \sin^2 \frac{\alpha}{2} + \left[(my^2) - \frac{(my)^2}{M} \right] \cdot \sin^2 \frac{\beta}{2},$$

$$F = (myz) \cdot \sin \frac{\beta}{2} \cdot \cos \frac{\beta}{2} - \frac{1}{M} \left\{ (my) \cdot \sin \frac{\beta}{2} \left[(mz_1) \cdot \cos \frac{\alpha}{2} - (mz_2) \cdot \cos \frac{\beta}{2} \right] \right\},$$

$$(mx^2) = 2m_V \cdot r_{XV}^2,$$

$$(my^2) = m_Z \cdot r_{XZ}^2 + m_V \cdot r_{XV}^2,$$

$$(my) = m_V \cdot r_{XV} - m_Z \cdot r_{XZ},$$

$$(mz_1) = 2m_V \cdot r_{XV},$$

$$(mz_2) = m_Z \cdot r_{XZ} + m_V \cdot r_{XV},$$

$$(myz) = m_Z \cdot r_{XZ} - m_V \cdot r_{XV},$$

$$\alpha = \angle Y - X - Y, \quad \beta = \angle Z - X - V.$$

XYZVW Molecules ($\sigma = 1$)

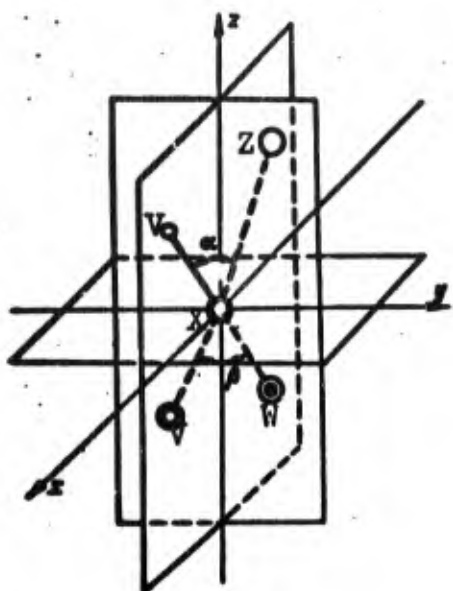


Fig. 44

$$I_A \cdot I_B \cdot I_C = ABC - 2DEF - BE^2 - CD^2 - AF^2 \quad (P3.40)$$

$$A = \left[(my^2) - \frac{(my)^2}{M} \right] \cdot \sin^2 \frac{\beta}{2} + (mx^2) \cdot \cos^2 \frac{\alpha}{2} + (my^2) \cdot \cos^2 \frac{\beta}{2} - \frac{1}{M} \left[(mz_1) \cdot \cos \frac{\alpha}{2} - (mz_2) \cdot \cos \frac{\beta}{2} \right]^2$$

$$B = \left[(mx^2) - \frac{(mx)^2}{M} \right] \cdot \sin^2 \frac{\alpha}{2} + (mx^2) \cdot \cos^2 \frac{\alpha}{2} + (my^2) \cdot \cos^2 \frac{\beta}{2} - \frac{1}{M} \left[(mz_1) \cdot \cos \frac{\alpha}{2} - (mz_2) \cdot \cos \frac{\beta}{2} \right]^2$$

$$C = \left[(mx^2) - \frac{(mx)^2}{M} \right] \cdot \sin^2 \frac{\alpha}{2} + \left[(my^2) - \frac{(my)^2}{M} \right] \cdot \sin^2 \frac{\beta}{2} \quad (P3.41)$$

$$D = -\frac{1}{M} (mx)(my) \cdot \sin \frac{\alpha}{2} \cdot \sin \frac{\beta}{2}$$

$$E = (mxz_1) \cdot \sin \frac{\alpha}{2} \cdot \cos \frac{\alpha}{2} - \frac{1}{M} \left\{ (mx) \cdot \sin \frac{\alpha}{2} \left[(mz_1) \cdot \cos \frac{\alpha}{2} - (mz_2) \cdot \cos \frac{\beta}{2} \right] \right\}$$

$$F = (myz_1) \cdot \sin \frac{\beta}{2} \cdot \cos \frac{\beta}{2} - \frac{1}{M} \left\{ (my) \cdot \sin \frac{\beta}{2} \left[(mz_1) \cdot \cos \frac{\alpha}{2} - (mz_2) \cdot \cos \frac{\beta}{2} \right] \right\}$$

$$(mx^2) = m_Y \cdot r_{XY}^2 + m_Z \cdot r_{XZ}^2 \quad (mx) = m_Y \cdot r_{XY} - m_Z \cdot r_{XZ}$$

$$(my^2) = m_Y \cdot r_{XY}^2 + m_W \cdot r_{XW}^2 \quad (my) = m_W \cdot r_{XW} - m_Y \cdot r_{XY}$$

$$(mz_1) = m_Y \cdot r_{XY} + m_Z \cdot r_{XZ} \quad (mxz_1) = m_Y \cdot r_{XY} - m_Z \cdot r_{XZ}$$

$$(mz_2) = m_Y \cdot r_{XY} + m_W \cdot r_{XW} \quad (myz_1) = m_Y \cdot r_{XY} - m_W \cdot r_{XW}$$

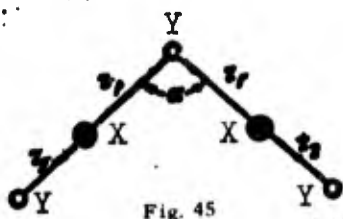
$$\alpha = \angle Y - X - Z, \quad \beta = \angle V - X - W$$

when $\alpha = \beta = \theta = 109^\circ 28'$, the equations of (P3.41), according to (P3.9),

assume the form

$$\begin{aligned}
 A &= (my^2) + \frac{1}{3}(mx^2) - \frac{2}{3M}(my)^2 - \frac{1}{M}[(mz_1) - (mz_2)]^2, \\
 B &= (mx^2) + \frac{1}{3}(my^2) - \frac{2}{3M}(mx)^2 - \frac{1}{3M}[(mz_1) - (mz_2)]^2, \\
 C &= \frac{2}{3} \left\{ (mx^2) + (my^2) - \frac{1}{M}[(mx)^2 + (my)^2] \right\}, \\
 D &= -\frac{2}{3M}(mx)(my), \\
 E &= \frac{2}{\sqrt{3}} \left\{ (mxz) - \frac{(mx)}{M}[(mz_1) - (mz_2)] \right\}, \\
 F &= \frac{2}{\sqrt{3}} \left\{ (myz) - \frac{(my)}{M}[(mz_1) - (mz_2)] \right\},
 \end{aligned} \tag{P3.42}$$

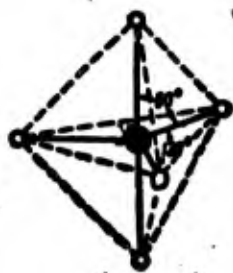
X_2Y_3 Molecules, Angular ($\sigma = 2$)



$$\begin{aligned}
 I_A &= 2m_X \left(r_{XY} \sin \frac{\alpha}{2} \right)^2 + 2m_Y \left[(r'_{XY} + r''_{XY}) \sin \frac{\alpha}{2} \right]^2, \\
 I_B &= m_Y x^2 + 2m_X \left(x - r_{XY} \cos \frac{\alpha}{2} \right)^2 + \\
 &\quad + 2m_Y \left[(r'_{XY} + r''_{XY}) \cos \frac{\alpha}{2} - x \right]^2, \\
 I_C &= I_A + I_B,
 \end{aligned} \tag{P3.43}$$

$$x = \frac{2m_X (r'_{XY} + r''_{XY}) + 2m_X r_{XY} \cdot \cos \frac{\alpha}{2}}{3m_Y + 2m_X}$$

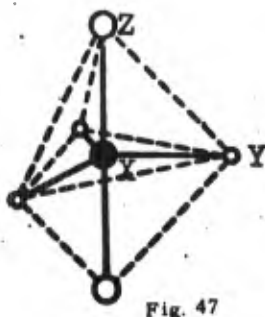
XY_5 Molecules, Bipyramidal ($\sigma = 6$)



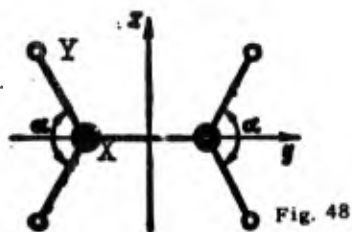
$$I_A \cdot I_B \cdot I_C = \frac{1}{4} m_Y \cdot r_{XY}^2 \tag{P3.44}$$

XY_3Z_2 Molecules, Bipyramidal ($\sigma = 6$)

$$\begin{aligned}
 I_A = I_B &= 2m_Z \cdot r_{XZ}^2 + \frac{3}{2} m_Y \cdot r_{XY}^2, \\
 I_C &= 3m_Y \cdot r_{XY}^2.
 \end{aligned} \tag{P3.45}$$



X_2X_4 Molecules, Plane, Symmetric ($\sigma = 4$)



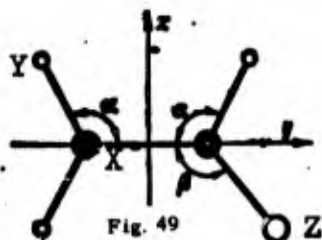
$$I_A = 4m_Y \cdot r_{XY}^2 \cdot \sin^2 \frac{\alpha}{2},$$

$$I_B = 4m_Y \left(\frac{1}{2} r_{XX} + r_{XY} \cdot \cos \frac{\alpha}{2} \right)^2 + \frac{1}{2} m_X \cdot r_{XX}^2. \quad (P3.46)$$

$$I_C = I_A + I_B,$$

$$\alpha = \angle Y - X - Y.$$

X_2Y_3Z Molecules, Plane ($\sigma = 1$)



$$I_A \cdot I_B \cdot I_C = (AB - D^2) \cdot C, \quad (P3.47)$$

$$A = 3m_Y \cdot y_1^2 + 2m_X \cdot y_2^2 + m_Z \cdot y_3^2 - \frac{1}{M} (m_Y \cdot y_1 - m_Z \cdot y_3)^2,$$

$$B = 3m_Y \cdot x_1^2 + 2m_X \cdot x_2^2 + m_Z \cdot x_3^2 - \frac{1}{M} (m_Z \cdot x_3 - m_Y \cdot x_1)^2,$$

$$C = A + B,$$

(P3.48)

$$D = m_Y \cdot x_1 \cdot y_1 - m_Z \cdot x_3 \cdot y_3 - \frac{1}{M} (m_Y \cdot y_1 - m_Z \cdot y_3) (m_Z \cdot x_3 - m_Y \cdot x_1),$$

$$x_1 = \frac{1}{2} r_{XX} - r_{XY} \cdot \cos \alpha, \quad x_2 = \frac{1}{2} r_{XX}, \quad x_3 = \frac{1}{2} r_{XX} - r_{XZ} \cdot \cos \beta$$

$$y_1 = r_{XY} \cdot \sin \alpha, \quad y_2 = 0, \quad y_3 = r_{XZ} \sin \beta, \quad \alpha = \angle X - X - Y, \quad \beta = \angle X - X - Z.$$

$X_2Y_2Z_2$ Molecules, Plane ($\sigma = 2$)

a) Trans type

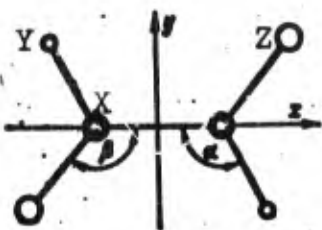


Fig. 50

$$I_A \cdot I_B \cdot I_C = (AB - D^2) C, \quad (P3.49)$$

$$A = 2(m_Y \cdot y_1^2 + m_Z \cdot y_2^2).$$

$$B = 2(m_X \cdot x_1^2 + m_Y \cdot x_2^2 + m_Z \cdot x_3^2), \quad (P3.50)$$

$$C = A + B,$$

$$D = 2(m_Z \cdot x_2 y_3 - m_Y \cdot x_1 y_1).$$

b) Asymmetric

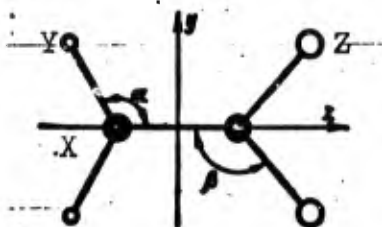


Fig. 51

$$I_A \cdot I_B \cdot I_C = ABC, \quad (P3.51)$$

$$A = 2(m_Y \cdot y_1^2 + m_Z \cdot y_2^2).$$

$$B = 2(m_X \cdot x_1^2 + m_Y \cdot x_2^2 + m_Z \cdot x_3^2) - \frac{4}{M}(m_Z x_3 - m_Y x_2)^2, \quad (P3.52)$$

$$C = A + B.$$

c) Cis type

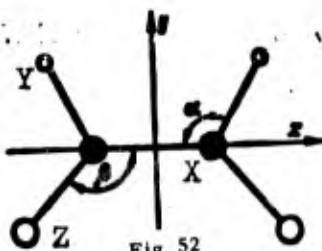


Fig. 52

$$I_A \cdot I_B \cdot I_C = ABC, \quad (P3.53)$$

$$A = 2(m_Y \cdot y_1^2 + m_Z \cdot y_2^2) - \frac{4}{M}(m_Y \cdot y_1 - m_Z \cdot y_2)^2.$$

$$B = 2(m_X \cdot x_1^2 + m_Y \cdot x_2^2 + m_Z \cdot x_3^2), \quad (P3.54)$$

$$C = A + B.$$

The symbols in Eqs. (P3.50), (P3.52), (P3.54) denote

$$x_1 = \frac{1}{2} r_{XX}, \quad x_2 = \frac{1}{2} r_{XX} - r_{XY} \cdot \cos \alpha, \quad | \quad x_3 = \frac{1}{2} r_{XX} - r_{XZ} \cdot \cos \beta,$$

$$y_1 = r_{XY} \cdot \sin \alpha, \quad y_2 = r_{XZ} \cdot \sin \beta.$$

$$\alpha = \angle Y - X - X, \quad \beta = \angle Z - X - X.$$

X_2Y_2ZV Molecules, Plane ($\sigma = 1$)

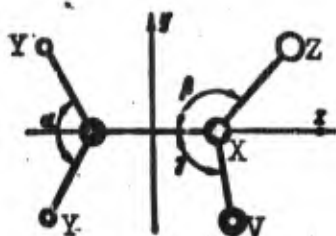


Fig. 53

$$I_A \cdot I_B \cdot I_C = (AB - D^2) \cdot C. \quad (P3.55)$$

$$A = 2m_Y \cdot y_1^2 + m_Z \cdot y_3^2 + m_V \cdot y_4^2 - \frac{1}{M} (m_Z y_3 - m_V y_4)^2,$$

$$B = 2m_Y x_1^2 + 2m_X x_2^2 + m_Z x_3^2 + m_V x_4^2 - \frac{1}{M} (m_Z x_3 + m_V x_4 - 2m_Y x_2)^2. \quad (P3.56)$$

$$C = A + B,$$

$$D = m_Z x_3 y_3 - m_V x_4 y_4 - \frac{1}{M} (m_Z x_3 + m_V x_4 - 2m_Y x_2) (m_Z y_3 - m_V y_4).$$

$$x_1 = \frac{1}{2} r_{XX}, \quad x_2 = \frac{1}{2} r_{XX} + r_{XY} \cdot \cos \frac{\alpha}{2}, \quad x_3 = \frac{1}{2} r_{XX} - r_{XZ} \cdot \cos \beta,$$

$$x_4 = \frac{1}{2} r_{XX} - r_{XV} \cdot \cos \gamma.$$

$$y_1 = r_{XY} \cdot \sin \frac{\alpha}{2}, \quad y_3 = r_{XZ} \cdot \sin \beta, \quad y_4 = r_{XV} \cdot \sin \gamma.$$

$$\alpha = \angle Y - X - Y, \quad \beta = \angle X - X - Z, \quad \gamma = \angle X - X - V.$$

XY_6 Molecules, Octahedral, Symmetric ($\sigma = 24$)

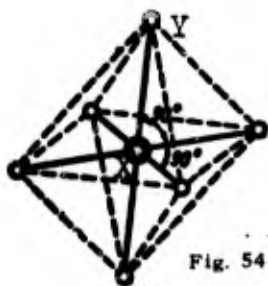


Fig. 54

$$I_A \cdot I_B \cdot I_C = 64m_Y^2 \cdot r_{XY}. \quad (P3.57)$$

X_4Y_6 Molecules, Tetrahedral ($\sigma = 12$)

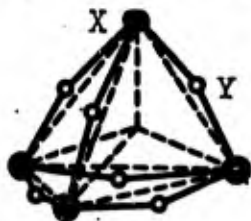


Fig. 55

$$I_A = I_B = I_C = \frac{8}{3} m_X \cdot r_1^2 + 4m_Y \cdot r_2^2. \quad (P3.58)$$

where

$$r_1 = 0,61237 \cdot r_{XX},$$

$$r_2 = r_{XY} \cdot \cos \frac{\angle X - Y - X}{2} + 0,35355 \cdot r_{XX}.$$

X_4Y_8 Molecules, Tetrahedral ($\sigma = 2$)

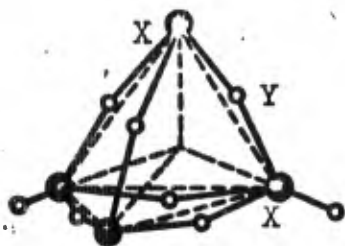


Fig. 56

$$I_A = \frac{8}{3} m_X \cdot r_X^2 + 4m_Y + \frac{2}{3} m_Y r_Y^2 - \frac{4m_Y^2}{3M} r_Y^2,$$

$$I_B = \frac{8}{3} m_X \cdot r_X^2 + 4m_Y \cdot r_Y^2 + 2m_Y r_Y^2 - \frac{4m_Y^2}{3M} r_Y^2, \quad (P3.59)$$

$$I_C = \frac{8}{3} m_X \cdot r_X^2 + 4m_Y \cdot r_Y^2 + \frac{4}{3} m_Y r_Y^2,$$

where

$$r_X = 0,61237 \cdot r_{XX},$$

$$r_Y = r_{XY} \cdot \cos \frac{\angle X-Y-X}{2} + 0,35355 \cdot r_{XX},$$

$$r_Y = r_X + r_{XY}.$$

r_{XY} is the distance from an X-atom to an inner Y-atom, r_{XY} is the distance from an X-atom to an outer Y-atom.

X_4Y_{10} Molecules, Tetrahedral ($\sigma = 12$)

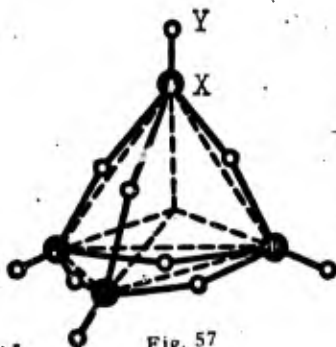


Fig. 57

$$I_A \cdot I_B \cdot I_C = \frac{8}{3} (m_X \cdot r_1^2 + m_Y \cdot r_2^2) + 4m_Y \cdot r_2^2, \quad (P3.60)$$

where

$$r_1 = 0,61237 \cdot r_{XX},$$

$$r_2 = r_{XY} \cdot \cos \frac{\angle X-Y-X}{2} + 0,35355 \cdot r_{XX},$$

$$r_2 = r_1 + r_{XY}.$$

r_{XY} is the distance from an X-atom to an inner Y-atom, r_{XY} is the distance from an X-atom to an outer Y-atom.

Appendix 4

CONCISE DATA FROM THE THEORY OF MOLECULAR VIBRATIONS AND FORMULAS FOR THE FREQUENCIES

Every molecule can be considered as a dynamical system in which the particles (atoms) are linked to one another by forces which tend to keep the entire system in a state of stable equilibrium. The configuration of such a system is defined by giving $3N$ coordinates, N being the number of atoms in the molecule. The orientation in space of a molecule as a whole in equilibrium is of no importance to the problem of the vibrational frequencies. The vibrational state of the system can therefore be characterized by giving $n = 3N - 6$ independent vibrational coordinates q_1 .* The latter may be the interatomic spacings and the angles between the bonds.

Let us describe the potential energy of a system by a function V which depends only on the location of the particles in this system, i.e.,

$$V = V(q_1, q_2, \dots, q_n). \quad (P4.1)$$

We shall consider vibrations caused by small deviations of the system from its stable equilibrium (the subscript zero indicates the value of the respective parameter when the system is at equilibrium). Let

$$q_i = q_{i0} + \eta_i. \quad (P4.2)$$

where η_i represents a small correction to the vibrational coordinate of the i th atom. Expanding the function V near its equilibrium into a Taylor series and omitting terms of higher than second degree we have

$$V(q_1, q_2, \dots, q_n) = V(q_{01}, q_{02}, \dots, q_{0n}) + \sum_i \left(\frac{\partial V}{\partial q_i} \right)_0 \eta_i + \frac{1}{2} \sum_{i,j} \left(\frac{\partial^2 V}{\partial q_i \partial q_j} \right)_0 \eta_i \eta_j. \quad (P4.3)$$

Furthermore, considering the balance condition and counting the potential energy from its minimum at equilibrium on, we find

$$2V = \sum_{i,j=1}^n \left(\frac{\partial^2 V}{\partial q_i \partial q_j} \right)_0 \eta_i \eta_j = \sum_{i,j=1}^n k_{ij} \eta_i \eta_j. \quad (P4.4)$$

where $k_{ij} = \left(\frac{\partial^2 V}{\partial q_i \partial q_j} \right)_0 = k_{ji}$. The kinetic energy of the system (with $q_{i0} = 0$) is (see, e.g., [161])

$$2T = \sum_{i,j=1}^n m_{ij} \dot{q}_i \dot{q}_j = \sum_{i,j=1}^n m_{ij} \dot{\eta}_i \dot{\eta}_j. \quad (P4.5)$$

where $m_{ij} = m_{ji}$.* The η_i can be taken as new vibrational coordinates (termed natural coordinates); the equations (P4.4) and (P4.5) will then give the potential and kinetic energies in natural coordinates.

When we set up the equation of motion in natural coordinates for each degree of freedom of the dynamical system, we arrive at a system of n linear second-order differential equations:

$$\sum_{j=1}^n (m_{ij} \ddot{\eta}_j + k_{ij} \eta_j) = 0, \quad i = 1, 2, \dots, n. \quad (P4.6)$$

We want a solution of the form

$$\eta_j = \eta_{j0} \cos(2\pi \omega t + \varphi). \quad (P4.7)$$

with the frequency ω , the amplitude η_{j0} , and the initial phase φ of the vibration.

After inserting the anticipated solution (P4.7) into (P4.6) we obtain a system of linear homogeneous algebraic equations

$$\sum_{j=1}^n (k_{ij} - m_{ij} \lambda) \eta_{j0} = 0, \quad i = 1, 2, \dots, n, \quad (P4.8)$$

with

$$\lambda = 4\pi^2 \omega^2. \quad (P4.9)$$

The condition that the system (P4.8) has a non-trivial solution is that

its determinant must vanish:

$$\begin{vmatrix} k_{11} - m_{11}\lambda & k_{12} - m_{12}\lambda & \dots & k_{1n} - m_{1n}\lambda \\ k_{21} - m_{21}\lambda & k_{22} - m_{22}\lambda & \dots & k_{2n} - m_{2n}\lambda \\ \dots & \dots & \dots & \dots \\ k_{n1} - m_{n1}\lambda & k_{n2} - m_{n2}\lambda & \dots & k_{nn} - m_{nn}\lambda \end{vmatrix} = 0. \quad (\text{P4.10})$$

This is called the secular equation, which has n roots determining the frequencies at which the solution (P4.7) satisfies the equation (P4.6). The problem of finding the frequencies of vibration of a dynamical system is thus reduced to solving the secular equation (P4.10). However, this form of the secular equation is very awkward to solve because the unknowns λ appear in every term of the determinant (P4.10).

In general it can be shown (see, e.g., [77]) that the kinetic and the potential energies of the system can be written as

$$2T = \sum_{i=1}^n \dot{Q}_i^2, \quad 2V = \sum_{i=1}^n \lambda_i Q_i^2, \quad (\text{P4.11})$$

where the λ_j represent the roots of Eq. (P4.10). The coordinates in which the kinetic and the potential energies have the form of (P4.11) are called normal coordinates. In terms of these, the equations of motion of the system are

$$\ddot{Q}_i + \lambda_i Q_i = 0, \quad i = 1, 2, \dots, n. \quad (\text{P4.12})$$

An oscillation described by Eq. (P4.12) is called a normal vibration. It follows from Eq. (P4.12) that each normal coordinate Q_i corresponds to the respective i th independent vibration of the system. The condition for stable equilibrium entails that the potential energy of the system must always be positive, i.e., that all λ_i are positive. According to Eq. (P4.9), each root λ_i of Eq. (P4.10) is related to one frequency of vibration ω_i , called the frequency of the i th normal vibration of the system. For normal vibrations, the secular equation has the form

$$\begin{vmatrix} \lambda_1 - 4\pi^2\omega^2 & 0 & \dots & 0 \\ 0 & \lambda_2 - 4\pi^2\omega^2 & \dots & 0 \\ \dots & \dots & \dots & \dots \\ 0 & 0 & \dots & \lambda_n - 4\pi^2\omega^2 \end{vmatrix} = 0, \quad (\text{P4.13})$$

and the problem of finding the normal frequencies of the system leads to solving n independent equations similar to Eq. (P4.9). Normal vibrations that are related to individual roots λ_1 of Eq. (P4.10) are called nondegenerate. Vibrations corresponding to multiple roots of Eq. (P4.10) are called degenerate, and the number of identical roots is called multiplicity of the degeneration. When the problem is degenerate not all the normal coordinates Q_1 are independent.

It can be proved (see, e.g., [4293]) that any set of coordinates describing the system can be written in terms of linear combinations between the normal coordinates, in other words, any type of vibration can be regarded as a superposition of a certain number of normal vibrations.

Hence, when we have the equations for the kinetic and the potential energies in some system of coordinates, we have to find linear combinations of these so that the kinetic and potential energies of the system can be expressed as a sum of square terms. Such a procedure is not difficult to accomplish when the molecule is composed of relatively few atoms. When the number of atoms is high, however, we have to use special methods. Wilson [4290, 4292, 4293] and Yel'yashevich [128, 185] have developed such methods of finding the secular equation. Both methods are basically the same although they may differ in minor aspects. The determinant (P4.10) can easily be transformed so that the unknown λ appears only in the diagonal terms.* To achieve this we only have to multiply the equations (P4.6) by the factors A_{l1} which satisfy the condition**

$$\sum_l A_{ll} m_{ll} = \delta_{ll}, \quad (\text{P4.14})$$

with the Kronecker symbol δ_{lj} equal to unity when $j = l$, and zero when $j \neq l$. After multiplication we have

$$\sum_i (A_{im} \ddot{\eta}_i + A_{ik} \eta_i) = 0. \quad (P4.15)$$

Taking the sum in this equation over i first, and then over j , and considering the condition (P4.14) we find the equation of motion giving explicitly the second derivative of the coordinates with respect to time:*

$$\ddot{\eta}_i = - \sum_j \sum_l A_{il} k_{lj} \eta_j = - \sum_l A_{il} \frac{\partial V}{\partial \eta_l}. \quad (P4.16)$$

Inserting a solution of the form (P4.7) in Eq. (P4.16) and considering (P4.9) we have

$$\sum_i [(\sum_l A_{il} k_{lj}) - \delta_{ij} \lambda] \eta_{0i} = 0. \quad (P4.17)$$

With the symbols

$$\sum_l A_{il} k_{lj} = D_{ij}, \quad (P4.18)$$

the equations of (P4.17) assume the form

$$\sum_i (D_{ij} - \delta_{ij} \lambda) \eta_{0i} = 0. \quad (P4.19)$$

The condition for a non-trivial solution of the system (P4.19) is that its determinant vanishes, i.e.,

$$\begin{vmatrix} D_{11} - \lambda & D_{12} & \dots & D_{1n} \\ D_{21} & D_{22} - \lambda & \dots & D_{2n} \\ \dots & \dots & \dots & \dots \\ D_{n1} & D_{n2} & \dots & D_{nn} - \lambda \end{vmatrix} = 0. \quad (P4.20)$$

Since in this determinant λ appears only in the diagonal terms, the diagonal elements D_{ll} must directly furnish the sought frequencies of vibration, if the off-diagonal elements are small. This is important when an approximate solution is to be constructed. According to (P4.18), the coefficients D_{lj} are determined only by the A_{lj} , since the k_{lj} are given quantities in this problem of calculating the frequencies.** Bas-

ically, the coefficients $A_{\ell 1}$ could be found by setting up the expression for the kinetic energy in natural coordinates and then applying the condition (P4.14). In practice, however, such a procedure is very complicated and lengthy.*

Yel'yashevich has shown [128, 185] that the coefficients $A_{\ell 1}$ can be derived directly from Eq. (P4.16). From Eq. (P4.14) it can be seen that the $A_{\ell 1}$ are completely defined as the coefficients of the m_{1j} in the expression (P4.5) for the kinetic energy, i.e., that they depend only on the masses of the atoms and on the equilibrium structural parameters of the molecule. Under sufficiently general assumptions about the nature of the intramolecular interactions, the coefficients $A_{\ell 1}$ can be tabulated in terms of natural coordinates [128].

Hence, in order to obtain the secular equation in natural coordinates, one has to give definite coefficients k_{1j} in the expression for the kinetic energy, to multiply them according to (P4.18) by the respective $A_{\ell 1}$ (using a table of the kinematic coefficients), and to assemble the determinant (P4.20) out of the resulting $D_{\ell j}$. Molecules containing a greater number of atoms have a determinant of high order; it is then very complicated to resolve Eq. (P4.20). Reducing the order of the secular determinant will therefore be an important factor. It can be achieved by introducing symmetry coordinates (for more details, see [152, 4292, 4293, 128, 77, 185]). When such symmetry coordinates are introduced, the secular determinant is decomposed into several determinants of lower order. The symmetry coordinates constitute an intermediate link between the natural and the normal coordinates. Yel'yashevich [128, 185] has compiled tables of the symmetry coefficients for several point groups, which make it easy to change from natural to symmetry coordinates and hence simplify the problem considerably.

Wilson's method [4290, 4292, 4293] is based on representing a sec-

ular equation of the type (P4.20) in the form of an algebraic equation and expressing the coefficients of the different powers of λ in terms of the internal coordinates.* In matrix form, the secular equation (P4.20) is

$$|FG - E\lambda| = 0, \quad \text{P4.21)}$$

where F represents the matrix of the force constants (or the matrix of the dynamic coefficients, as referred to by Yel'yashevich), E the unity matrix, and G the inverse kinetic energy matrix.

The internal coordinates S_t are related to the Cartesian coordinates ξ_1 by the linear transformation

$$S_t = \sum_{i=1}^{3N} B_{ti} \xi_i, \quad t = 1, 2, \dots, n. \quad \text{(P4.22)}$$

It can be shown that the elements of the G matrix are

$$G_{t't} = \sum_{i=1}^{3N} \frac{1}{m_i} B_{ti} B_{t'i}, \quad t, t' = 1, 2, \dots, n. \quad \text{(P4.23)}$$

When the secular equation (P4.21) is represented in the form of an algebraic equation with respect to λ , the formula for the sth term of the equation, as shown by Wilson [4290, 4292, 4293] is

$$n_s = (-1)^s \lambda^{3N-s} \sum G^{(s)} F^{(s)}, \quad \text{(P4.24)}$$

where $G^{(s)}$ is an arbitrary minor of order s of the determinant $|G|$, and $F^{(s)}$ the corresponding minor of the determinant $|F|$. Summation is to be taken over all possible minors of the order s.**

To set up the explicit secular equation we therefore have to find all the coefficients B_{t1} of the transformation from Cartesian to internal coordinates (set up the transformation matrix B), then from Eq. (P4.23) to find the elements of the matrix G, and from Eq. (P4.24) to calculate the products between the corresponding minors of the determinants $|G|$ and $|F|$. It must be said that the number of required minors

rises rapidly with N . In the formulas of (P4.24) Wilson [4290] has assumed extensive simplifications for the case that the molecule consists of atoms having identical masses. For symmetric molecules, the equation can be solved easily by introducing symmetry coordinates. The equation for λ in this case is decomposed to several equations of lower order. The explicit form of the secular equation is convenient because it is easily accessible to methods of approximate solution. One of these is to separate the high frequencies [4290, 4292, 4293]. This method is based on the empirical fact that certain frequencies of vibration are actually determined only by a small number of force constants and depend only very slightly on the remaining ones (existence of characteristic frequencies, great difference between the frequencies of one molecule, etc.). The equation in this case can be solved for the high and the low frequencies separately. For the low frequencies, the equation must be divided by the product of all great force constants contained therein on the condition that the great force constants tend to infinity. All terms whose denominator contains a great force constant will then vanish, and the degree of the equation for the low frequencies will have been reduced (by the number of great force constants). The corresponding equation for the high frequencies can be derived by setting all the small force constants equal to zero. Also in this case the degree of the equation reduces.

Techniques of approximation have been developed for resolving the secular determinants. An outline of these methods is given in monographs by Wilson, Decius, and Cross [4293], and by Vol'kenshteyn, Yel'yashevich, and Stepanov [128]. Among these methods are the linear interpolation by James and Coolidge [2207, 2208] and iteration methods for solving equations of the type (P4.18), developed by Gopshteyn [163] and Mayants [294, 294a]. Vol'kenshteyn, Yel'yashevich, and Stepanov

[128] have also worked out a method for a rapid and approximate calculation of the frequencies of complex molecules and have compiled tables on the over-all interaction coefficients for some hydrocarbon compounds.

When the problem of calculating the frequencies of vibration had been set it was assumed that the coefficients k_{1j} were known force constants in the expression for the kinetic energy, (P4.4). Actually, these force constants have been determined accurately only for a small number of diatomic and simple polyatomic molecules since the inverse problem is of more practical importance, namely the problem of finding the force constants from the known frequencies or the problem of finding the potential function. The expression (P4.4) for the potential energy shows that generally, for unsymmetric molecules, the number of independent force constants is equal to

$$\frac{(3N-6)(3N-5)}{2}.$$

This number is less for symmetric molecules as the symmetry gives rise to additional relations between certain force constants.* The number of independent force constants is 6 for an asymmetric triatomic molecule, and 4 for a symmetric triatomic molecule.** It is evident that from the above expression, generally, the number of force constants is greater than the number of fundamental frequencies.*** When therefore the force constants are determined from spectral data, one has to consider subsidiary relations or to adopt simplifications concerning the values of the force constants.

More relations between the frequencies and the force constants can be derived from data on isotopes of the molecule whose force constants are to be found. Assuming that the fields (as well as the potential functions) in isotopic molecules are identical to one another, we may

also take it for granted that their force constants are identical. Simplifying assumptions mostly neglect one or another constant in order that the number of sought constants may not be greater than the number of frequencies of the molecule under investigation. This is what is known as approximations or various models of the field of the molecular forces.

One of the more simple approximations is that of a field of radial forces which is based on the assumption that the forces, keeping the molecule balanced, act only along the lines connecting each pair of atoms. When, in this case, the complete set of variations of the interatomic spacings is used as internal coordinates (radial-force field coordinates), the expression (P4.4) for the potential energy will contain only the squares of these coordinates but not the mixed terms.* It must be said that in practical calculations this approximation is completely inadequate for linear molecules, and that the force constants calculated in this approximation make sense only for purely ionic interactions.

Another approximation which is more apt to render the valence idea of the chemists is the approximation of the valence-force field. This model suggests forces that are opposed to any change in the length of the valence bonds and in the angles between these (the forces between free atoms are not directly taken into consideration). Hence, apart from changes in the spacings, angular changes too enter the valence-force field coordinates. In this simple valence-force field approximation, the mixed terms in the expression for the potential energy are omitted so that the matrix of the force constants may be diagonal. The fact that the bonds have characteristic frequencies, i.e., frequencies which have approximately the same value in different molecules and depend only slightly on the other bonds, suggests that the force constants of such bonds are correct when calculated in the simple valence-

force field approximation. Hence, the force constants of corresponding bonds of other molecules can be used in calculating the frequencies of vibration of a certain molecule. Although this method has been successfully applied in many cases, it cannot be applied always because the omission of the interaction force constants will very often lead to a completely wrong representation of the real field within the molecule.

A more exact description of the intramolecular forces requires in the potential function also the mixed interaction terms of the different coordinates. A potential function comprising all interaction force constants may be called a generalized potential function.* The matrix of the force constants is then no longer diagonal. Apart from the linear symmetric triatomic molecules there is only a small number of other molecules whose generalized potential function is known. In practice, the force constants are calculated mostly from a set of such constants that would match the observed frequency. The secular equation can for this purpose be written as

$$\lambda^n + A_1\lambda^{n-1} + A_2\lambda^{n-2} + \dots + (-1)^n A_n = 0.$$

From this we can at once obtain the relations

$$\begin{aligned} -\sum \lambda_i &= A_1, \\ \sum \sum \lambda_i \lambda_j &= A_2, \\ \dots & \\ (-1)^n \lambda_1 \lambda_2 \dots \lambda_n &= A_n. \end{aligned}$$

which can also be applied to calculating the force constants from the known frequencies. Up to this time the theoretical methods of calculating the force constants of interaction have not been sufficiently developed to be used for practical purposes. For this reason, the theory of the molecular vibrations is still semi-empirical.

At the end of this appendix equations will be given relating the vibration frequencies of several types of polyatomic molecules to their structural parameters and their supposed system of force constants. For

practical purposes, which have been pursued in this Handbook, it was sometimes sufficient in the calculation of the frequencies not to employ the whole set of force constants but to use a more or less approximate potential function. The following three kinds of approximation have been used altogether: 1) The simple valence-force field approximation in which all the force constants of interaction are taken equal to zero, 2) the more general valence-force field approximation in which only part of the interaction force constants are taken into consideration, and 3) the generalized potential function which takes account of all interaction constants. The methods of Wilson [4290, 4293] and Yel'yashevich [128, 185] have been used for deducing the respective equations in the last case. Accordingly, the symbols used by Wilson and Yel'yashevich for the force constants have been adopted also in this Handbook.*

In the formulas deduced by the Yel'yashevich method, the nondimensional parameters $\epsilon_i = m_H/m_i$ and $\sigma_\beta = s_{C-H}/s_\beta$, have been used, where \underline{i} is the symbol for the atom, β the spacing number, $m_H = 1.088$ is the spectroscopic mass of the hydrogen atom, and $s_{C-H} = 1.09 \cdot 10^{-8}$ cm. In all the equations below $\mu_i = 1/m_i$, where m_i is the mass of the i th atom in atomic weight units, the frequencies ν_i are given in cm^{-1} , the interatomic spacings d_i in cm, and Δd and $\Delta \alpha$ are the changes in bond length and angle, respectively. The force constants have the dimension of $\text{dynes} \cdot \text{cm}^{-1}$, and the factor A^* in the equations below is then equal to $16.976 \text{ dyne}^{-1} \cdot \text{cm}^{-1}$. In various publications the force constants are given in cm^{-2} for simpler calculation; in such cases the factor A is equal to unity.**

The potential function is given in all cases for clarity.

XYZ Molecules, Angular, Asymmetric (see Fig. 32)*

$$\begin{aligned}
 2V &= f_a(\Delta d_1)^2 + f_a(\Delta d_2)^2 + f_a(\Delta \alpha)^2 \\
 v_1^2 + v_2^2 + v_3^2 &= A \left\{ (\mu_x + \mu_y) f_a + (\mu_x + \mu_z) f_a + B \frac{f_a}{d_1 d_2} \right\}, \\
 v_1^2 v_2^2 + v_1^2 v_3^2 + v_2^2 v_3^2 &= A^2 \left\{ (\mu_x \mu_y + \mu_x \mu_z + \mu_y \mu_z + \mu_x^2 \sin^2 \alpha) f_a f_a + \right. \\
 &+ \left. [(\mu_x + \mu_y) B - \mu_x^2 d_1^2 \sin^2 \alpha] \frac{f_a}{d_1 d_2} f_a + [(\mu_x + \mu_z) B - \mu_x^2 d_2^2 \sin^2 \alpha] \frac{f_a}{d_1 d_2} f_a \right\}, \\
 v_1^2 v_2^2 v_3^2 &= A^3 B (\mu_x \mu_y + \mu_x \mu_z + \mu_y \mu_z) \frac{f_a}{d_1 d_2} f_a f_a.
 \end{aligned} \tag{P4.25}$$

$B = \mu_x d_1^2 + \mu_y d_2^2 + \mu_x (d_1^2 + d_2^2 - 2d_1 d_2 \cos \alpha)$, and f_a, f_a, f_a are the force constants of the bonds X - Y (d_1), X - Z (d_2) and of the angle between Y - X - Z (α).

XYZ Molecules, Linear, Asymmetric (see Fig. 31)**

$$\begin{aligned}
 \text{a) } 2V &= f_a(\Delta d_1)^2 + f_a(\Delta d_2)^2 + 2f_{ad}(\Delta d_1)(\Delta d_2) + f_a(\Delta \alpha)^2 \\
 v_1^2 + v_2^2 &= A \{ (\mu_x + \mu_y) f_a + (\mu_y + \mu_z) f_a - 2\mu_y f_{ad} \}, \\
 v_1^2 v_2^2 &= A^2 (\mu_x \mu_y + \mu_x \mu_z + \mu_y \mu_z) (f_a f_a - f_{ad}^2), \\
 v_3^2 &= A (\mu_x d_1^2 + \mu_x d_2^2 + \mu_y (d_1 + d_2)^2) \frac{f_a}{d_1 d_2}.
 \end{aligned} \tag{P4.26}$$

$$\begin{aligned}
 \text{b) } 2V &= f_a(\Delta d_1)^2 + f_a(\Delta d_2)^2 + f_a(\Delta \alpha)^2 \\
 v_1^2 + v_2^2 &= A \{ (\mu_x + \mu_y) f_a + (\mu_y + \mu_z) f_a \}, \\
 v_1^2 v_2^2 &= A^2 (\mu_x \mu_y + \mu_x \mu_z + \mu_y \mu_z) f_a f_a, \\
 v_3^2 &= A (\mu_x d_1^2 + \mu_x d_2^2 + \mu_y (d_1 + d_2)^2) \frac{f_a}{d_1 d_2}.
 \end{aligned} \tag{P4.27}$$

f_a, f_a, f_a, f_{ad} are the force constants of the bonds X - Y (d_1), Y - Z (d_2), of the angle between X - Y - Z (α), and of the interaction between the bonds (d_1 and d_2).

XY₂ Molecules, Angular, Symmetric (see Fig. 27)***

$$\begin{aligned}
 \text{a) } 2V &= f_a[(\Delta d_1)^2 + (\Delta d_2)^2] + 2f_{ad}(\Delta d_1)(\Delta d_2) + 2f_{ad}d(\Delta d)(\Delta \alpha) + f_a d^2(\Delta \alpha)^2 \\
 v_1^2 + v_2^2 &= A \left[(\mu_y + 2\mu_x \cos^2 \frac{\alpha}{2}) (f_d + f_{ad}) - 4\mu_x \sin \alpha \cdot f_{ad} + 2(\mu_y + 2\mu_x \sin^2 \frac{\alpha}{2}) f_a \right], \\
 v_1^2 v_2^2 &= 2A^2 \mu_y (\mu_y + 2\mu_x) [(f_d + f_{ad}) f_a - 2f_{ad}^2], \\
 v_3^2 &= A (\mu_y + 2\mu_x \sin^2 \frac{\alpha}{2}) (f_d - f_{ad}).
 \end{aligned} \tag{P4.28}$$

$$\begin{aligned}
 \text{b)} \quad 2V &= f_d[(\Delta d_1)^2 + (\Delta d_2)^2] + 2f_{dd}(\Delta d_1)(\Delta d_2) + f_\alpha d^2(\Delta \alpha)^2 \\
 v_1^2 + v_2^2 &= A \left[\left(\mu_Y + 2\mu_X \cos^2 \frac{\alpha}{2} \right) (f_d + f_{dd}) + 2 \left(\mu_Y + 2\mu_X \sin^2 \frac{\alpha}{2} \right) f_\alpha \right], \quad (\text{P4.29}) \\
 v_1^2 v_2^2 &= 2A^2 \mu_Y (\mu_Y + 2\mu_X) (f_d + f_{dd}) f_\alpha, \\
 v_3^2 &= A \left(\mu_Y + 2\mu_X \sin^2 \frac{\alpha}{2} \right) (f_d - f_{dd}).
 \end{aligned}$$

$$\begin{aligned}
 \text{c)} \quad 2V &= f_d[(\Delta d_1)^2 + (\Delta d_2)^2] + f_\alpha d^2(\Delta \alpha)^2 \\
 v_1^2 + v_2^2 &= A \left[\left(\mu_Y + 2\mu_X \cos^2 \frac{\alpha}{2} \right) f_d + 2 \left(\mu_Y + 2\mu_X \sin^2 \frac{\alpha}{2} \right) f_\alpha \right], \quad (\text{P4.30}) \\
 v_1^2 v_2^2 &= 2A^2 \mu_Y (\mu_Y + 2\mu_X) f_d f_\alpha, \\
 v_3^2 &= A \left(\mu_Y + 2\mu_X \sin^2 \frac{\alpha}{2} \right) f_\alpha.
 \end{aligned}$$

$f_d, f_\alpha, f_{dd}, f_{d\alpha}$ are the force constants of the bond X - Y (d), of the angle Y - X - Y (α), of the interaction between the bonds, and of the interaction between bond and angle.

XY_2 Molecules, Linear, Symmetric (see Fig. 26)*

$$\begin{aligned}
 \text{a)} \quad 2V &= f_d[(\Delta d_1)^2 + (\Delta d_2)^2] + 2f_{dd}(\Delta d_1)(\Delta d_2) + f_\alpha(\Delta \alpha)^2 \\
 v_1^2 &= A\mu_Y (f_d + f_{dd}), \\
 v_2^2 &= 2A (\mu_Y + 2\mu_X) \frac{f_\alpha}{d^2}, \\
 v_3^2 &= A (\mu_Y + 2\mu_X) (f_d - f_{dd}).
 \end{aligned} \quad (\text{P4.31})$$

$$\begin{aligned}
 \text{b)} \quad 2V &= f_d[(\Delta d_1)^2 + (\Delta d_2)^2] + f_\alpha(\Delta \alpha)^2 \\
 v_1^2 &= A\mu_Y f_d, \\
 v_2^2 &= 2A (\mu_Y + 2\mu_X) \frac{f_\alpha}{d^2}, \\
 v_3^2 &= A (\mu_Y + 2\mu_X) f_d.
 \end{aligned} \quad (\text{P4.32})$$

f_d, f_α, f_{dd} are the force constants of the bond X - Y (d), of the angle Y - X - Y (α), and of the interaction between the bonds.

XY_2Z Molecules, Pyramidal (see Fig. 39)**

Over-all interaction matrix

a) for the symmetry type A'

$$\begin{vmatrix}
 A_0 k_{xy} - \lambda & A_1 k_{xz} & 2A_2 k_\alpha & A_4 k_\beta \\
 A_1 k_{xy} & A_2 k_{xz} - \lambda & 0 & A_5 k_\beta \\
 2A_3 k_{xy} & 0 & A_6 k_\alpha - \lambda & A_6 k_\beta \\
 A_4 k_{xy} & A_5 k_{xz} & A_6 k_\alpha & A_7 k_\beta - \lambda
 \end{vmatrix} = 0; \quad (\text{P4.33a})$$

b) for the symmetry type A''

$$\begin{vmatrix} B_0 k_{xy} - \lambda & B_1 k_\beta \\ B_1 k_{xy} & B_2 k_\beta - \lambda \end{vmatrix} = 0, \quad (P4.33b)$$

where

$$A_0 = e_y + e_x (1 + \cos \alpha); \quad A_1 = 2e_x \cos \alpha, \quad A_2 = e_x + e_z, \quad A_3 = -e_x \sigma_{xy} \sin \alpha,$$

$$A_4 = -e_x \sigma_{xz} \sin \alpha, \quad A_5 = 2\sigma_{xy} [e_x (1 - \cos \alpha) + e_y],$$

$$A_6 = \frac{2\sigma_{xy}}{1 + \cos \alpha} [\sigma_{xy} (e_x + e_y) \cos \alpha - e_x (\sigma_{xy} + \sigma_{xz}) \cos^2 \alpha + e_x \sigma_{xz} (\cos \alpha + \sin^2 \alpha)],$$

$$A_7 = e_x (\sigma_{xy}^2 + \sigma_{xz}^2 - 2\sigma_{xy} \sigma_{xz} \cos \alpha) + e_y \sigma_{xy}^2 + e_z \sigma_{xz}^2 +$$

$$+ \frac{\sigma_{xy}}{1 + \cos \alpha} [\sigma_{xy} (e_x + e_z) \cos \alpha - e_x (\sigma_{xy} + \sigma_{xz}) \cos^2 \alpha + e_x \sigma_{xz} (\cos \alpha + \sin^2 \alpha)],$$

$$B_0 = e_y + e_x (1 - \cos \alpha), \quad B_1 = e_x \sigma_{xz} \sin \alpha,$$

$$B_2 = e_x (\sigma_{xy}^2 + \sigma_{xz}^2 - 2\sigma_{xy} \sigma_{xz} \cos \alpha) + e_y \sigma_{xy}^2 + e_z \sigma_{xz}^2 -$$

$$- \frac{\sigma_{xy}}{1 + \cos \alpha} [\sigma_{xy} (e_x + e_z) \cos \alpha - e_x (\sigma_{xy} + \sigma_{xz}) \cos^2 \alpha + e_x \sigma_{xz} (\cos \alpha + \sin^2 \alpha)].$$

$$e_x = \frac{m_H}{m_X}, \quad e_y = \frac{m_H}{m_Y}, \quad e_z = \frac{m_H}{m_Z},$$

$$\sigma_{xy} = \frac{r_{C-H}}{r_{X-Y}}, \quad \sigma_{xz} = \frac{r_{C-H}}{r_{X-Z}},$$

k_{xy} , k_{xz} , k_α , k_β are the force constants of the bonds X - Y, X - Z, and of the angles Y - X - Y and Y - X - Z, α is the angle between Y - X - Z, and $\lambda = 4\pi^2 c v^2$. All the force constants are given in cm^{-2} .

X₂YZ Molecules, Linear, Asymmetric (see Fig. 35)*

$$2V = f_d (\Delta d_1)^2 + f_d (\Delta d_2)^2 + f_d (\Delta d_3)^2 + 2f_{d,d_1} (\Delta d_1) (\Delta d_2) +$$

$$+ 2f_{d,d_2} (\Delta d_2) (\Delta d_3) + f_\alpha (\Delta \alpha_1)^2 + f_\alpha (\Delta \alpha_2)^2$$

$$v_1^2 + v_2^2 + v_3^2 = A (\mu_x + \mu_y) f_d + 2\mu_x f_d + (\mu_x + \mu_z) f_d - 2\mu_x (f_{d,d_1} - f_{d,d_2}),$$

$$v_1^2 v_2^2 + v_1^2 v_3^2 + v_2^2 v_3^2 = A^2 \{ \mu_x (\mu_x + 2\mu_y) (f_d f_{d_1} - f_{d,d_1}^2) + \mu_x (\mu_x + 2\mu_z) (f_d f_{d_2} - f_{d,d_2}^2) +$$

$$+ (\mu_x + \mu_y) (\mu_x + \mu_z) f_d f_d - 2\mu_x (\mu_x + \mu_z) f_d f_{d,d_1} - 2\mu_x (\mu_x + \mu_y) f_d f_{d,d_2} + 2\mu_x^2 f_{d,d_1} f_{d,d_2} \},$$

$$v_1^2 v_2^2 v_3^2 = A^3 [2\mu_x \mu_y \mu_z + \mu_x^2 (\mu_y + \mu_z)] (f_d f_{d_1} f_{d_2} - f_{d,d_1} f_{d,d_2} - f_{d,d_2} f_{d,d_1}). \quad (P4.34)$$

$$v_1^2 = A (\mu_y d_2^2 + \mu_x d_1^2 + \mu_x (d_1 + d_2)^2) \frac{f_{d_1}}{d_1^2 d_2^2},$$

$$v_2^2 = A (\mu_z d_3^2 + \mu_x d_2^2 + \mu_x (d_2 + d_3)^2) \frac{f_{d_2}}{d_2^2 d_3^2}.$$

$f_{d_1}, f_{d_2}, f_{d_3}, f_{\alpha_1}, f_{\alpha_2}, f_{d_1 d_2}, f_{d_2 d_3}$ are the force constants of the bonds X - Y (d_1), X - X (d_2), X - Z (d_3), of the angle Y - X - Y (α_1), of the angle Z - X - X (α_2), and of the interaction between the bonds.

X_2Y_2 Molecules, Linear, Symmetric (see Fig. 34)*

a)

$$2V = f_d [(\Delta d_1)^2 + (\Delta d_2)^2] + f_a (\Delta d_2)^2 + 2f_{da} (\Delta d_2) [(\Delta d_1) + (\Delta d_3)] + 2f_{\alpha\alpha} (\Delta \alpha_1) (\Delta \alpha_2) + f_\alpha [(\Delta \alpha_1)^2 + (\Delta \alpha_2)^2]$$

$$v_1^2 + v_2^2 = A \{2\mu_X (f_d - 2f_{da}) + (\mu_X + \mu_Y) f_d\}$$

$$v_1^2 v_2^2 = 2A^2 \mu_X \mu_Y (f_d f_a - 2f_{da}) \quad (P4.35)$$

$$v_3^2 = A (\mu_X + \mu_Y) f_a$$

$$v_4^2 = A (\mu_Y d_2^2 + \mu_X (d_2 + 2d_1)^2) \frac{f_a - f_{\alpha\alpha}}{d_1^2 d_2^2}$$

$$v_5^2 = A (\mu_X + \mu_Y) \frac{f_a + f_{\alpha\alpha}}{d_1^2}$$

b)

$$2V = f_d [(\Delta d_1)^2 + (\Delta d_2)^2] + f_a (\Delta d_2)^2 + f_\alpha [(\Delta \alpha_1)^2 + (\Delta \alpha_2)^2]$$

$$v_1^2 + v_2^2 = A \{2\mu_X f_d + (\mu_X + \mu_Y) f_d\}$$

$$v_1^2 v_2^2 = 2A^2 \mu_X \mu_Y f_d f_a$$

$$v_3^2 = A (\mu_X + \mu_Y) f_a \quad (P4.36)$$

$$v_4^2 = A (\mu_Y d_2^2 + \mu_X (d_2 + 2d_1)^2) \frac{f_a}{d_1^2 d_2^2}$$

$$v_5^2 = A (\mu_X + \mu_Y) \frac{f_a}{d_1^2}$$

$f_{d_1}, f_{d_2}, f_\alpha, f_{dd}, f_{\alpha\alpha}$ are the force constants of the bonds X - Y (d_1), X - X (d_2), of the angle Y - X - X (α), and of the interaction between bonds and angles.

XY_3 Molecules, Pyramidal, Symmetric (see Fig. 38)**

a)

$$2V = f_d [(\Delta d_1)^2 + (\Delta d_2)^2 + (\Delta d_3)^2] + 2f_{da} [(\Delta d_1) (\Delta d_2) + (\Delta d_1) (\Delta d_3) + (\Delta d_2) (\Delta d_3)] + 2f_{\alpha\alpha} \cdot d [(\Delta \alpha_{12}) \Delta d_3 + (\Delta \alpha_{13}) \Delta d_2 + (\Delta \alpha_{23}) \Delta d_1] + 2f_{\alpha\alpha} \cdot d^2 [(\Delta \alpha_{12}) (\Delta \alpha_{13}) + (\Delta \alpha_{13}) (\Delta \alpha_{23}) + (\Delta \alpha_{12}) (\Delta \alpha_{23})] + f_\alpha \cdot d^2 [(\Delta \alpha_{12})^2 + (\Delta \alpha_{13})^2 + (\Delta \alpha_{23})^2]$$

$$v_1^2 + v_2^2 = A \left\{ [\mu_Y + \mu_X (1 + 2 \cos \alpha)] (f_d + 2f_{da}) - \frac{8\mu_X}{\sin \alpha} (1 - \cos \alpha) (1 + 2 \cos \alpha) f_{\alpha\alpha} + \frac{2(1 + 2 \cos \alpha)}{1 + \cos \alpha} [\mu_Y + 2\mu_X (1 - \cos \alpha)] (f_\alpha + 2f_{\alpha\alpha}) \right\}$$

$$\begin{aligned}
v_1^2 v_2^2 &= 2A^2 \frac{1+2\cos\alpha}{1+\cos\alpha} \mu_Y (\mu_Y + 3\mu_X) [(f_d + 2f_{dd}) (f_a + 2f_{aa}) - 4f_{da}], \\
v_3^2 + v_4^2 &= A \left\{ [\mu_Y + \mu_X (1 - \cos\alpha)] (f_d - f_{dd}) - \frac{\mu_X (1 - \cos\alpha)^2}{2\sin\alpha} f_{da} + \right. \\
&\quad \left. + \frac{[\mu_Y (2 + \cos\alpha) + \mu_X (1 - \cos\alpha)^2]}{1 + \cos\alpha} (f_a - f_{aa}) \right\}, \quad (P4.37) \\
v_3^2 v_4^2 &= \frac{A^2}{1 + \cos\alpha} \left[\mu_Y^2 (2 + \cos\alpha) + 3\mu_X \mu_Y (1 - \cos\alpha) + \frac{3}{4} \mu_X^2 (1 - \cos\alpha)^2 \right] \times \\
&\quad \times \left[f_d - f_{dd} (f_a - f_{aa}) - \frac{1}{4} f_{da}^2 \right].
\end{aligned}$$

$$\begin{aligned}
b) \quad 2V &= f_d [(\Delta d_1)^2 + (\Delta d_2)^2 + (\Delta d_3)^2] + 2f_{dd} [(\Delta d_1)(\Delta d_2) + (\Delta d_1)(\Delta d_3) + (\Delta d_2)(\Delta d_3)] + \\
&\quad + 2f_{aa} \cdot d^2 [(\Delta \alpha_{12})(\Delta \alpha_{13}) + (\Delta \alpha_{12})(\Delta \alpha_{23}) + (\Delta \alpha_{13})(\Delta \alpha_{23})] + \\
&\quad + f_a \cdot d^2 [(\Delta \alpha_{12})^2 + (\Delta \alpha_{13})^2 + (\Delta \alpha_{23})^2]
\end{aligned}$$

$$\begin{aligned}
v_1^2 + v_2^2 &= A \left\{ [\mu_Y + \mu_X (1 + 2\cos\alpha)] (f_d + 2f_{dd}) + \frac{2(1+2\cos\alpha)}{1+\cos\alpha} [\mu_Y + 2\mu_X (1 - \cos\alpha)] f_a \right\} (f_a + 2f_{aa}), \\
v_1^2 v_2^2 &= 2A^2 \frac{1+2\cos\alpha}{1+\cos\alpha} \mu_Y (\mu_Y + 3\mu_X) (f_d + 2f_{dd}) (f_a + 2f_{aa}), \quad (P4.38)
\end{aligned}$$

$$v_3^2 + v_4^2 = A \left\{ [\mu_Y + \mu_X (1 - \cos\alpha)] (f_d - f_{dd}) + \frac{[\mu_Y (2 + \cos\alpha) + \mu_X (1 - \cos\alpha)^2]}{1 + \cos\alpha} (f_a - f_{aa}) \right\}.$$

$$v_3^2 v_4^2 = \frac{A^2}{1 + \cos\alpha} \left[\mu_Y^2 (2 + \cos\alpha) + 3\mu_X \mu_Y (1 - \cos\alpha) + \frac{3}{4} \mu_X^2 (1 - \cos\alpha)^2 \right] (f_d - f_{dd}) (f_a - f_{aa}).$$

$$c) \quad 2V = f_d [(\Delta d_1)^2 + (\Delta d_2)^2 + (\Delta d_3)^2] + f_a \cdot d^2 [(\Delta \alpha_{12})^2 + (\Delta \alpha_{13})^2 + (\Delta \alpha_{23})^2]$$

$$v_1^2 + v_2^2 = A \left\{ [\mu_Y + \mu_X (1 + 2\cos\alpha)] f_d + \frac{2(1+2\cos\alpha)}{1+\cos\alpha} [\mu_Y + 2\mu_X (1 - \cos\alpha)] f_a \right\},$$

$$v_1^2 v_2^2 = 2A^2 \frac{1+2\cos\alpha}{1+\cos\alpha} \mu_Y (\mu_Y + 3\mu_X) f_d f_a. \quad (P4.39)$$

$$v_3^2 + v_4^2 = A \left\{ [\mu_Y + \mu_X (1 - \cos\alpha)] f_d + \frac{\mu_Y (2 + \cos\alpha) + \mu_X (1 - \cos\alpha)^2}{1 + \cos\alpha} f_a \right\},$$

$$v_3^2 v_4^2 = \frac{A^2}{1 + \cos\alpha} \left[\mu_Y^2 (2 + \cos\alpha) + 3\mu_X \mu_Y (1 - \cos\alpha) + \frac{3}{4} \mu_X^2 (1 - \cos\alpha)^2 \right] f_d f_a.$$

$f_d, f_a, f_{dd}, f_{aa}, f_{da}$ are the force constants of the bond X - Y (d), of the angle Y - X - Y (α), and of the interaction among the bonds, among the angles, and between bonds and angles.

XY₃ Molecules, Plane, Symmetric (see Fig. 37)*

$$\begin{aligned}
a) \quad 2V &= f_d [(\Delta d_1)^2 + (\Delta d_2)^2 + (\Delta d_3)^2] + 2f_{dd} [(\Delta d_1)(\Delta d_2) + (\Delta d_1)(\Delta d_3) + (\Delta d_2)(\Delta d_3)] + \\
&\quad + 2f_{aa} \cdot d^2 [(\Delta \alpha_1)(\Delta \alpha_2 + \Delta \alpha_3) + (\Delta \alpha_2)(\Delta \alpha_3)] + \\
&\quad + f_a \cdot d^2 [(\Delta \alpha_1)^2 + (\Delta \alpha_2)^2 + (\Delta \alpha_3)^2] + f_r \cdot d^2 (\Delta \gamma)^2
\end{aligned} \quad (P4.40)$$

$$v_1^2 = A \mu_Y (f_d + 2f_{dd}),$$

$$v_2^2 = A (\mu_Y + 3\mu_X) f_r.$$

$$v_3^2 + v_4^2 = A \left(\mu_Y + \frac{3}{2} \mu_X \right) [(f_d - f_{dd}) + 3(f_a - f_{aa})].$$

$$v_3^2 v_4^2 = 3A^2 \mu_Y (\mu_Y + 3\mu_X) (f_d - f_{dd}) (f_a - f_{aa}).$$

$$b) 2V = f_d[(\Delta d_1)^2 + (\Delta d_2)^2 + (\Delta d_3)^2] + f_\alpha \cdot d^2[(\Delta \alpha_1)^2 + (\Delta \alpha_2)^2 + (\Delta \alpha_3)^2] + f_\gamma \cdot d^2(\Delta \gamma)^2$$

$$v_1^2 = A\mu_Y f_d.$$

$$v_2^2 = A(\mu_Y + 3\mu_X) f_\gamma. \quad (P4.41)$$

$$v_3^2 + v_4^2 = A\left(\mu_Y + \frac{3}{2}\mu_X\right)(f_d + 3f_\alpha).$$

$$v_5^2 v_6^2 = 3A^2 \mu_Y (\mu_Y + 3\mu_X) f_d f_\alpha.$$

$f_d, f_\alpha, f_{dd}, f_{\alpha\alpha}$ are the force constants of the bond X - Y (d), of the angle Y - X - Y (α), and of the interaction between bonds and angles, f_γ is the force constant related to vibrations out of the plane.

XY₅ Molecules, Bipyramidal (see Fig. 46)*

$$2V = f_d[(\Delta d_{12})^2 + (\Delta d_{13})^2 + (\Delta d_{14})^2] + f_\alpha[(\Delta \alpha_{12})^2 + (\Delta \alpha_{13})^2] + \\ + 2f_{d\alpha}[(\Delta d_{12})(\Delta \alpha_{12}) + (\Delta d_{13})(\Delta \alpha_{13}) + (\Delta d_{14})(\Delta \alpha_{14})] + \\ + f_\alpha \cdot d^2[(\Delta \alpha_{212})^2 + (\Delta \alpha_{213})^2 + (\Delta \alpha_{214})^2] + f_\alpha \cdot d^2[(\Delta \alpha_{312})^2 + (\Delta \alpha_{313})^2 + (\Delta \alpha_{314})^2] + \\ + (\Delta \alpha_{216})^2 + (\Delta \alpha_{316})^2 + (\Delta \alpha_{416})^2]$$

$$v_1^2 = A\mu_Y (f_d + 2f_{d\alpha}).$$

$$v_2^2 = A\mu_Y f_\alpha. \quad (P4.42)$$

$$v_3^2 + v_4^2 = A[(2\mu_X + \mu_Y) f_d + 2(3\mu_X + \mu_Y) f_\alpha].$$

$$v_5^2 v_6^2 = 2A^2 \mu_Y (5\mu_X + \mu_Y) f_d f_\alpha.$$

$$v_6^2 + v_7^2 + v_8^2 = A\left[\left(\frac{3}{2}\mu_X + \mu_Y\right)(f_d - f_{d\alpha} + 3f_\alpha) + \frac{3}{2}(2\mu_X + \mu_Y) f_\alpha\right].$$

$$v_5^2 v_6^2 + v_5^2 v_7^2 + v_5^2 v_8^2 = 3A^2\left[\frac{1}{2}\left(\frac{7}{2}\mu_X + \mu_Y\right)(f_d - f_{d\alpha} + 3f_\alpha) f_\alpha + (3\mu_X + \mu_Y)(f_d - f_{d\alpha}) f_\alpha\right].$$

$$v_6^2 v_7^2 v_8^2 = \frac{9}{2} A^3 \mu_Y^2 (5\mu_X + \mu_Y) (f_d - f_{d\alpha}) f_\alpha f_\alpha. \quad v_9 = \frac{7}{2} A\mu_Y f_\alpha.$$

$f_{d_1}, f_{d_2}, f_{\alpha_1}, f_{\alpha_2}, f_{dd}$ are the force constants related to the base and axial bonds X - Y, to the angles Y - X - Y in the base (α_1) and the axial (α_2) planes of the molecule, and to the interaction between the bonds.

- 2003 For a linear molecule with five translational and rotational degrees of freedom, $n = 3N - 5$ coordinates must be given.
- 2004 The coefficients of the m_{ij} depend on the mass of the atoms on the equilibrium-structure parameters of the molecule.
- 2006 Here and in what follows we shall use the terminology and presentation as given by Yel'yashevich [128, 185]. A more exact presentation of this problem has been published by Mayants [294a].
- 2006 The $A_{\ell i}$ are basically the elements of the inverse kinetic energy matrix.
- 2007 Tatevskiy [408] has proved Eq. (P4.16) for an arbitrary system of coordinates.
- 2007 Yel'yashevich has called the $A_{\ell i}$ the coefficients of kinematic interaction, the k_{ij} (force constants) coefficients of dynamic interaction, and the $D_{\ell j}$ coefficients of over-all interaction.
- 2008 Tatevskiy [408] has shown that the calculations can be simplified by using the momenta as variables.
- 2009 The terminology and symbols used by Wilson [4290, 4292, 4293] have been adopted here. The internal coordinates are basically the same as the natural coordinates (see page 2004).
- 2009 According to definition, the coefficient of λ^{3N-6} is equal to unity and the coefficient of λ^0 is equal to the product of the determinants $|G| \cdot |F|$.
- 2011 Owing to the symmetry there are subsidiary relations between some of the internal coordinates.
- 2011 The number of independent force constants does not depend on the coordinates chosen to describe the potential function.
- 2011 Except for linear symmetric triatomic molecules and certain other symmetric molecules.
- 2012 I.e., the matrix of the force constants is diagonal in the radial-force field approximation.
- 2013 Bhagavantam and Venkatarayudu [77] have introduced the term "intravalent forces" for this case. Herzberg [152] does not give it a special name but calls it a "more general form of force field," a "more general potential function," etc.

2014

The interrelation between the denotations for the force constants, as suggested by Wilson, Yel'yashevich, and Herzberg, is as follows:

1	Силавая постоянная	Вильсон [4293] 2	Ельясевич [128] 3	Герцберг [152] 4
5	валентная	f_{d_i}	k_{d_i}	k_i
6	деформационная	f_{a_i}	k_{γ}	k_{θ}, k_{Δ}
7	взаимодействия связей	$f_{d_i d_j}$	A	k_{ij}
8	взаимодействия углов	$f_{a_i a_j}$	I	k_{θ}
9	взаимодействия связи с углом . .	$f_{d_i a_j}$	a_i	—

1) Force constant; 2) Wilson [4293]; 3) Yel'yashevich [128]; 4) Herzberg [152]; 5) valence; 6) deformation; 7) interaction among bonds; 8) interaction among angles; 9) interaction between bond and angle.

2014 $A = 16/4\pi^2 m_0$, where m_0 is the mass in grams of an oxygen atom, c is the velocity of light in centimeters per second.

2014 The constants f_{α} and $f_{\alpha\alpha}$ in Eqs. (P4.25)-(P4.27), P4.31), (P4.32), (P4.34)-(P4.36) have the dimension of radian^{-1} with $A = 1$, and $\text{dyne}\cdot\text{cm}/\text{radian}$ with $A = 16.976 \text{ dyne}^{-1}\cdot\text{cm}^{-1}$.

2015 The formulas (P4.25) are given in accordance with [2579, 4290].

2015 The formulas (P4.26) have been deduced by Milevskaya, the formulas (P4.27) are given in accordance with [152].

2015 The formulas (P4.28) are given according to [2847a], the formulas (P4.29) and (P4.30) according to [152].

2016 The formulas (P4.31) and P4.32) are given according to [152].

2016 The formulas (P4.33) have been deduced by Veyts after the Yel'yashevich method [128].

2017 The formulas (P4.34) have been deduced by Milevskaya.

2018 The formulas (P4.35) and (P4.36) are given according to [152].

2018 The formulas (P4.37) are given according to [4087], the formulas (P4.38) and (P4.39) according to [152].

2019 The formulas (P4.40) are given according to [3250a], the formulas (P4.41) according to [152].

2020

The formulas (P4.42) are given according to [3719].

Appendix 5

CONSIDERATION OF THE INTERMOLECULAR INTERACTIONS IN THE CALCULATION OF THE THERMODYNAMIC FUNCTIONS OF GASES

In Chapter 2 of this Handbook we have examined methods of calculating the thermodynamic functions of ideal gases from their molecular constants. The thermodynamic functions of the gases calculated in this way are sufficiently accurate in the range of high temperature and low pressure. At low temperatures and high pressures, the thermodynamic functions of gases calculated without the intermolecular interaction may differ greatly from the thermodynamic functions of the respective real gases. A method of considering the intermolecular interactions in the calculation of the thermodynamic functions of gases is examined in this Appendix; the p-V-T data and the force constants of the intermolecular potential of some gases that are considered in this Handbook, and whose experimental p-V-T data are known, are given.

1. THE CALCULATION OF THE THERMODYNAMIC FUNCTIONS OF REAL GASES FROM AN EQUATION OF STATE IN VIRIAL FORM

Calculation of the thermodynamic functions from the equation of state of a real gas. The thermodynamic properties of a real gas can be calculated from the general thermodynamic relations if the equation of state of this gas is known. The method is to determine the partial derivative $(\partial p/\partial T)_V$ from the equation of state of the gas, and then to calculate the internal energy and the entropy of the gas using the relations

$$\left(\frac{\partial E}{\partial V}\right)_T = T\left(\frac{\partial p}{\partial T}\right)_V - p. \quad (P5.1)$$

$$\therefore \left(\frac{\partial S}{\partial V}\right)_T = \left(\frac{\partial p}{\partial T}\right)_V \quad (\text{P5.2})$$

It must be kept in mind that in this case not the entire values of E or S but only the corrections to the respective thermodynamic functions of an ideal gas, that are due to the intermolecular interaction, are determined. From the internal energy found in this way it is possible to calculate the enthalpy $H = E + pV$ of the gas as well as the specific heat capacities at constant pressure and constant volume.*

The equation of state in virial form. Various equations of state are used for calculating the thermodynamic properties of real gases. The most comprising synopsis on equations of state used at present is in the book by Vukalovich and Novikov [138].

It must be said that the known equations of state are either empirical or, at the best, semi-empirical. The range of applicability of such equations is therefore limited by the temperatures at which the respective experimental data can be gotten, i.e., not over 1000-1200°K. Using these equations of state at high temperatures may be unjustified since there are no relevant experimental data.

The only equation of state which can be deduced theoretically by means of statistical mechanics is one in virial form. That makes it possible to apply such an equation of state at temperatures for which there are no experimental p-V-T data. A deduction and theoretical reasoning of the equation of state in virial form has been given by Bogolyubov [93] and Mayer and Goeppert-Mayer [2822]. These authors have worked out a general method of calculating the virial coefficients for any temperature from the data characterizing the properties of the gas molecules and their interaction energy.

We may point out the publications [1182, 1350, 4328] as examples of how the equation of state in virial form is employed in the calcula-

tion of the thermodynamic properties of real gases at high temperatures.

Two varieties of the equation of state in virial form are known:

$$pV = RT \left(1 + \frac{B}{V} + \frac{C}{V^2} + \frac{D}{V^3} + \dots \right), \quad (\text{P5.3})$$

$$pV = RT (1 + B'p + C'p^2 + D'p^3 + \dots), \quad (\text{P5.4})$$

where B, C, D, ... are the virial coefficients of the second, third, fourth, etc., terms, respectively. The virial coefficients B, C, D, ... of pure gases turn out to be functions of temperature solely and are determined by the interaction between groups of two, three, or four molecules, respectively. Hence, consideration of the pair interaction between the molecules leads to the second virial coefficient, consideration of triple interaction to the third virial coefficient, etc.

It can be shown that the virial coefficients B, C, and D are interrelated with the virial coefficients B', C', and D' as follows:

$$B' = \frac{B}{RT}; \quad (\text{P5.5})$$

$$C' = \frac{C - B^2}{R^2 T^2}; \quad (\text{P5.6})$$

$$D' = \frac{D - 3BC + 2B^3}{R^3 T^3}. \quad (\text{P5.7})$$

The equation of state (P5.3) is used in this Handbook in the calculation of the thermodynamic properties of various gases.

Calculation of the thermodynamic functions with the equation of state in virial form. As was shown above, the dependence of the fundamental thermodynamic functions on pressure is easy to find through the general thermodynamic relations and the equation of state. For an equation of state in virial form (P5.3) the fundamental thermodynamic functions can be written as follows:

- 1) Internal energy

$$E - E^\circ = -RT \left(\frac{T}{V} \frac{dB}{dT} + \frac{T}{2V} \frac{dC}{dT} + \dots \right). \quad (P5.8)$$

2) Enthalpy

$$H - H^\circ = RT \left[\frac{1}{V} \left(B - T \frac{dB}{dT} \right) + \frac{1}{V^2} \left(C - \frac{1}{2} T \frac{dC}{dT} \right) + \dots \right]. \quad (P5.9)$$

3) Entropy

$$S - S^\circ = -R \ln p - R \left(\frac{T}{V} \frac{dB}{dT} + \frac{B^2}{2V} + \frac{T}{2V^2} \frac{dC}{dT} + \dots \right) \quad (P5.10)$$

4) Specific heat capacity at constant volume

$$C_v - C_v^\circ = -R \left(\frac{2T}{V} \frac{dB}{dT} + \frac{T^2}{V} \frac{d^2B}{dT^2} + \frac{T}{V^2} \frac{dC}{dT} + \frac{T^2}{2V^2} \frac{d^2C}{dT^2} + \dots \right). \quad (P5.11)$$

5) Specific heat capacity at constant pressure

$$C_p - C_p^\circ = -R \left(\frac{T^2}{V} \frac{d^2B}{dT^2} - \frac{B - T \frac{dB}{dT} - C + T \frac{dC}{dT} - \frac{1}{2} T^2 \frac{d^2C}{dT^2}}{V^2} + \dots \right). \quad (P5.12)$$

Here, $E - E^\circ$, $H - H^\circ$, $S - S^\circ$, $C_v - C_v^\circ$ and $C_p - C_p^\circ$ are the corrections accounting for the departures from the properties of an ideal gas for internal energy, enthalpy, entropy, specific heat capacity at constant volume, and specific heat capacity at constant pressure, respectively.

Under certain conditions the thermodynamic properties can be calculated with an equation of state having only three virial coefficients:

$$pV = RT \left(1 + \frac{B}{V} + \frac{C}{V^2} \right). \quad (P5.13)$$

As Nelson, Obert, and Evanston [3088] have shown, the error in the thermodynamic quantities when calculated by means of the equation of state (P5.13) is appreciable only at pressures twice as high as the critical and at temperatures below the critical.

At temperatures above 2000°K and pressures up to 250 atm the molal specific volume of a gas is of the order of $10^3 \text{ cm}^3/\text{mole}$. As B amounts to $10\text{-}30 \text{ cm}^3/\text{mole}$ and C to $100\text{-}500 \text{ cm}^6/\text{mole}^2$, we have $B/V = 0.03$, $C/V^2 = 0.0002$. These quantities are small as compared to unity; therefore it can be assumed in the equation of state (P5.13) that the volume

of the gas is equal to that of an ideal gas, and the term C/V^2 can be neglected. Thus

$$V = \frac{RT}{p} \left(1 + \frac{Bp}{RT} \right). \quad (\text{P5.14})$$

It is expedient to use such a simple relation between volume and pressure for calculations with Eqs. (P5.8)-(P5.12) within the above ranges of temperature and pressure. Moreover, the volume calculated from the equation of state of an ideal gas may be inserted in the terms with C . After some other simplifications, Eqs. (P5.9) and (P5.10) assume the following form which is convenient for calculations:

$$H_T - H_T^\circ = 0.0242p \left(B - T \frac{dB}{dT} \right) + \frac{0.000295}{T} \left(C - B^2 - \frac{1}{2} T \frac{dC}{dT} \right) p^2, \quad (\text{P5.15})$$

$$S_T - S_T^\circ = -0.0242p \frac{dB}{dT} - 0.000295 \frac{p^2}{T^2} \left(\frac{B^2}{2} - \frac{C}{2} + \frac{T}{2} \frac{dC}{dT} \right), \quad (\text{P5.16})$$

$$\Phi_T^\circ - \Phi_T^{\circ\text{MA}} = -0.0242 \frac{p}{T} B - 0.000147 \frac{p^2}{T} (C + B^2), \quad (\text{P5.17})$$

where

$$\Phi_T^{\circ\text{MA}} = S_T^{\circ\text{MA}} - \frac{H_T^\circ - H_0^\circ}{T}, \quad (\text{P5.18})$$

$$S_T^{\circ\text{MA}} = S_T^\circ - R \ln p. \quad (\text{P5.19})$$

The derivatives of the virial coefficients of nonpolar gases, which are necessary for calculating the thermodynamic quantities, can be calculated from the tables 311 and 312. For polar gases these quantities have been determined graphically.

Volume II of this Handbook contains tables of the virial coefficients and their derivatives for all gases examined in this Appendix. The corrections to be added to the thermodynamic functions of an ideal gas at various temperatures and pressures have been calculated for illustration from these tables and from Eqs. (P5.9) and (P5.10) for Ar , H_2 , O_2 , H_2O , CO_2 , and CH_4 ; they are presented in the tables 305-310.

TABLE 305

Corrections for the Nonideal Behavior to the Thermodynamic Functions of Ar

T, °K	1 $H_T - H_T^0$, кал/моль			2 $S_T - S_T^{id}$, кал/моль·град		
	3 p, атм					
	1	100	250	1	100	250
500	-0,6	-51,9	-107,7	-0,0015	-0,1427	-0,3287
1000	0,2	24,5	66,3	-0,0003	-0,0272	-0,0599
1500	0,4	43,8	111,2	-0,0001	-0,0104	-0,0249
2000	0,5	54,3	136,0	—	-0,0042	-0,0103
3000	0,6	64,4	160,8	—	-0,0021	-0,0053
4000	0,7	66,6	166,3	—	-0,0016	-0,0040
5000	0,7	69,5	169,5	—	-0,0012	-0,0031
6000	0,7	67,6	169,4	—	-0,0010	-0,0026

1) cal/mole; 2) cal/mole·deg; 3) atm.

TABLE 306

Corrections for the Nonideal Behavior to the Thermodynamic Functions of H₂

T, °K	1 $H_T - H_T^0$, кал/моль			2 $S_T - S_T^{id}$, кал/моль·град		
	3 p, атм					
	1	100	250	1	100	250
500	0,3	28,0	71,7	-0,0002	-0,0167	-0,0375
1000	0,4	41,2	102,6	—	0,0008	0,0020
1500	0,4	42,8	106,6	—	0,0020	0,0052
2000	0,4	43,0	107,3	—	0,0009	0,0021
3000	0,4	42,0	104,9	—	0,0017	0,0043
4000	0,4	41,2	102,9	—	0,0014	0,0035
5000	0,4	41,2	102,9	—	0,0011	0,0028
6000	0,4	41,3	103,0	—	0,0010	0,0024

1) cal/mole; 2) cal/mole·deg; 3) atm.

2. METHODS OF DETERMINING THE VIRIAL COEFFICIENTS

Experimental methods. The virial coefficients can be gotten from experimental p-V-T data in several ways. In one of these, Eq. (P5.3) is changed to the form

$$A = \left(\frac{pV}{RT} - 1\right)V = B + \frac{C}{V} + \dots \quad (\text{P5.20})$$

Here it can be seen that the second virial coefficient is given by the expression

$$B(T) = \lim_{p \rightarrow 0} \left(\frac{pV}{RT} - 1\right)V = \lim_{p \rightarrow 0} A. \quad (\text{P5.21})$$

It is easy to obtain from Eq. (P5.20) also the expression for the third virial coefficient

$$C(T) = \lim_{p \rightarrow 0} (A - B)V. \quad (\text{P5.22})$$

TABLE 307

Corrections for the Nonideal Behavior to the Thermodynamic Functions of O_2

T, °K	1 $H_T - H_T^\circ$, кал/моль			2 $S_T - S_T^{\text{ид}}$, кал/моль·град		
	3 p, атм					
	1	100	250	1	100	250
500	-0,6	-57,8	-104,6	-0,0016	-0,1501	-0,3378
1000	0,3	28,3	76,5	-0,0003	-0,0286	-0,0663
1500	0,5	48,7	124,7	-0,0001	-0,0108	-0,0256
2000	0,6	60,0	150,3	—	-0,0042	-0,0104
3000	0,7	70,4	175,9	—	0,0002	-0,0005
4000	0,7	72,7	181,5	—	0,0010	0,0024
5000	0,7	73,5	183,6	—	0,0011	0,0029
6000	0,7	73,6	183,8	—	0,0012	0,0029

1) cal/mole; 2) cal/mole·deg; 3) atm.

This method of determining B and C requires very accurate data on the compressibility of the gases at very low densities.

Another method is used when the experimental data on the compressibility of a gas are available in the form of a finite polynomial in powers of $1/V$:

$$\frac{pV}{RT} = 1 + \frac{A_1(T)}{V} + \frac{A_2(T)}{V^2} + \dots + \frac{A_n(T)}{V^n}. \quad (\text{P5.23})$$

Here, different values of the constants A_j are taken for different

temperatures. These values are determined by the least-squares method or by another method of finding an equation for a curve rendering experimental data. The resulting quantities $A_j(T)$ are directly taken to be the virial coefficients. When the experimental data are evaluated

TABLE 308

Corrections for the Nonideal Behavior to the Thermodynamic Functions of H_2O

T, °K	1 $H_T - H_T^\circ$, ккал/моль			2 $S_T - S_T^\circ$, ккал/моль·град		
	3 p, атм					
	1	100	250	1	100	250
1000	-2,5	-249,0	-634,6	-0,0016	-0,1963	-0,4202
1500	-1,0	-96,2	-237,9	-0,0006	-0,0538	-0,1271
2000	-0,4	-39,7	-97,4	-0,0002	-0,0217	-0,0538
3000	-0,1	-5,5	-13,2	-0,0001	-0,0079	-0,0197
4000	0,1	11,1	27,9	—	-0,0030	-0,0075
5000	0,1	14,0	34,7	—	-0,0022	-0,0054
6000	0,2	16,9	41,9	—	-0,0015	-0,0039

1) cal/mole; 2) cal/mole·deg; 3) atm.

TABLE 309

Corrections for the Nonideal Behavior to the Thermodynamic Functions of CO_2

T, °K	1 $H_T - H_T^\circ$, ккал/моль			2 $S_T - S_T^\circ$, ккал/моль·град		
	3 p, атм					
	1	100	250	1	100	250
1000	-0,5	-42,2	-83,2	-0,0006	-0,0573	-0,1302
1500	0,2	21,8	64,0	-0,0005	-0,0468	-0,0984
2000	0,5	51,3	131,4	-0,0002	-0,0161	-0,0395
3000	0,7	75,0	188,3	-0,0001	-0,0059	-0,0144
4000	0,9	89,9	233,9	—	-0,0014	-0,0035
5000	1,0	96,1	238,1	—	—	—
6000	1,0	98,4	243,4	—	0,0005	0,0013

1) cal/mole; 2) cal/mole·deg; 3) atm.

TABLE 310

Corrections for the Nonideal Behavior to the Thermodynamic Functions of CH_4

T, °K	1 $H_T - H_T^\circ$, кал/моль			2 $S_T - S_T^{\text{ид}}$, кал/моль·град		
	3 p, атм					
	1	100	250	1	100	250
1000	0,1	11,8	40,7	-0,0006	-0,0556	-0,1192
1500	0,3	32,5	91,5	-0,0002	-0,0192	-0,0449
2000	0,6	65,1	143,7	-0,0001	-0,0081	-0,0189
3000	0,8	84,7	211,8	—	-0,0015	-0,0001
4000	0,9	91,7	228,9	—	0,0002	0,0011
5000	0,9	93,6	233,8	—	0,0009	0,0057
6000	0,9	94,7	236,4	—	0,0011	0,0069

1) cal/mole; 2) cal/mole·deg; 3) atm.

in this way one has to be careful because the $A_j(T)$ depend on the pressure range in which the measurements have been made as well as on the number of terms in the series (P5.23). Hence, as long as the experimental data do not cover sufficiently low pressures, the function A_1 cannot be the same as the second virial coefficient. As most of the precise measurements of the compressibility of gases are made at medium and high pressures it is difficult to get reliable values of the virial coefficients out of such measurements. Moreover, the coefficient $A_2(T)$ and the subsequent coefficients are very responsive to the degree of the polynomial chosen; in the calculation of the third virial coefficient from experimental data one is therefore likely to encounter great difficulties.

Experimenters often express their results in the form of a polynomial other than (P5.23). Dutch scientists (Michels and collaborators in Amsterdam and Kamerling Onnes and collaborators at Leyden) take the physical atmosphere to be the unit of pressure, and the molal specific volume of a gas at the pressure of 1 physical atmosphere at a tempera-

ture of 0°C to be the unit of volume. This standard unit of volume is known as the Amagat unit of volume; it is different for each gas since the gases are not ideal at a pressure of 1 atm.

The volume of a gas at any pressure and temperature is given by the ratio of the actual gas volume to the standard unit volume

$$V_a = \frac{V}{V_{\text{станд}}}, \quad (\text{P5.24})$$

where V_a is the volume in Amagat units. Similarly, the density of a gas in Amagat units is

$$\rho_a = \frac{1}{V_a} = \frac{V_{\text{станд}}}{V}, \quad (\text{P5.25})$$

and the equation of state can be written in the form

$$pV_a = a + bp_a + cp_a^2 + dp_a^3 + \dots \quad (\text{P5.26})$$

By comparing this equation with Eq. (P5.3) we can see that the virial coefficients B, C, D are related to the coefficients a, b, c, d in the following way:

$$B = \frac{b}{a} V_{\text{станд}}, \quad (\text{P5.27})$$

$$C = \frac{c}{a} V_{\text{станд}}^2, \quad (\text{P5.28})$$

$$D = \frac{d}{a} V_{\text{станд}}^3, \quad (\text{P5.29})$$

where

$$V_{\text{станд}} = \frac{RT}{a}. \quad (\text{P5.30})$$

In general these relations can be considered only as an approximation for determining the virial coefficients because they hold true only when Eq. (P5.26) is an infinite series.

The German scientists (in particular, Holborn and Otto) use the pressure of 100 cm Hg for a unit. The unit volume they use is that of a mole of gas at 100 cm Hg and 0°C.

The equation of state is written in the familiar form

$$pV = \bar{A}' + \bar{B}'p + \bar{C}'p^2 + \dots, \quad (\text{P5.31})$$

where \bar{A}' , \bar{B}' , \bar{C}' are functions of temperature. The virial coefficients in this case can be expressed in terms of the quantities \bar{A}' , \bar{B}' , \bar{C}' as follows:

$$B = 0.76 RT \frac{\bar{B}'}{\bar{A}'}, \quad (\text{P5.32})$$

$$C = 0.76 RT \left(\frac{\bar{C}'}{\bar{A}'} + \bar{B}'^2 \right). \quad (\text{P5.33})$$

Like the former, the expressions (P5.32) and (P5.33) are just approximations when the series (P5.31) is not infinite.

Empirical relations. It is sometimes convenient to use approximate formulas for B as a function of temperature to estimate the magnitude of the second virial coefficient as well as to carry out interpolations:

$$\frac{B}{V_{sp}} = 0.438 - 0.881 \frac{T_{sp}}{T} - 0.757 \left(\frac{T_{sp}}{T} \right)^2 \quad (\text{P5.34})$$

or

$$\frac{B}{V_{sp}} = 0.461 - 1.158 \frac{T_{sp}}{T} - 0.503 \left(\frac{T_{sp}}{T} \right)^2. \quad (\text{P5.35})$$

The formula (P5.34), which was suggested by Stockmayer and Beattie [3870], as well as the formula (P5.35) suggested by Beattie and Bridgeman [703] for nonpolar gases above critical temperature produce the same values of the second virial coefficients. To estimate B at temperatures below the critical the known Berthelot equation

$$\frac{Bp_{kr}}{RT_{sp}} = \frac{9}{128} \left[1 - 6 \left(\frac{T_{sp}}{T} \right)^2 \right]. \quad (\text{P5.36})$$

is often used. Assigning a value of 0.282 to the term $(p_{kr} \cdot V_{kr})/RT_{kr}$ in this equation, which is justified for quite a large number of compounds [1590, 1879, 2547], we obtain

$$B = V_{sp} \left[0.25 - 1.5 \left(\frac{T_{sp}}{T} \right)^2 \right]. \quad (\text{P5.37})$$

As shown by Fox and Lambert [1590], this equation is sufficiently accu-

rate. The values of B for nonpolar vapors, as calculated from the above equations, are in good agreement with the experimental values of B for reduced temperatures T/T_{kr} ranging from 0.6 to 1.0. The B values for polar vapors, calculated from the critical constants according to Eq. (P5.36) are always considerably lower than the experimental values. If this departure is caused by a slight dimerization then the experimental values of the second virial coefficients can be written as

$$B_{\text{non}} = B + B_g \quad (\text{P5.38})$$

where B is the value to be calculated from Eq. (P5.36) and B_g a quantity representing the effect of reversible dimerization. The value of B_g can be calculated from the formula [2550]

$$B_g = -\frac{RT}{K_p} \quad (\text{P5.39})$$

where K_p is the dissociation constant of the dimer.

Theoretical calculation of the virial coefficients. Classical statistical mechanics supplies the following expressions for the second and the third virial coefficient:

$$B(T) = -\frac{N}{4} \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} f_{12} \sin \theta_1 d\theta_1 \sin \theta_2 d\theta_2 d(\varphi_2 - \varphi_1) r_{12}^2 dr_{12} \quad (\text{P5.40})$$

$$C(T) = -\frac{8\pi^3 N^2}{3} \iiint f_{12} f_{13} f_{23} r_{12} r_{13} r_{23} dr_{12} dr_{13} dr_{23} \quad (\text{P5.41})$$

(r_{12} , r_{13} , and r_{23} together form a triangle), where $f_{ij} = e^{-\frac{U_{ij}}{kT}} - 1$, U is the potential function of intermolecular interaction (intermolecular potential), N is Avogadro's number, k the Boltzmann constant, and r_{ij} the distance from the i th to the j th molecule.

Several different kinds of intermolecular potential are known at present (Buckingham (6-exp), Buckingham-Corner, Stockmayer, Lennard-Jones, etc.).

The Lennard-Jones (6-12) intermolecular potential*

$$U(r) = 4\epsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right]. \quad (\text{P5.42})$$

is often used in the calculation of the virial coefficients of nonpolar gases; ϵ is the minimum potential energy of interaction between the gas molecules, corresponding to a state of equilibrium, and σ is the collision diameter of a molecule.

The virial coefficients of polar gases can be calculated from the Stockmayer intermolecular potential:

$$U(r, \theta_1, \theta_2, \varphi_2 - \varphi_1) = 4\epsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right] - \frac{\mu^2}{r^3} [2 \cos \theta_1 \cos \theta_2 - \sin \theta_1 \sin \theta_2 \cos(\varphi_2 - \varphi_1)]. \quad (\text{P5.43})$$

Here, ϵ and σ have the same physical meaning like in the Lennard-Jones potential, μ is the dipole moment, θ_1 and θ_2 are the angles enclosed between the dipoles and the lines connecting the centers of the dipoles, and φ_2 and φ_1 are the angles between the planes going through the central line and the axis of the dipoles.

Apart from these two potential functions also other types of intermolecular potentials are used in the calculation of the virial coefficients. A detailed survey on various types of intermolecular potentials is given in a book by Hirschfelder, Curtiss, and Bird [2082]. In this Handbook the virial coefficients of nonpolar gases have been calculated by means of the Lennard-Jones (6-12) intermolecular potential.

The choice of this potential function is based upon the fact that numerous experiments showed good agreement between the values of the virial coefficients calculated by means of a Lennard-Jones potential and the values immediately obtained from the experiment for many nonpolar molecules. We have, indeed, to be aware of the fact that a potential function with the parameters ϵ and σ determined from the experimental values of the second virial coefficient obtained at low temperatures generally cannot be applied to the calculation of the values of

TABLE 311

The Second Virial Coefficient of Nonpolar Gases and Its Derivative With Respect to T^*

T^*	B^*	$T^* \frac{dB^*}{dT^*}$	T^*	B^*	$T^* \frac{dB^*}{dT^*}$
0,30	-27,880581	76,607256	2,70	-0,22358628	1,1000353
0,35	-18,754895	45,247713	2,80	-0,18450728	1,0494802
0,40	-13,798835	30,267080	2,90	-0,14850215	1,0029572
0,45	-10,754975	21,989482	3,00	-0,11523390	0,9600031
0,50	-8,720205	13,823690	3,10	-0,08441245	0,9202229
0,55	-7,2740858	13,582156	3,20	-0,05578896	0,8832774
0,60	-6,1979708	11,248849	3,30	-0,02913997	0,8488746
0,65	-5,3681918	9,5455096	3,40	-0,00428086	0,8167606
0,70	-4,7100370	8,2571145	3,50	+0,01895684	0,7867145
0,75	-4,1759283	7,2540135	3,60	0,04072012	0,7585430
0,80	-3,7342254	6,4541400	3,70	0,06113882	0,7320758
0,85	-3,3631193	5,8034061	3,80	0,08032793	0,7071630
0,90	-3,0471143	5,2649184	3,90	0,09839014	0,6836715
0,95	-2,7749102	4,8127607	4,00	0,11541691	0,6614830
1,00	-2,5380814	4,4282816	4,10	0,13149021	0,6404922
1,05	-2,3302208	4,0976659	4,20	0,14668372	0,6206045
1,10	-2,1463742	3,8106421	4,30	0,16106381	0,6017352
1,15	-1,9826492	3,5592925	4,40	0,17469039	0,5838082
1,20	-1,8359492	3,3374893	4,50	0,18761774	0,5667545
1,25	-1,7037784	3,1404074	4,60	0,19989511	0,5505118
1,30	-1,5841047	2,9642040	4,70	0,21156728	0,5350237
1,35	-1,4752571	2,8057826	4,80	0,22267507	0,5202387
1,40	-1,3758479	2,6626207	4,90	0,23325577	0,5051101
1,45	-1,2847160	2,5326459	5,0	0,24334351	0,4925951
1,50	-1,2006832	2,4141403	6,0	0,32390437	0,3839722
1,55	-1,1235183	2,3056683	7,0	0,37608846	0,3082566
1,60	-1,0519115	2,2060215	8,0	0,41343396	0,2524801
1,65	-0,98545337	2,1141772	9,0	0,44059784	0,2097011
1,70	-0,92361639	2,0292621	10,0	0,46087529	0,1758670
1,75	-0,86594279	1,9505276	20,0	0,52537420	0,0286638
1,80	-0,81203328	1,8773287	30,0	0,52692546	-0,0174929
1,85	-0,76153734	1,8091057	40,0	0,51857502	-0,0303115
1,90	-0,71414733	1,7453722	50,0	0,50836143	-0,0516478
1,95	-0,66959030	1,6857016	60,0	0,49821261	-0,0593621
2,00	-0,62762535	1,6297207	70,0	0,48865069	-0,0645039
2,10	-0,55063308	1,5275444	80,0	0,47979009	-0,0680819
2,20	-0,48170997	1,4366294	90,0	0,47161504	-0,0706470
2,30	-0,41967761	1,3552188	100,0	0,46406948	-0,0725244
2,40	-0,36357566	1,2819016	200,0	0,41143168	-0,0775400
2,50	-0,31261340	1,2155320	300,0	0,39012787	-0,0765245
2,60	-0,26613345	1,1551691	400,0	0,35835117	-0,0747534

TABLE 312

The Third Virial Coefficient of Nonpolar Gases and Its Derivative With Respect to T^*

T^*	C^*	$T^* \frac{dC^*}{dT^*}$	T^*	C^*	$T^* \frac{dC^*}{dT^*}$
0,70	-3,37664	28,68	3,10	0,34842	-0,1148
0,75	-1,79197	18,05	3,20	0,34491	-0,1060
0,80	-0,84953	11,60	3,30	0,34177	-0,09826
0,85	-0,27657	7,561	3,40	0,33894	-0,09133
0,90	+0,07650	4,953	3,50	0,33638	-0,08510
0,95	0,29509	3,234	3,60	0,33407	-0,07963
1,00	0,42966	2,078	3,70	0,33196	-0,07462
1,05	0,51080	1,292	3,80	0,33002	-0,07024
1,10	0,55762	0,7507	3,90	0,32825	-0,06634
1,15	0,58223	0,3760	4,00	0,32662	-0,06286
1,20	0,59240	+0,1159	4,10	0,32510	-0,05989
1,25	0,59326	-0,0646	4,20	0,32369	-0,05709
1,30	0,58815	-0,1889	4,30	0,32238	-0,05458
1,35	0,57933	-0,2731	4,40	0,32115	-0,05237
1,40	0,56831	-0,3288	4,50	0,32000	-0,05040
1,45	0,55611	-0,3641	4,60	0,31891	-0,04865
1,50	0,54339	-0,3845	4,70	0,31788	-0,04712
1,55	0,53059	-0,3943	4,80	0,31690	-0,04579
1,60	0,51803	-0,3963	4,90	0,31596	-0,04461
1,65	0,50587	-0,3929	5,00	0,31508	-0,04359
1,70	0,49425	-0,3858	6,0	0,30771	-0,03893
1,75	0,48320	-0,3759	7,0	0,30166	-0,03489
1,80	0,47277	-0,3643	8,0	0,29618	-0,04231
1,85	0,46296	-0,3516	9,0	0,29103	-0,04529
1,90	0,45376	-0,3382	10,0	0,28610	-0,04825
1,95	0,44515	-0,3245	20,0	0,24643	-0,06437
2,00	0,43710	-0,3109	30,0	0,21954	-0,06753
2,10	0,42260	-0,2840	40,0	0,20012	-0,06714
2,20	0,40999	-0,2588	50,0	0,18529	-0,06566
2,30	0,39900	-0,2355	60,0	0,17347	-0,06388
2,40	0,38943	-0,2142	70,0	0,16376	-0,06203
2,50	0,38108	-0,1950	80,0	0,15560	-0,06025
2,60	0,37378	-0,1777	90,0	0,14860	-0,05857
2,70	0,36737	-0,1621	100,0	0,14251	-0,05700
2,80	0,36173	-0,1482	200,0	0,10679	-0,04599
2,90	0,35675	-0,1352	300,0	0,08943	-0,03970
3,00	0,35234	-0,1247	400,0	0,07862	-0,03551

TABLE 313

The Second Virial Coefficient B* for Polar Gases

T*	P*						
	0.1	0.2	0.3	0.4	0.5	0.6	0.7
0.30	-31,129	-42,968	-72,01				
0.35	-20,355	-25,879	-38,07	-64,11			
0.40	-14,717	-17,777	-24,090	-36,28	-60,4		
0.45	-11,339	-13,241	-16,985	-23,733	-35,92	-58.8	
0.50	-9,1199	-10,401	-12,841	-17,026	-24,11	-36,36	-59,0
0.55	-7,5631	-8,4786	-10,181	-12,996	-17,53	-24,91	-37,3
0.60	-6,4159	-7,1001	-8,3495	-10,360	-13,477	-18,33	-26,0
0.65	-5,5381	-6,0677	-7,0213	-8,5234	-10,789	-14,185	-19,34
0.70	-4,8460	-5,2675	-6,0183	-7,1813	-8,8965	-11,394	-15,05
0.75	-4,2871	-4,6304	-5,2364	-6,1627	-7,5043	-9,413	-12,13
0.80	-3,8268	-4,1116	-4,6110	-5,3659	-6,4433	-7,9478	-10,040
0.85	-3,4414	-3,6815	-4,1000	-4,7271	-5,6113	-6,8267	-8,486
0.90	-3,1142	-3,3193	-3,6738	-4,2045	-4,9432	-5,9457	-7,292
0.95	-2,8330	-3,0103	-3,3168	-3,7695	-4,3961	-5,2373	-6,3523
1.00	-2,5889	-2,7437	-3,0102	-3,4021	-3,9406	-4,6567	-5,5953
1.05	-2,3750	-2,5114	-2,7455	-3,0881	-3,5559	-4,1732	-4,9744
1.10	-2,1862	-2,3072	-2,5145	-2,8167	-3,2271	-3,7649	-4,4572
1.15	-2,0183	-2,1265	-2,3113	-2,5799	-2,9430	-3,4160	-4,0203
1.20	-1,8680	-1,9653	-2,1312	-2,3716	-2,6952	-3,1146	-3,6471
1.25	-1,7328	-1,8208	-1,9706	-2,1870	-2,4773	-2,8519	-3,3249
1.30	-1,6105	-1,6905	-1,8264	-2,0223	-2,2844	-2,6211	-3,0442
1.35	-1,4994	-1,5724	-1,6963	-1,8746	-2,1124	-2,4168	-2,7976
1.40	-1,3980	-1,4649	-1,5784	-1,7413	-1,9581	-2,2348	-2,5795
1.45	-1,3051	-1,3667	-1,4710	-1,6502	-1,8190	-2,0717	-2,3854
1.50	-1,2197	-1,2766	-1,3728	-1,5106	-1,6931	-1,9247	-2,2115
1.55	-1,1410	-1,1937	-1,2827	-1,4101	-1,5785	-1,7917	-2,0549
1.60	-1,0681	-1,1171	-1,1998	-1,3179	-1,4738	-1,6708	-1,9133
1.65	-1,0006	-1,0462	-1,1232	-1,2330	-1,3778	-1,5604	-1,7846
1.70	-0,93775	-0,98038	-1,0523	-1,1547	-1,2896	-1,4594	-1,6674
1.75	-0,87917	-0,91908	-0,98633	-1,0821	-1,2079	-1,3662	-1,5597
1.80	-0,82445	-0,86190	-0,92498	-1,0147	-1,1325	-1,2804	-1,4610
1.85	-0,77322	-0,80844	-0,86772	-0,95197	-1,0625	-1,2011	-1,3699
1.90	-0,72516	-0,75834	-0,81417	-0,89345	-0,99736	-1,1275	-1,2858
1.95	-0,67998	-0,71130	-0,76398	-0,83873	-0,93662	-1,0590	-1,2078
2.00	-0,63745	-0,66707	-0,71686	-0,78747	-0,87985	-0,99526	-1,1353
2.10	-0,55947	-0,58607	-0,63076	-0,69408	-0,77679	-0,87993	-1,0048
2.20	-0,48969	-0,51373	-0,55409	-0,61121	-0,68570	-0,77850	-0,89060
2.30	-0,42893	-0,44877	-0,48540	-0,53721	-0,60472	-0,68865	-0,78989
2.40	-0,37020	-0,39012	-0,42354	-0,47076	-0,53224	-0,60858	-0,70049
2.50	-0,31868	-0,33695	-0,36578	-0,41079	-0,46702	-0,53676	-0,62063
2.60	-0,27172	-0,28852	-0,31668	-0,35642	-0,40807	-0,47205	-0,54292
2.70	-0,22875	-0,24428	-0,27025	-0,30692	-0,35455	-0,41347	-0,48420
2.80	-0,18929	-0,20366	-0,22774	-0,26167	-0,30572	-0,36020	-0,42552
2.90	-0,15295	-0,16630	-0,18867	-0,22018	-0,26106	-0,31159	-0,37210
3.00	-0,11937	-0,13182	-0,15266	-0,18200	-0,22005	-0,26705	-0,32330
3.10	-0,08828	-0,09991	-0,11927	-0,14677	-0,18229	-0,22612	-0,27854
3.20	-0,06941	-0,07030	-0,0885	-0,11417	-0,14740	-0,18639	-0,23738

TABLE 313 (continued)

0.8	0.9	1.0	1.1	1.2	1.3	1.4	1.5
-58.8							
-38.5	-59.7						
-27.3	-40.0						
-20.50	-28.2						
-16.05	-21.78	-41.8					
-12.973	-17.14	-30.4	-43.5				
-10.759	-13.90	-23.2	-32.0	-45.8			
-9.103	-11.612	-18.4	-24.7	-34.4	-47.0		
-7.828	-9.790	-14.92	-19.61	-26.3	-35.7	-50.3	
-6.820	-8.440	-12.42	-16.00	-21.0	-27.9	-37.8	-52.0
-6.008	-7.349	-10.54	-13.36	-17.2	-22.3	-29.8	-40.0
-5.3413	-6.4696	-9.08	-11.35	-14.3	-18.4	-24.3	-31.0
-4.7855	-5.7520	-7.915	-9.780	-12.21	-15.42	-19.6	-25.4
-4.3161	-5.1537	-6.976	-8.534	-10.53	-13.13	-16.5	-21.1
-3.9152	-4.6483	-6.203	-7.524	-9.20	-11.34	-14.1	-17.7
-3.5690	-4.2164	-5.559	-6.693	-8.11	-9.90	-12.2	-15.1
-3.2676	-3.8438	-5.014	-5.998	-7.21	-8.74	-10.7	-13.1
-3.0030	-3.5193	-4.5484	-5.4111	-6.471	-7.780	-9.41	-11.46
-2.7691	-3.2345	-4.1466	-4.9092	-5.839	-6.976	-8.38	-10.12
-2.5609	-2.9329	-3.7970	-4.4762	-5.298	-6.296	-7.51	-9.00
-2.3745	-2.7591	-3.4963	-4.0994	-4.831	-5.714	-6.78	-8.08
-2.2069	-2.5589	-3.2100	-3.7688	-4.426	-5.212	-6.15	-7.29
-2.0554	-2.3788	-2.9783	-3.4768	-4.069	-4.774	-5.612	-6.612
-1.9180	-2.2165	-2.7629	-3.2173	-3.7548	-4.3909	-5.145	-6.045
-1.7923	-2.0685	-2.5697	-2.9860	-3.4760	-4.0532	-4.735	-5.541
-1.6775	-1.9340	-2.3913	-2.7770	-3.2256	-3.7517	-4.370	-5.097
-1.5720	-1.8110	-2.2357	-2.5889	-3.0014	-3.4831	-4.046	-4.706
-1.4749	-1.6981	-2.0912	-2.4183	-2.7990	-3.2419	-3.758	-4.359
-1.3852	-1.5941	-1.9591	-2.2630	-2.6156	-3.0244	-3.498	-4.049
-1.3021	-1.4982	-1.8380	-2.1211	-2.4487	-2.8273	-3.265	-3.771
-1.1531	-1.4082	-1.7265	-1.9910	-2.2963	-2.6479	-3.053	-3.520
-1.0234	-1.3268	-1.6285	-1.7610	-2.0282	-2.3342	-2.6846	-3.0857
-0.90957	-1.1785	-1.5380	-1.5643	-1.8002	-2.0693	-2.3758	-2.7248
-0.80895	-1.0490	-1.4100	-1.3943	-1.6044	-1.8431	-2.1138	-2.4204
-0.71944	-0.93509	-1.2803	-1.2461	-1.4345	-1.6479	-1.8889	-2.1606
-0.63934	-0.83413	-1.06584	-1.1159	-1.2860	-1.4780	-1.6940	-1.9368
-0.56729	-0.74412	-0.86421	-1.0008	-1.1551	-1.3289	-1.5239	-1.7423
-0.50216	-0.66342	-0.77343	-0.89828	-1.0391	-1.1972	-1.3742	-1.5719
-0.44304	-0.59072	-0.69191	-0.80654	-0.93559	-1.0802	-1.2416	-1.4215
-0.38917	-0.52492	-0.61833	-0.72400	-0.84275	-0.97554	-1.1235	-1.2876
-0.33999	-0.46511	-0.55165	-0.64940	-0.75908	-0.86132	-1.0177	-1.1687
-0.29496	-0.41054	-0.49096	-0.58163	-0.66333	-0.79663	-0.92240	-1.0616
	-0.36057	-0.43552	-0.51998	-0.61449	-0.71967	-0.83625	-0.96505

TABLE 313 (continued)

T°	t°						
	0.1	0.2	0.3	0.4	0.5	0.6	0.7
3.30	-0,03254	-0,04277	-0,05986	-0,08393	-0,11509	-0,15351	-0,19940
3.40	-0,00748	-0,01710	-0,03318	-0,05580	-0,08509	-0,12118	-0,16427
3.50	+0,01594	+0,00698	-0,00828	-0,02959	-0,05717	-0,09115	-0,13169
3.60	0,03787	0,02931	+0,01501	-0,00511	-0,03113	-0,06318	-0,10140
3.70	0,05844	0,05035	0,03682	+0,01780	-0,00680	-0,03708	-0,07318
3.80	0,07778	0,07011	0,05729	0,03928	+0,01596	-0,01268	-0,04684
3.90	0,09597	0,08869	0,07653	0,05944	0,03736	+0,01018	-0,02219
4.00	0,11312	0,10620	0,09465	0,07841	0,05744	0,03463	+0,00091
4.10	0,12930	0,12272	0,11173	0,09628	0,07633	0,05180	0,02259
4.20	0,14460	0,13833	0,12786	0,11314	0,09414	0,07078	0,04299
4.30	0,15907	0,15309	0,14310	0,12907	0,11095	0,08868	0,06218
4.40	0,17279	0,16708	0,15754	0,14414	0,12684	0,10558	0,08029
4.50	0,18580	0,18034	0,17122	0,15841	0,14187	0,12155	0,09740
4.60	0,19815	0,19293	0,18420	0,17194	0,15611	0,13668	0,11357
4.70	0,20990	0,20489	0,19652	0,18478	0,16962	0,15101	0,12888
4.80	0,22107	0,21627	0,20825	0,19699	0,18245	0,16461	0,14340
4.90	0,23172	0,22711	0,21941	0,20860	0,19465	0,17752	0,15718
5.0	0,24187	0,23744	0,23004	0,21965	0,20625	0,18990	0,17026
6.0	0,32187	0,31877	0,31360	0,30634	0,29699	0,28552	0,27191
7.0	0,37532	0,37302	0,36918	0,36380	0,35687	0,34838	0,33832
8.0	0,41284	0,41106	0,40809	0,40393	0,39857	0,39201	0,38424
9.0	0,44012	0,43870	0,43633	0,43301	0,42873	0,42349	0,41729
10.0	0,46049	0,45932	0,45738	0,45463	0,45116	0,44687	0,44179
20.0	0,52527	0,52495	0,52441	0,52367	0,52271	0,52153	0,52014
30.0	0,52687	0,52672	0,52647	0,52611	0,52568	0,52510	0,52444
40.0	0,51854	0,51845	0,51830	0,51809	0,51782	0,51749	0,51710
50.0	0,50834	0,50828	0,50818	0,50804	0,50876	0,50764	0,50738
60.0	0,49820	0,49815	0,49808	0,49798	0,49785	0,49769	0,49750
70.0	0,48864	0,48861	0,48855	0,48847	0,48838	0,48826	0,48811
80.0	0,47978	0,47976	0,47971	0,47965	0,47957	0,47948	0,47937
90.0	0,47181	0,47159	0,47155	0,47150	0,47144	0,47136	0,47127
100.0	0,46406	0,46405	0,46402	0,46398	0,46392	0,46386	0,46379
200.0	0,41143	0,41142	0,41142	0,41140	0,41139	0,41137	0,41135
300.0	0,38013	0,38012	0,38012	0,38011	0,38011	0,38010	0,38009
400.0	0,35835	0,35835	0,35835	0,35834	0,35834	0,35833	0,35833

TABLE 313 (continued)

t°							
0.8	0.9	1.0	1.1	1.2	1.3	1.4	1.5
-0,25302	-0,31468	-0,38472	-0,46355	-0,55167	-0,64962	-0,75801	-0,87758
-0,21459	-0,27239	-0,33800	-0,41179	-0,49417	-0,58562	-0,68671	-0,79805
-0,17900	-0,23332	-0,29493	-0,36415	-0,44135	-0,52697	-0,62149	-0,72548
-0,14598	-0,19713	-0,25510	-0,32018	-0,39270	-0,47305	-0,56164	-0,65899
-0,11527	-0,16352	-0,21818	-0,27949	-0,34776	-0,42333	-0,50657	-0,59792
-0,08664	-0,13225	-0,18388	-0,24176	-0,30615	-0,37736	-0,45574	-0,54166
-0,05989	-0,10308	-0,15193	-0,20667	-0,26752	-0,33476	-0,40870	-0,48968
-0,03486	-0,07582	-0,12213	-0,17397	-0,23158	-0,29519	-0,36507	-0,44155
-0,01140	-0,05030	-0,09426	-0,14345	-0,19807	-0,25834	-0,32451	-0,39687
+0,01064	-0,02636	-0,06815	-0,11490	-0,16677	-0,22397	-0,28673	-0,35530
0,03137	-0,00387	-0,04366	-0,08814	-0,13747	-0,19184	-0,25145	-0,31654
0,05089	+0,01729	-0,02064	-0,06302	-0,11000	-0,16175	-0,21846	-0,28033
0,06932	0,03723	+0,00103	-0,03941	-0,08421	-0,13353	-0,18755	-0,24645
0,08672	0,05605	0,02145	-0,01717	-0,05995	-0,10702	-0,15854	-0,21469
0,10318	0,07383	0,04073	+0,00380	-0,03709	-0,08207	-0,13127	-0,18486
0,11877	0,09065	0,05896	0,02360	-0,01554	-0,05856	-0,10560	-0,15681
0,13355	0,10659	0,07620	0,04232	+0,00483	-0,03637	-0,08140	-0,13039
0,14758	0,12170	0,09255	0,06004	0,02409	-0,01540	-0,05855	-0,10548
0,25614	0,23818	0,21799	0,19554	0,17077	+0,14364	+0,11409	+0,08206
0,32667	0,31341	0,29853	0,28199	0,26379	0,24389	0,22225	0,19885
0,37524	0,36502	0,35355	0,34082	0,32682	0,31154	0,29494	0,27702
0,41012	0,40197	0,39283	0,38270	0,37157	0,35942	0,34625	0,33203
0,43593	0,42927	0,42180	0,41353	0,40444	0,39453	0,38379	0,37221
0,51854	0,51672	0,51468	0,51243	0,50996	0,50728	0,50438	0,50125
0,52367	0,52281	0,52184	0,52077	0,51970	0,51833	0,51695	0,51547
0,51665	0,51613	0,51556	0,51493	0,51423	0,51348	0,51266	0,51179
0,50707	0,50673	0,50635	0,50593	0,50546	0,50496	0,50441	0,50383
0,49729	0,49704	0,49676	0,49643	0,49613	0,49576	0,49537	0,49495
0,48795	0,48776	0,48755	0,48732	0,48707	0,48679	0,48650	0,48618
0,47924	0,47909	0,47893	0,47874	0,47854	0,47833	0,47810	0,47784
0,47117	0,47105	0,47091	0,47077	0,47061	0,47043	0,47024	0,47004
0,46370	0,46360	0,46349	0,46337	0,46323	0,46309	0,46293	0,46276
0,41132	0,41129	0,41126	0,41123	0,41119	0,41115	0,41110	0,41105
0,38008	0,38006	0,38005	0,38003	0,38001	0,37999	0,37997	0,37994
0,35832	0,35831	0,35830	0,35829	0,35828	0,35827	0,35825	0,35824

the virial coefficients at high temperatures and may give rise to considerable errors. The calculation of the virial coefficients by means of the force constants of the intermolecular potential obtained from measurements of viscosity, diffusion or thermal conductivity may in some cases also yield values that differ essentially from the experimental ones. Many authors [539, 540, 541, 543, 544, 2801] assume that a potential function calculated from scattering experiments in molecular beams is the most reliable intermolecular potential function for high temperatures. Unfortunately, such a calculation of potential functions encounters considerable difficulties and, up to now, similar calculations have only been carried out for some of the simplest atomic systems. As shown by Amdur and Ross [544], the difference in the values of the second virial coefficients calculated by means of different intermolecular potentials does not exceed 20% for temperatures up to 6000°K. Besides, although the error in the determination of the virial coefficients grows with increasing temperature, at the same time (if the pressure is constant) the relative magnitude of the nonideality correction to the thermodynamic functions is reduced. These two circumstances permit the use of the intermolecular Lennard-Jones potential with values of the force constants obtained at temperatures of 300-700°K, in order to calculate the virial coefficients at temperatures up to 6000°K.

As shown above, the second and third virial coefficients can be calculated with the help of Eqs. (P5.40) and (P5.41). In the general case, the solution of the equations (P5.40) and (P5.41) can be represented in the form [2082]

$$B(T) = b_0 B^*, \quad (\text{P5.44})$$

$$C(T) = b_0^2 C^*, \quad (\text{P5.45})$$

where $b_0 = \frac{2}{3} \pi N \sigma^3$, and B^* and C^* are functions of kT/ϵ for nonpolar gases and functions of kT/ϵ and $t^* = \frac{\mu^2}{\sqrt{8\epsilon\sigma^3}}$ for polar gases. The quantities

b_0 , ϵ/k and t^* are usually called the force constants of the intermolecular potential. Hirschfelder, McClure and Weeks [2084] have compiled tables of the values of B^* for both nonpolar and polar gases within a comparatively narrow interval of values of $T^* = kT/\epsilon$. In later works [821, 2082, 3543] the values of the function B^* were calculated for a wide interval of values of T^* . In the present Handbook Table 311 shows the values of B^* and its derivatives for a T^* -interval from 0.30 to 400, calculated by Bird and Spatz [821] with the help of an intermolecular Lennard-Jones potential.

Table 312 gives the values of C^* used in the calculation of the third virial coefficients of nonpolar gases. This table was borrowed from a paper of Bird, Spatz and Hirschfelder [822].

The virial coefficients of polar gases were calculated by means of Tables 313 and 314, compiled by Rowlinson [3543].

Quantum effects have to be taken into account in calculating the virial coefficients of light gases (He , H_2 , D_2). As shown by Hirschfelder, Curtiss and Bird [2082], allowing for the quantum corrections to the second virial coefficient is possible with the help of the formula

$$B^* + \Delta B_{\text{qm}}^* = B^* + \lambda^3 B_1^* + \lambda^6 B_{11}^* + \dots \quad (\text{P5.46})$$

where $\lambda^* = \frac{h}{\sigma \sqrt{m\epsilon}}$ is a quantum-mechanical parameter, m is the molecular mass, h is Planck's constant, and the functions B_1^* and B_{11}^* are determined by the equations

$$B_1^* = \sum_{l=1}^{\infty} b_l^* T^{-\frac{2l+1}{15}} \quad (\text{P5.47})$$

$$B_{11}^* = \sum_{l=1}^{\infty} b_{11}^* T^{-\frac{2l+1}{15}} \quad (\text{P5.48})$$

The values of b_l^* and b_{11}^* are given in Table 315 borrowed from a paper by De-Boer [1280]. Table 316 contains the accepted values of the quan-

tum-mechanical parameter λ^* for some light gases considered in the Handbook.

Table 314

Third Virial Coefficient C^* for Polar Gases

T^*	C^*											
	0,0	0,1	0,2	0,3	0,4	0,5	0,6	0,7	0,8	1,0	1,2	
1,0	0,4297	0,4440	0,5304	0,740								
1,2	0,5924	0,6177	0,7162	0,9216	1,268	1,78	2,5	3,4	4,6	7,0		
1,4	0,5883	0,5900	0,6679	0,8221	1,075	1,451	2,0	2,7	3,7	6,3	6,0	
1,6	0,5180	0,5351	0,5940	0,7075	0,8899	1,158	1,53	2,03	2,69	4,72	7,0	
1,8	0,4728	0,4861	0,5311	0,6161	0,7507	0,9455	1,214	1,572	2,03	3,36	5,2	
2,0	0,4371	0,4478	0,4826	0,5478	0,6496	0,7957	0,995	1,257	1,595	2,46	4,0	
2,5	0,3811	0,3873	0,4078	0,4445	0,5195	0,5807	0,6871	0,825	0,999	1,482	2,19	
3,0	0,3523	0,3563	0,3692	0,3924	0,4275	0,4761	0,5403	0,6223	0,7248	1,002	1,401	
4,0	0,3268	0,3286	0,3350	0,3463	0,3630	0,3859	0,4156	0,4529	0,4986	0,6194	0,7857	
6,0	0,3077	0,3085	0,3109	0,3151	0,3213	0,3298	0,3401	0,3532	0,3690	0,4095	0,4640	
8,0	0,2962	0,2966	0,2978	0,3000	0,3031	0,3072	0,3124	0,3188	0,3265	0,3459	0,3715	
10,0	0,2881	0,2883	0,2871	0,2884	0,2902	0,2926	0,2957	0,2995	0,3039	0,3151	0,3297	

TABLE 315

Values of Coefficients b_I^J and b_{II}^J

I	b_I^J	b_{II}^J	I	b_I^J	b_{II}^J
0	$8,297 \cdot 10^{-3}$	$-2,630 \cdot 10^{-3}$	7	$1,10 \cdot 10^{-3}$	$-5,02 \cdot 10^{-3}$
1	$6,11 \cdot 10^{-3}$	$-9,037 \cdot 10^{-3}$	8	$5,66 \cdot 10^{-3}$	$-2,941 \cdot 10^{-3}$
2	$7,65 \cdot 10^{-3}$	$-2,549 \cdot 10^{-3}$	9	$2,692 \cdot 10^{-3}$	$-1,584 \cdot 10^{-3}$
3	$6,79 \cdot 10^{-3}$	$-1,494 \cdot 10^{-3}$	10	$1,237 \cdot 10^{-3}$	$-8,03 \cdot 10^{-4}$
4	$5,903 \cdot 10^{-3}$	$-1,40 \cdot 10^{-3}$	11	$5,39 \cdot 10^{-3}$	$-3,839 \cdot 10^{-4}$
5	$3,362 \cdot 10^{-3}$	$-1,12 \cdot 10^{-3}$	12	$2,234 \cdot 10^{-3}$	$-1,74 \cdot 10^{-4}$
6	$2,01 \cdot 10^{-3}$	$-6,28 \cdot 10^{-3}$	13	$8,90 \cdot 10^{-4}$	$-7,53 \cdot 10^{-3}$

TABLE 316

Quantum Mechanical Parameter λ^* for Some Gases

1) Вещество	λ^*	1) Вещество	λ^*	1) Вещество	λ^*	1) Вещество	λ^*
He ^d	3,08	Kr	0,102	D ₂	1,223	CH ₄	0,239
He ^e	2,87	Xe	0,064	HT	1,223	N ₂	0,026
Ne	0,593	H ₂	1,724	DT	1,095	CO	0,220
Ar	0,186	HD	1,412	T ₂	1,00	O ₂	0,201

1) Substance.

3. CALCULATION OF THE THERMODYNAMIC PROPERTIES OF REAL GAS MIXTURES

The problem of calculating the thermodynamic functions of real gas mixtures for high pressures has not completely been solved, as yet. In many cases, however, satisfactory results can be obtained by making use of the calculation method given below.

In the case of a gas mixture, we may also use a series expansion of the virial coefficients B_m , C_m , etc.:

$$pV = RT \left(1 + \frac{B_m}{V} + \frac{C_m}{V^2} + \dots \right), \quad (\text{P5.49})$$

where the coefficients B_m , C_m , etc., are determined by the equations (see [2593])

$$B_m = \sum_{ik} x_i x_k B_{ik}, \quad (\text{P5.50})$$

$$C_m = \sum_{ikl} x_i x_k x_l C_{ikl}, \quad (\text{P5.51})$$

x_i , x_k , x_l are here the molar fractions of the components, and B_{ik} , B_{kk} , C_{iii} , etc., the virial coefficients of the pure gases.

The corrections to the thermodynamic properties of gases chemically not reacting with each other are calculated according to the formulas (P5.8)-(P5.12), with the difference that in the case considered the virial coefficients B_m and C_m of the mixture are applied, instead of B and C . The correction to the thermodynamic properties of the mixture as a whole is calculated.

In the calculation of corrections to the thermodynamic functions of mixtures of chemically interacting gases (e.g., in the calculation of the composition of combustion products at high pressures) the fact that in this case the equilibrium constant K_p is not only a function of temperature, but also of pressure has to be taken into account. A general method of calculating the composition of a chemically reacting gas mixture consists in obtaining the composition of the mixture under the

assumption that each of its components as well as the mixture as a whole are ideal. In this case, approximate values of the molar fractions of the components x_i are obtained. In order to take the influence of pressure on the equilibrium constants into account, "effective" second virial coefficients are calculated according to the formula

$$B_{k,ef} = \sum_i x_i B_{ki} \quad (P5.52)$$

where $B_{k,ef}$ is the "effective" second virial coefficient of the kth component, x_i is the molar fraction of the ith component, B_{ki} is the second virial coefficient due to pair interaction of the particles of the kth and ith components.

The following phase of calculation is the determination of the values of the equilibrium constants at a pressure equal to the pressure p of the whole mixture. This calculation is rendered necessary by the fact that the particles of each component interact not only with one another, but also with the particles of all other components of the mixture. This interaction is determined by the pressure of the whole mixture; consequently, also the magnitude of the constant depends on the total pressure of the whole mixture. In order to calculate the equilibrium constants the values of the functions H and Φ^* are determined according to the formulas (P5.15) and (P5.17), and the correction to the value of K_p is then determined according to the formula

$\delta \ln K_p = \frac{\delta \Delta \Phi^*}{4.57584}$. The new calculation of the composition is carried out on the basis of the values obtained for K_p . If necessary calculations are also carried out in the following approximations, in an analogous way. In order to calculate the composition of combustion products, however, it is, as a rule, sufficient to restrict oneself to the first approximation, even at pressures of the order of 400-600 atm.

Another method of calculating the compositions of mixtures of

chemically interacting real gases is the well-known method of volatilities. The principle of this method consists in the fact that the thermodynamic relations describing the equilibria of perfect gases are also applied to mixtures of real gases. This is achieved by substituting the partial pressures in these relations by special state functions f , called volatilities. The volatilities of the gases may be calculated in terms of the virial coefficients according to the formula [1588]

$$\ln \frac{f}{p} = \frac{2B}{V} + \frac{3}{2} \frac{C}{V^2} - \ln \frac{pV}{RT}. \quad (P5.53)$$

We have to note that the calculation of the virial coefficients of gas mixtures has, as yet, no serious theoretical foundation and is carried out by means of semiempirical formulas.

In the following, only the most widespread methods of calculating B_m and B_{ik} will be considered. The coefficients B_m depend on both the quantitative composition of the mixture and the temperature, but the coefficients B_{ik} are only functions of temperature. Consequently, the problem of calculating the virial coefficients B_m is essentially a problem of calculating the coefficients B_{ik} . All methods proposed have no rigorous theoretical foundation, and their acceptability, considered from the point of view of statistical mechanics, is different.

The simplest method of calculating B_{ik} is the method of additivity [283, 4372]. It is based on the assumption that the value of B_{ik} is equal to the arithmetical mean of B_{ii} and B_{kk} at the same temperature, i.e.,

$$B_{ik} = \frac{B_{ii} + B_{kk}}{2}. \quad (P5.54)$$

In this case

$$B_m = \sum x_i B_{ii}. \quad (P5.55)$$

This method of calculating B_{ik} - except for the case where the values of B_{ii} and B_{kk} are almost equal - has neither a theoretical nor an ex-

perimental foundation.

The method of combining the force constants [1465] is a widespread method of calculating the interaction coefficients B_{1k} . The application of this method consists in calculating the interaction coefficient according to Eq. (P5.44), and the force constants of the intermolecular potential of the two-component gas mixture are determined from the force constants of the pure gases according to the equations

$$\sigma_{12} = \frac{1}{2} (\sigma_1 + \sigma_2), \quad (\text{P5.56})$$

$$\epsilon_{12} = \sqrt{\epsilon_1 \epsilon_2}, \quad (\text{P5.57})$$

$$r^* = \frac{\mu_1 \mu_2}{\sqrt{8 \epsilon_{12} \sigma_{12}^3}} \approx \sqrt{r_1^* r_2^*}. \quad (\text{P5.58})$$

In the case of mixtures of polar gases with nonpolar ones the interaction between the polar and nonpolar molecules must be taken into account. As shown by Hirschfelder, Curtiss and Bird [2082], the expression for the interaction energy between a polar and a nonpolar molecule has the same form as for nonpolar molecules. In this case, the force constants of the mixture are determined from the following empirical formulas:

$$\sigma_{np} = \frac{1}{2} (\sigma_n + \sigma_p) \xi^{-\frac{1}{6}}, \quad (\text{P5.59})$$

$$\epsilon_{np} = \sqrt{\epsilon_n \cdot \epsilon_p} \xi^2, \quad (\text{P5.60})$$

where

$$\xi = 1 + \frac{\alpha_N \mu_p^2}{4 \sigma_p^3} \sqrt{\frac{\epsilon_n}{\epsilon_p}} \approx 1 + 0.892 \frac{\alpha_N r_p^*}{(h)_n} \cdot \sqrt{\frac{\epsilon_n}{\epsilon_p}}, \quad (\text{P5.61})$$

where α_N is the polarizability of the nonpolar molecules, and the subscripts "N" and "P" refer to nonpolar and polar molecules, respectively.

Tables 317 and 318 give the values of the dipole moments and the polarizabilities for some molecules. The tables presented are taken

from the book by Hirschfelder, Curtiss and Bird [2082]. Like the additivity method the combination method is theoretically founded only in the case of identical molecules.

TABLE 317
Polarizability α of Molecules

1 Молекула	$\alpha \cdot 10^{24}, \text{cm}^3$	2 Молекула	$\alpha \cdot 10^{24}, \text{cm}^3$	3 Молекула	$\alpha \cdot 10^{24}, \text{cm}^3$
H ₂	7,9	N ₂ O	30,0	CH ₄	28,0
N ₂	17,5	CO	19,5	C ₂ H ₄	42,6
O ₂	16,0	CO ₂	26,5	C ₂ H ₂	33,3
Cl ₂	46,1	C ₂ N ₂	50,1	CCl ₄	105

1) Molecule; 2) molecule; 3) molecule.

TABLE 318
Dipole Moments of Molecules

1 Молекула	2 $\mu, \text{дебай}$	3 Молекула	4 $\mu, \text{дебай}$	5 Молекула	6 $\mu, \text{дебай}$
H ₂ O	1,83	COS	0,72	CHF ₃	1,59
HF	1,91	HCN	2,93	CHCl ₃	1,05
HCl	1,03	CH ₃ F	1,82	CHBr ₃	1,30
HBr	0,79	CH ₂ Cl	1,89	CHF ₂ Cl	1,40
HJ	0,33	CH ₃ Br	1,78	CHFCI ₂	1,29
H ₂ S	0,93	CH ₃ J	1,59	CF ₂ Cl ₂	0,51
SO ₂	1,60	CH ₂ Cl ₂	1,58		
NH ₃	1,47	CH ₂ Br ₂	1,43		

1) Molecule; 2) Debye; 3) molecule; 4) Debye;
5) molecule; 6) Debye.

Using this method of calculating B_{12} , Beattie and Stockmayer [707] have obtained results, for a methane-butane mixture, and Reuss and Beenakker [3424] for H₂-Co and H₂-N₂ mixtures, which are in good agreement with the experimental ones. In the works of Gorski and Miller [1827] and Cottrell and collaborators [1191, 1192, 4234], besides results which are in good agreement with the experimental data (mixture of O₂ with N₂) also results are obtained that differ considerably from

the experimental ones (mixtures of CO₂ with O₂). Moreover, these authors established that for many mixtures investigated the temperature dependence of the calculated second virial coefficients differs widely from the temperature dependence of the values of B_m found by experiment. At present, no final conclusion can be drawn as to the range of applicability of the given method since the number of studies carried out as yet is insufficient. But as is shown by the results of works mentioned above, the application of the method of combining the force constants to mixtures of gases composed of nonpolar molecules approximately equal in diameter yields values of B_m that are in good agreement with the experimental ones.

Another widespread method of calculating the interaction coefficients B_m is the method proposed by Guggenheim and McGlashan [1879] which is based on the application of the principle of corresponding states. According to this principle the same function exists for all substances

$$\varphi\left(\frac{p}{p_{kr}}; \frac{V}{V_{kr}}; \frac{T}{T_{kr}}\right) = 0. \quad (P5.62)$$

In other words, if the gas properties are not expressed in terms of p, T and V, but in terms of the nondimensional parameters p/p_{kr}, T/T_{kr} and V/V_{kr}, it is possible to obtain a reduced equation of state (P5.62) which does not contain any of the quantities characterizing the real state of the gas. The constancy of the expression p_{kr}V_{kr}/RT_{kr} for all substances is the applicability criterion for the principle of corresponding states. In reality, however, this expression yields only approximately equal values for the individual groups of substances, and the principle of corresponding states can only be applied at the boundaries of these groups. Applying the principle of corresponding states to the calculation of the interaction coefficients B₁₂, we obtain:

$$\frac{B_{11}}{V_{11,sp}} = \varphi \left(\frac{T}{T_{11,sp}} \right), \quad (P5.63)$$

$$\frac{B_{22}}{V_{22,sp}} = \varphi \left(\frac{T}{T_{22,sp}} \right). \quad (P5.64)$$

Hence

$$\frac{B_{12}}{V_{12,sp}} = \varphi \left(\frac{T}{T_{12,sp}} \right). \quad (P5.65)$$

Guggenheim and McGlashan suggested the determination of the quantities $T_{12,kr}$ and $V_{12,kr}$ from the equations

$$T_{12,sp} = (T_{11,sp} \cdot T_{22,sp})^{\frac{1}{2}}, \quad (P5.66)$$

$$(V_{12,sp})^{\frac{1}{2}} = \frac{1}{2} (V_{11,sp})^{\frac{1}{2}} + \frac{1}{2} (V_{22,sp})^{\frac{1}{2}}, \quad (P5.67)$$

where the subscripts 11 and 22 denote quantities referring to the pure components.

The values of the second virial coefficients of nonpolar vapor mixtures calculated on the basis of the principle of corresponding states are, as shown in the works [192, 194, 1591, 3544, 3545], in satisfactory agreement with the experimental values. Studying the second virial coefficients of methane mixtures with $C(CH_3)_4$, $Si(CH_3)_4$ and SF_6 , Hamann, Lambert and Thomas [1938] have, however, obtained results that differ considerably from the values of B_m , calculated by means of the principle of corresponding states. In the case of mixtures of nonpolar vapors with polar ones, but also for mixtures of polar vapors, the molecular association caused by either dipole interaction or hydrogen bond must be taken into account in calculating the second virial coefficients of the mixtures. The proposed methods of calculating the second virial coefficients of such mixtures [1590, 2548, 2549] can only be applied if the degree of association is low.

We have to note that with high temperatures the application of this method of calculating B_{ik} to a calculation of the second virial

coefficients of the mixtures has neither a theoretical nor an experimental foundation.

Generally, the problem whether any of the methods of calculating the second virial coefficients of gas mixtures is applicable at high temperatures has not been solved, as yet. When this edition of the Handbook was prepared, calculations of the second virial coefficients of some mixtures of H_2 , O_2 , CO , CO_2 , H_2O and CH_4 in a temperature interval from 2000-6000°K were carried out by various methods. On the basis of these calculations it was found that the greatest discrepancy occurs between the quantities B_m calculated by the additivity method and those obtained with the method of combining the force constants in which case this discrepancy attains 40% for some mixtures in the temperature interval mentioned above. In all cases the values of B_m , calculated by the additivity method, were higher than the values of B_m , calculated by the method of combining the force constants. The corresponding discrepancy in the values of the enthalpy of the mixture at a pressure of 200 atm amounted only to some hundredths, and in the values of the mixture entropy to some thousandths of one percent.

4. p-V-T-DATA AND VIRIAL COEFFICIENTS OF SOME GASES

In this division, a survey is given of the works dealing with the determination of the p-V-T-data, the virial coefficients and force constants of the intermolecular potential. On the basis of this survey the values of the force constants of the intermolecular Lennard-Jones and Stockmayer potentials given in the Tables 319 and 320 were accepted in the Handbook for such gases for which reliable values are found in literature; in the second volume, tables of the virial coefficients and their derivatives are given for these gases.

O_2 (gas). A review of works published until 1948 on the investigation of the p-V-T-data of O_2 is given in the paper by Gratch [1842].

The most accurate p-V-T-data have been obtained by Meyers [2868] in the interval from 123-473°K and by Michels and Nederbragt [2889] in the interval from 273-423°K for pressures up to 136 atm. The papers by the Dutch investigators [2453, 3144, 4064] comprise the low-temperature interval from 121-293°K for pressures up to 20 atm.

TABLE 319

Accepted Values of the Force Constants of the Intermolecular Lennard-Jones Potential

Вещество 1	$\sigma/\text{Å}, \text{°K}$ 2	$b_0, \text{см}^3/\text{моль}$ 3	Вещество 4	$\sigma/\text{Å}, \text{°K}$ 5	$b_0, \text{см}^3/\text{моль}$ 6
O ₂	117,04	54,37	CF ₂ Cl ₂	296	179
O ₃	205	80,95	CFCI ₃	235	466
H ₂	37,00	31,67	C ₆ H ₆	199,2	116,7
F ₂	121	59,4	C ₆ H ₂	117	607
SF ₆	195	234,85	C ₆ N ₂	175,7	397,8
N ₂	95,36	64,13	SiF ₄	148,7	220,70
NO	131	40	BF ₃	178	108
N ₂ O	189	122	BCl ₃	346	118,36
CO	100,8	69,22	He	10,22	21,07
CO ₂	{ 187,5 ^a	{ 113,0 ^a	Ne	35,60	26,21
	{ 203,3 ^b	{ 75,20 ^b	Ar	119,49	49,92
CH ₄	148,2	70,16	Kr	166,67	62,92
CF ₄	152,1	131,31	Xe	225,3	84,96

1) Substance; 2) cm³/mole; 3) substance; 4) cm³/mole; 5) for; 6) for.

TABLE 320

Accepted Values of the Force Constants of the Intermolecular Stockmayer Potential

1 Вещество	$\sigma/\text{Å}, \text{°K}$ 2	$b_0, \text{см}^3/\text{моль}$ 3	ρ^*	3 Вещество	$\sigma/\text{Å}, \text{°K}$ 4	$b_0, \text{см}^3/\text{моль}$ 5	ρ^*
H ₂ O	380	23,42	1,2	CH ₂ Br	486,4	52,48	0,4
SO ₂	471	35,48	0,5	CH ₂ J	590,6	69,65	0,2
NH ₃	317	22,12	1,0	CHFCI ₃	381	141	0,1
CH ₃ F	207	47,85	1,07	CHCl ₃	1082	33,45	0,1
CH ₂ Cl	378	50,73	0,6				

1) Substance; 2) cm³/mole; 3) substance; 4) cm³/mole.

The virial coefficients of oxygen were repeatedly determined both from the p-V-T-data [1935, 2076, 2100, 2868, 2890, 3062] and from measurements of sonic speed [2346, 2552, 4059]. Meyers [2868] determined the values of the second, third and fourth virial coefficients in the interval from 123-473°K on the basis of experimental p-V-T-data. An analogous work was carried out by the US National Bureau of Standards [2076]. The data of these works for the second virial coefficient are in good agreement with each other and with the results of other authors.

On the basis of the experimental data on the second virial coefficient Holborn and Otto [2100], Newitt [3062], and Hall and Ibele [1934] determined the force constants of the intermolecular Lennard-Jones potential equal to $\epsilon/k = 118^\circ\text{K}$, $b_0 = 52.26 \text{ cm}^3/\text{mole}$, $\epsilon/k = 117.5^\circ\text{K}$, $b_0 = 57.75 \text{ cm}^3/\text{mole}$ and $\epsilon/k = 117.04^\circ\text{K}$, $b_0 = 54.37 \text{ cm}^3/\text{mole}$, respectively. The most accurate values of the force constants are the values suggested by Hall and Ibele [1935], which were obtained on the basis of the most reliable experimental values of B.

With the help of the experimental values of the virial coefficients for temperatures up to 500°K and the values calculated for higher temperatures, tables of the thermodynamic properties of molecular oxygen for temperatures up to 3000°K and for pressures up to 100 atm were compiled on the basis of the equation of state $pV = RT(1 + B'_p + C'_p + D'_p)$ in the US National Bureau of Standards [2076]. These data are in good agreement with the data given in Table 307 of this Handbook.

O_3 (gas). Birdsall, Jenkins and DiPaolo [823] determined the compressibility of ozone in the interval from 161-398°K for pressures up to 10 atm. On the basis of these data we calculated the force constants of the intermolecular Lennard-Jones potential. Birdsall, Jenkins and DiPaolo [823] tabulated the thermodynamic properties of ozone in the

interval from 200-350°K for pressures up to 10 atm.

H_2 (gas). The most comprehensive surveys of works on the investigation of the p-V-T properties of hydrogen published up to 1948 were given by Gratch [1842] and Wolley, Scott and Brickwedde [4329]. Among the works of this period we have to mention the works by Deming and Shupe [1313] and Michels and collaborators [2879]. Deming and Shupe gave data on the compressibility of hydrogen in the interval from 58-773°K for pressures up to 1200 atm. The most accurate data on the compressibility of hydrogen in the interval from 93-423°K for pressures up to 3000 atm were obtained by Michels and collaborators. Wilkinson [4270] studied the p-V-T properties in the interval from 20-3000°K for pressures from 1 to 200 atm. Johnston and collaborators [2271, 2286] determined the p-V-T data of hydrogen also in the interval from 20-300°K, but for pressures up to 1000 atm. Michels and Gerver [2878] calculated the values of the second virial coefficient from the data on hydrogen compressibility in the interval from 273-373°K. Deming and Shupe [1313] calculated the values of B also from compressibility data in the interval from 58-773°K and proposed an empirical formula for B. Schaefer [3604] determined the values of the second virial coefficient for ortho-, para- and normal hydrogen at low temperatures. Keyes [2384] reviewed the values of B obtained by various authors in order to introduce correctives into the temperature scale of hydrogen and proposed a formula which can be applied in the interval from 23-473°K.

There are several collections of the force constants of the intermolecular Lennard-Jones potential in literature. Corner [1182] carried out a detailed investigation and comparing the calculated and experimental values of B concluded that the values suggested by Michels and Goudekot [2879] are the most accurate values of the force constants. Bird, Spatz and Hirschfelder [822] proved that a quantum-mechanical

formula must be used to calculate the second virial coefficient of hydrogen with a quantum-mechanical parameter having the value $\lambda^* = 1.729$. On the basis of the compressibility data of H₂ Michels and collaborators [2876, 2882] tabulated the thermodynamic properties of hydrogen for the temperature interval from 98-423°K and pressures up to 3000 atm. Woolley, Scott and Brickwedde [4329] evaluated the p-V-T data of H₂, partly analytically, partly graphically, and tabulated the thermodynamic properties of hydrogen for the interval from 20-873°K and for pressures up to 3000 atm. With the help of the most reliable p-V-T data tables of the thermodynamic properties of hydrogen for temperatures up to 600°K and pressures up to 100 atm were compiled in the US National Bureau of Standards [2076].

D₂ (gas). The compressibility of deuterium was investigated by Michels and collaborators [2875, 2880] in the interval from 93-423°K at pressures up to 3000 atm. Schaefer [3603, 3604] experimentally determined the difference between the values of the second virial coefficients of hydrogen and deuterium in the interval from 23-46°K. Van Itterbeck and collaborators [4058, 4060, 4061] redetermined the values of B for deuterium and on the basis of these measurements gave an empirical formula for calculating the values of this quantity which is in good agreement with Schaefer's measurements in the temperature interval from 19-42°K. Michels and Goudekot [2880] also experimentally determined the difference in the values of B of the hydrogen isotopes.

On the basis of all these measurements of the second virial coefficient Bird, Spatz and Hirschfelder [822] found out that the force constants of the intermolecular Lennard-Jones potential for deuterium are the same as those for hydrogen, but the quantum-mechanical parameter λ^* is equal to 1.223. Michels and collaborators [2876, 2882] tabulated the thermodynamic properties of deuterium for the temperature in-

terval from 273-423°K and for pressures up to 3000 atm.

H_2O (gas). A great number of works on the experimental investigation of the thermodynamic properties of water and water vapor are known.

A considerable number of works was devoted to experimental investigations of the specific heat C_p of water vapor. Experimental investigations of C_p in the works by Smith, Keyes and Gerry [3787, 3788, 3789] and Timrot and collaborators [421] comprise a pressure interval up to 300 atm at temperatures up to 800°K. Vukalovich, Rasskazov and Sheyndlin determined C_p of water vapor in a pressure interval from 300 to 700 atm for temperatures up to 973°K in their works [139, 343, 461]. Sheyndlin, Shpil'rayn and Sychev [462] calculated the specific heat C_p of water vapor on the saturation line making use of several experimental works of other authors.

The specific heat at constant volume C_p of water and water vapor was investigated by Amirkhanov and Kerimov [71] in a rather wide temperature and pressure interval including also the critical domain.

Sirota and Timrot [377, 378] calculated the enthalpy of water vapor for pressures up to 150 atm and temperatures up to 873°K making use of their experimental measurements of the specific heat C_p of water vapor. Callendar and Egerton [1043] determined the entropy of water vapor for pressures up to 150 atm in a temperature interval from 573-823°K with an accuracy of 0.1%. An analogous work carried out by Havlicek and Miskovsky [1976] comprises a pressure interval up to 400 atm where the accuracy is 0.25%. In the work by Vukalovich, Zubarev and Prusanov [136] the results of previous works on the determination of water vapor enthalpy have been rendered more accurate.

The most comprehensive survey of works up to 1948 on the determination of the p-V-T data of water vapors was published by Keyes [2385].

In a paper by Timrot et al. [421] the p-V-T data were investigated in a temperature interval from 643-813°K at pressures from 110 to 300 atm. Kirillin and Romyantsev [226, 227] obtained experimental data for water vapor in a wide pressure interval at temperatures from 573 to 923°K. Kennedy [2370] investigated the p-V-T properties at temperatures up to 1273°K and pressures up to 98 atm.

Keenan and Keyes [2341] tabulated the thermodynamic properties of water vapor for temperatures from 311 to 733°K and pressures up to 360 atm on the basis of their experimental p-V-T data. Koch [2453] published tables of thermodynamic properties in metric units for the interval from 373-823°K and for pressures from 0.01 to 3000 atm. For low pressures Goff and Gratch [1785] compiled accurate tables of the thermodynamic properties of water vapors in the interval from 167-375°K.

In the US National Bureau of Standards [2076] tables of the thermodynamic properties of water vapor for temperatures up to 900°K and pressures up to 100 atm were compiled based on the experimental data of Keyes [2386], Kennedy [2368], and Kirillin and Romyantsev [226]. Analogous tables were published by Fano, Hubbel and Beckett [1527].

Having analyzed the experimental works of several authors Kirillin, Sheyndlin and Shpil'rayn [228] compiled tables of the basic values of enthalpies and specific volumes of water vapors in the temperature interval from 673-923°K for pressures up to 500 atm.

On the basis of the experimental data Sarukhanian [3584] published tables of the thermodynamic properties of water vapor for temperatures up to 1273°K and pressures up to 1000 atm.

With the help of the works by Sheyndlin [461] and Kirillin and Romyantsev [227] Rivkin and Sirota [404] compiled tables of the thermodynamic properties of water vapor which, together with tables compiled earlier [403], comprise a pressure interval up to 400 atm for tempera-

tures up to 1023°K. Analogous tables compiled by Vukalovich [135] comprise a pressure interval up to 1000 atm for temperatures up to 1273°K. In all these works the Centigrade temperature scale is used. Where the intervals overlap, the tables of Rivkin and Sirota [404] and Vukalovich [135] are almost identical and quite reliable.

Keenan and Keyes [2341] have experimentally determined the values of the second virial coefficient in the interval from 311-733°K and proposed an empirical equation which reproduces well the experimental values of B in the temperature interval indicated:

$$B = 18,016 \left[1,89 - \frac{2641,6}{T} e^{-\frac{1,858 \cdot 10^6}{T^2}} \right].$$

With the help of the experimental values of B the force constants of the intermolecular Stockmayer potential were calculated [1878].

F_2 (gas). White, Jih-Heng and Johnston [4237] determined the second virial coefficient of diatomic fluorine in the interval from 80-300°K and on the basis of their experimental data calculated the force constants of the intermolecular Lennard-Jones potential.

Cl_2 (gas). Pellaton [3214] investigated the p-V-T properties of chlorine vapors in the temperature interval from 273-417°K. Ross and Maass [3488] measured the density of Cl_2 in the interval from 288-348°K at pressures up to 2 atm and proposed an equation of state for the temperature interval indicated. Martin and Hou [2789] derived an equation of state of the chlorine vapors for a wide temperature interval, on the basis of experimental p-V-T data.

Measurements of the second virial coefficient carried out by Pier [3242], Jaquerod and Tourpain [4387], and Eucken and Hoffmann [1501] comprise an interval from 244-457°K.*

On the basis of empirical equations of state Arnold and Kobe [569], Hulme [2152] and Ziegler [192] tabulated the thermodynamic prop-

erties of chlorine in the interval from 231-477°] for pressures from 0.7 to 27 atm. Kapoor and Martin [2324] compiled tables of thermodynamic properties in the interval from 211-555°K for pressures up to 95 atm, making use of an equation of state of chlorine that had been proposed by Martin and Hou [2789].

J_2 (gas). Perlman and Rollefson [3223] studied the p-V-T properties of iodine in the interval from 723-1274°K.

HF (gas). As far back as 1888, Thorpe and Humbly [3987] measured the temperature dependence of the density of gaseous HF at a pressure of 745 mm Hg, by means of the Dumas method (in platinum vessels), but also the pressure dependence of density at several temperatures. The authors proved the presence of HF association whose degree is strongly dependent on the concrete conditions. The phenomenon of hydrogen fluoride being strongly associated was verified in several later works [940, 941, 942, 943, 944, 945, 946, 1600, 2217, 2644, 3737]. Briegleb and Strohmeier [946] accurately measured the isothermals of the density of gaseous HF in the interval from 299-329°K at medium and low temperatures. Franck and Spalthoff [1593] measured the specific heat of hydrogen fluoride vapors and its density at temperatures up to 573°K and pressures up to 300 atm.

HCl (gas). Schlatter [3624] measured the compressibility of HCl at 273°K for pressures higher than 1 atm. Curtiss and Hirschfelder [1237] calculated the compressibility of HCl for a wide temperature interval from the heat of vaporization and the pressure of the vapors. On the basis of experimental data, Arnold and Kobe [569] tabulated the thermodynamic properties of hydrogen chloride.

HBr (gas). Guye [1906] measured the compressibility of HBr at 273°K for pressures lower than 1 atm. Curtiss and Hirschfelder [1237] calculated the compressibility of HBr from the heat of vaporization and

the pressure of the vapors for a wide temperature interval.

HJ (gas). Curtiss and Hirschfelder [1237] calculated the compressibility of HJ from the heat of vaporization and the pressure of the vapors.

S, S₂, S₄, S₆, S₈ (gas). Evens and Wagman [1516] tabulated the thermodynamic properties of sulfur in the interval from 273-1773°K on the basis of experimental data. The tables of the thermodynamic properties of sulfur compiled by West [4217] comprise a temperature interval from 393-919°K for pressures up to 10 atm.

SO₂ (gas). Cawood and Patterson [1076] measured the compressibility of sulfur dioxide in the interval from 303-323°K at low pressure. Riedel [3440] obtained the p-V-T data of SO₂ in the interval from 278-323°K. Cooper, B. Maass and O. Maass [1170] experimentally determined the values of the second virial coefficient of sulfur dioxide in the interval from 273-345°K.

The values of B obtained by Riedel [3440] comprise an interval from 278-318°K. On the basis of these experimental values Baybuz [80] determined the force constants of the intermolecular Stockmayer potential.

Making use of experimental p-V-T data Rynning and Hurd [3569] and West and Giusti [4218] tabulated the thermodynamic properties of SO₂ in the interval from 200-1366°K for pressures up to 68 atm.

H₂S (gas). The compressibility of hydrogen sulfide at 273°K was measured by Maverick [2815] at a pressure of about 1 atm. Reamer, Sage and Lacey [3407] investigated the p-V-T properties of H₂S in the interval from 278-444°K at a pressure up to 68 atm. Curtiss and Hirschfelder [1237] calculated the deviation of the properties of hydrogen sulfide from the properties of a perfect gas from the heat of vaporization and the pressure of the vapors. Pitzer and collaborators [3262] calculated

the compressibility of H_2S in the interval from 298-1494°K at pressures up to 800 atm. On the basis of the experimental p-V-T data West [4216] tabulated the thermodynamic properties of hydrogen sulfide.

SF_6 (gas). McCormack and Schneider [2692] measured the compressibility of sulfur hexafluoride in the interval from 273-523°K for pressures up to 50 atm and calculated the values of the second virial coefficient with an accuracy of 1.5%. Hamann, McManamey and Pearse [1939] measured the values of the second virial coefficient of SF_6 in the interval from 293-448°K with an accuracy of ± 4 cm³/mole. McCormack and Schneider [2692] calculated the force constants of the intermolecular Lennard-Jones potential on the basis of the experimental values of B: $\epsilon/k = 188.7^\circ K$, $b_0 = 211.6$ cm³/mole. An analogous work was carried out by Hamann, McManamey and Pearse [1939] who obtained $\epsilon/k = 200.9^\circ K$ and $b_0 = 260$ cm³/mole. Evidently, the most accurate values of the force constants were obtained by Hamann and Lambert [1937].

N_2 (gas). A great number of works on the investigation of p-V-T data of nitrogen is known. A survey and a graphic evaluation of the earliest works were performed by Deming and Shupe [1311], but the most comprehensive survey of works carried out up to 1948 was given by Gratch [1842]. In a later work by Bloomer and Rao [845] the p-V-T properties of nitrogen were studied for temperatures up to 530°K.

Making use of ultrasonic speed measurements Noury and collaborators [3105] calculated the compressibility of nitrogen from the critical temperature up to 523°K for pressures up to 1200 atm.

In 1949, Claitor and Crawford [1120] determined the values of the second and third virial coefficients by evaluating low-temperature p-V-T data. Michels, Lunbeck and Wolkers [2884] calculated the values of the second virial coefficient with the help of data on the compressibility of nitrogen in the interval from 273-423°K. Keyes [2384] pro-

posed an empirical formula for calculating the values of the second virial coefficient in the interval from 123-1273°K. The experimental values of the second virial coefficient obtained by Holborn and Otto [2103] are well described by Keyes' formula. Hall and Ibele [1934, 1935] evaluated the p-V-T data for high temperatures and calculated the values of the second and third virial coefficients.

Amdur and Mason [542] calculated the values of the second virial coefficient of N₂ for the interval from 1000-15,000°K. On the basis of the experimental data on the second virial coefficient Holborn and Otto [2103] calculated the force constants of the intermolecular Lennard-Jones potential: $\epsilon/k = 95.9^\circ\text{K}$ and $b_0 = 64.42 \text{ cm}^3/\text{mole}$. Corner [1181] found $\epsilon/k = 95.3^\circ\text{K}$ and $b_0 = 61.49 \text{ cm}^3/\text{mole}$, relying both on experimental values of the second virial coefficient and on measurements of the Joule-Thomson effect. Using his own measurements of the second virial coefficient Michels [2897] obtained $\epsilon/k = 95.05^\circ\text{K}$ and $b_0 = 63.78 \text{ cm}^3/\text{mole}$. The most reliable values of the force constants calculated by Hall and Ibele are equal to $\epsilon/k = 95.36^\circ\text{K}$ and $b_0 = 64.13 \text{ cm}^3/\text{mole}$.

Using data taken from literature Rybkin [352] tabulated the thermodynamic properties of N₂ for pressures up to 25 atm. Analogous tables for temperatures up to 3000°K and pressures up to 100 atm were compiled in the US National Bureau of Standards [2076] on the basis of the equation of state $pV = (RT(1 + B'_p + C'_p2 + D'_p3))$. The accuracy of the calculated values of enthalpy and entropy lies within 10% of the values of the corrections to nonideality.

Saurel and Lecocq [3595] investigated the pressure dependence of the functions $E - TS$ and $H - TS$ and of the entropy of nitrogen for temperatures from 573 to 1073°K at pressures up to 900 atm. On the basis of data given by the US National Bureau of Standards [2076] the same authors calculated the values of the function $H - TS$ and of volatility

of nitrogen for the same temperature and pressure interval. The calculated values of the function $H - TS$ are in good agreement with the experimental Data.

Woolley [4327] tabulated the thermodynamic properties of nitrogen in the interval from 100-3000°K at pressures up to 100 atm on the basis of the experimental p-V-T data.

NO (gas). Batuecas [680] measured the compressibility of nitrogen oxide for pressures from 230 to 840 mm Hg. Eucken and D'Or [1500] studied the p-V-T properties in the interval from 125-273°K for low pressures. Briner, Biedermann and Rother [948, 949] investigated the compressibility of NO in the interval from 193-283°K for pressures up to 160 atm. Johnston and Weimer [2285] investigated the p-V-T properties for low pressure in the interval from 122-308°K. More accurate p-V-T data were obtained by Golding and Sage [1794] in the interval from 278-378°K for pressures up to 170 atm. The accuracy of these data by Golding and Sage is 0.3%. Eucken and D'Or [1500] determined the values of the second virial coefficient in the interval from 125-273°K. Johnston and Weimer [2285] calculated the values of B for nitrogen oxide in the temperature interval from 122-308°K on the basis of the p-V-T data at low pressures with an accuracy of ± 2 cm³/mole. For the calculation of the values of B in the interval from 122-308°K the authors proposed a formula by Eucken and D'Or in a form that had been rendered more accurate by them:

$$B = 20 + \frac{5881.5}{T} - \frac{5.7639 \cdot 10^6}{T^2} + \frac{8.4301 \cdot 10^{10}}{T^4} - \frac{9.2783 \cdot 10^{14}}{T^6} \text{ cm}^3/\text{mole}.$$

On the basis of the experimental values of the second virial coefficient, Hirschfelder, McClure, Curtiss and Osborne [2083] calculated the force constants of the intermolecular Lennard-Jones potential:

$\epsilon/k = 131^\circ\text{K}$ and $b_0 = 40 \text{ cm}^3/\text{mole}$. Using p-V-T data published earlier

Opfell, Schlinger and Sage [3145] tabulated the thermodynamic properties of nitrogen oxide in the interval from 210-377°K for pressures from 0.7 to 204 atm.

N_2O (gas). The compressibility of nitrogen monoxide at pressures from 1-3 mm Hg in the temperature interval from 273-310°K was determined by Cawood and Patterson [1076]. Johnston and Weimer [2285] investigated the p-V-T properties at low pressures in the interval from 197-298°K. In the works by Batuecas [680, 681, 682, 684], Sutton, Ambler and Williams [3908] and Bottomley [871] the compressibility of N_2O was measured at different pressures, but only at several fixed temperatures. Making use of ultrasonic speed measurements Noury and collaborators [3105] calculated the compressibility of N_2O in the interval from 310-523°K for pressures up to 1200 atm. On the basis of p-V-T data Johnston and Weimer [2285] calculated the values of the second virial coefficient in the interval from 197-298°K with an accuracy of ± 2 $cm^3/mole$. For the calculation of B in the mentioned temperature interval the authors proposed an equation that reproduces well the experimental data:

$$B = 32 - \frac{56115}{T} + 3,9424 \cdot \frac{10^6}{T^2} - 3,9145 \cdot \frac{10^{11}}{T^3} + 3,0747 \cdot \frac{10^{16}}{T^4}.$$

Hirschfelder, McClure, Curtiss and Osborne [2083] calculated the force constants of the intermolecular Lennard-Jones potential: $a/k = 189^\circ K$ and $b_0 = 122 \text{ cm}^3/mole$.

NH_3 (gas). The first accurate compressibility measurements were carried out by Holst [2109] in the interval from 307-373°K for pressures from 3 to 6 atm. In the work by Bridgman [936] the compressibility of ammonia was investigated for pressures up to 15,000 atm in the interval from 303-368°K, with an accuracy up to some percents. Initially Keyes [2381] obtained the p-V-T data essentially from the equation

of state $p = \frac{RT}{V-a} - \frac{A}{(V-b)^2}$; the data were obtained in the temperature interval from 330 to 440°K at pressures from 20 to 40 atm. The accuracy of these data lies within 1% by volume since ammonia was adsorbed by the walls of the piezometer. In a later work by Keyes [2382] the compressibility was investigated in the interval from 273-483°K for pressures from 100 to 1100 atm with an accuracy of 0.25%. Meyers and Jessup [2869] investigated the p-V-T data of ammonia in the interval from 238-578°K for pressures from 0.9 to 28 atm. These data were obtained with the high accuracy of the order of 0.1%, whereas the accuracy for high temperatures is 0.04%. The data of Beattie and Lawrence [706] comprise the interval from 323-600°K at pressures from 14 to 120 atm and, by means of the Beattie-Bridgman equation, can be connected with the data of Meyers and Jessup. Kazarnovskiy [201] obtained p-V-T data for the interval from 473-573°K at pressures from 100 to 1600 atm with a maximum error of 0.5%. Kazarnovskiy's data correspond to Keyes' data within 0.7% at 473°K. Tsiklis [453] investigated the compressibility at pressures from 1000 to 10,000 atm in the interval from 323-423°K and suggested an empirical equation of state for these temperature and pressure intervals. The maximum difference between the experimental data and the results of calculation by means of the equation of state is 0.5%. Kazarnovskiy and Karapet'yants [202] compiled diagrams and tables of the thermodynamic properties of NH₃ in the interval from 423-573°K for pressures from 20 to 2000 atm with an accuracy of 3%, making use of the p-V-T data obtained by Keyes, Beattie and Lawrence and Kazarnovskiy. Besides the aforementioned equations obtained by Keyes, Beattie-Bridgman and Tsiklis, a great number of other equations of state were suggested. All these equations of state are based on experimental data and are correct in one or another narrow range of temperatures and pressures. Meyers and Jessup [2869], and then Beattie and Lawrence

[706] determined the values of the second virial coefficient B experimentally. Keyes [2383] suggested an empirical equation for B

$$B = 17,031 \left[24 - \frac{2316}{T} \right] e^{-\frac{774 \cdot 10^6}{T^2}} \text{ cm}^3/\text{mole},$$

which reproduces well the experimental data in the interval from 238-598°K. Hirschfelder, McClure and Weeks [2084] also determined the values of the second virial coefficient on the basis of the p-V-T data obtained by Meyers and Jessup

$$B = 6,28 \left[1 - 0,509 e^{-\frac{88}{T^2}} \right] \text{ cm}^3/\text{mole}.$$

Lambert et al. [2550, 2551] obtained an equation for the second virial coefficient on the basis of the experimental data

$$B = 20,8 - \frac{2,05}{T^2} \cdot 10^7 \text{ cm}^3/\text{mole}.$$

The authors showed that the values of B obtained from this formula and the values obtained from the formulas of Keyes and Hirschfelder, McClure and Weeks are in a satisfactory agreement. On the basis of the experimental values of B Meyers [3016] also suggested an empirical formula for the second virial coefficient. Using the experimental values of the second virial coefficient, Rowlinson [3543], and then Guggenheim [1878] calculated the force constants of the intermolecular Stockmayer potential $t^* = 1,0$, $e/k = 317^\circ\text{K}$ and $b_0 = 22.12 \text{ cm}^3/\text{mole}$. Fink [1560] proposed an empirical equation for the enthalpy describing well the existing experimental data, with an accuracy of 0.1%. The same author [1559] proposed enthalpy and entropy equations for the metastable state. Grahl [1839] compiled a table of the values of enthalpy for NH_3 in the interval from 200-644°K for pressures up to 1088 atm. Din [1350] published tables of thermodynamic functions in the interval from 300-580°K for pressures from 1 to 1100 atm on the basis of the most reliable experimental data.

NF_3 (gas). The compressibility of NF_3 at room temperature was measured by Ramaswamy [3354].

CO (gas). The most comprehensive survey of works on the investigation of the p-V-T data of carbon monoxide published up to 1948 is given in the work by Gratch [1842]. Dilley [1349] obtained data in the interval from 200-300°K for pressures up to 1000 atm. The work by Deming and Shupe [1312] comprises the widest temperature interval from 200-473°K for pressures up to 120 atm. The data obtained by Bartlett et al. [669] comprise the interval from 200-473°K for pressures up to 1000 atm and are in good agreement with the data obtained by other authors. The most reliable p-V-T data for carbon monoxide were obtained in a work by Michels, Lupton, Wassenaar and DeGraaff [2885] in 1952, for an interval from 273-423°K and for pressures up to 3000 atm. Using these data for the mentioned temperature interval the authors determined the values of the second virial coefficient. Van Itterbeck and Van Doninck [4058] calculated the values of B for low temperatures from sonic speed measurements. Maron and Turnbull [2779] calculated the values of the second, third, fourth and fifth virial coefficients, making use of the virial coefficients of nitrogen and the critical constants of CO for 200-473°K.

Corner [2779] determined the force constants of the intermolecular Lennard-Jones potential: $\epsilon/k=100.2\text{ K}$ and $b_0 = 67.22\text{ cm}^3/\text{mole}$, by means of an investigation of the Joule-Thomson effect. On the basis of more accurate p-V-T data, the values $\epsilon/k = 100.8^\circ\text{K}$ and $b_0 = 69.22\text{ cm}^3/\text{mole}$ which are the most reliable were obtained in the US National Bureau of Standards [2076]. The values of the force constants $\epsilon/k = 126.3^\circ\text{K}$ and $b_0 = 64.68\text{ cm}^3/\text{mole}$ obtained in the work by Bahadur and Madam [607a] on the basis of the well-known values of the third virial coefficient of carbon monoxide cannot be considered reliable enough since the values

of the third virial coefficient of CO themselves were obtained with low accuracy. Deming and Shupe [1312] graphically evaluated the p-V-T data obtained by Scott [3668], Goig [1788, 1789] and Bartlett et al. [669] and tabulated the thermodynamic functions for temperatures up to 673°K and pressures up to 100 atm. Using his own p-V-T data Michels [2872, 2873] determined the thermodynamic functions of carbon monoxide for temperatures from 273-423°K and pressures up to 3000 atm. Making use of the equation of state $pV = RT(1 + B'p + C'p^2)$, in the US National Bureau of Standards [2076] tables of the thermodynamic properties of CO for temperatures up to 3000°K and pressures up to 100 atm were calculated; the accuracy of the nonideality correction for temperatures higher than 400°K and pressures up to 10 atm is 10%, and 20% for lower temperatures and pressures higher than 10 atm. On the basis of the experimental p-V-T data Din [1350] tabulated the thermodynamic properties for the interval from 273-673°K and for pressures from 0.1 to 1200 atm.

CO₂ (gas). Carbon dioxide is one of the first gases for which accurate compressibility measurements were carried out. For the first time, the p-V-T data of CO₂ were measured by Amagat [532, 533] in the interval from 273-523°K for pressures up to 100 atm. Keesom [2345] and Lowry and Erickson [2668] investigated the compressibility of CO₂ in the critical domain.

The compressibility of carbon dioxide at temperatures lower than 273°K was determined only in the work by Jenkin [2223]. Very accurate compressibility measurements at a pressure below 1 atm were carried out by Batuecas [683], Guye and Batuecas [1907] and Kennedy [2369]. Maass and Menni [2683], Cooper and Maass [1171, 1172], Cawood and Patterson [1076] and Schaefer [3604] determined the compressibility of CO₂ for pressures below 20 atm. For pressures up to 36 atm and temperatures from 273-373°K the compressibility and volatility of CO₂ were deter-

mined by Houghton, McLeon and Pitchie [2131]. The work by McCormack and Schneider [2690] comprises the widest temperature interval from 273-873°K for pressures up to 50 atm. Dinglinger [1351] determined the specific volume of CO₂ and, thus, measured the compressibility in the interval from 273-403°K at pressures up to 230 atm. Kendall and Sage [2367] determined the compressibility of carbon dioxide in the interval from 277-501°K at pressures up to 680 atm. In the works by Michels and collaborators [2874, 2886, 2887, 2888] the most accurate p-V-T data for the temperature interval from 273-423°K and a pressure interval from 16 to 3000 atm are obtained. Even for higher pressures the error of experiment does not exceed 0.05%. For pressures below 1000 atm the measurements were undertaken with an accuracy of 0.02%.

Several equations of state were proposed for carbon dioxide. The equations of state suggested by Onnes and Crommelin [3142], Plank and Kuprianoff [3264] and Beattie-Bridgman [703] prove to be the most accurate. All these equations are satisfactory for the whole domain in which gaseous carbon dioxide exists. Probably, the Beattie-Bridgman equation is the most accurate in the interval from 273-373°K for pressures up to 100 atm.

Michels [2887] experimentally determined the values of the second virial coefficient in the interval from 273-423°K and, in detail, considered the problem whether the Kamerlingh-Onnes equation could be applied to CO₂ in the aforementioned temperature interval. He established that this equation reproduces the p-V-T data for pressures above 500 atm better than the other equations of state. Margenau [2767] critically compared the theoretical and experimental values of the second virial coefficient. Zaalishvili [192] calculated the values of the second virial coefficient of CO₂ in the interval from 310-510°K, by means of various data on compressibility and making use of the Beattie-Bridgman

equation. McCormack and Schneider [2961] calculated the values of the second virial coefficient in the interval from 273-873°K, with an accuracy of 1%. The data obtained by McCormack and Schneider are in good agreement with Michels' data within the mentioned experimental accuracy. The values of the second virial coefficient for temperatures from 213-243°K obtained by Cook [1164] agree with the data found by the other research workers.

On the basis of the experimental data on the second virial coefficient Michels [2886] determined the force constants of the intermolecular Lennard-Jones potential: $\epsilon/k = 189^\circ\text{K}$, $b_0 = 113.9 \text{ cm}^3/\text{mole}$. Making use of experimental data for a wider temperature interval, Corner [1182] obtained the values $\epsilon/k = 198^\circ\text{K}$ and $b_0 = 110 \text{ cm}^3/\text{mole}$. McCormack and Schneider [2693] found that for CO_2 there are two sets of values for the force constants of the intermolecular Lennard-Jones potential: $\epsilon/k = 203.3^\circ\text{K}$ and $b_0 = 113.0 \text{ cm}^3/\text{mole}$ for the interval from 273-473°K, and $\epsilon/k = 187.5^\circ\text{K}$ and $b_0 = 75.20 \text{ cm}^3/\text{mole}$ for the interval from 573-1473°K. These values of the force constants are the most reliable.

Making use of the p-V-T data published in literature Rybkin [352] compiled tables of the thermodynamic properties of the real gas at pressures up to 25 atm. Using the data obtained by Kennedy [2369] Price [3320] calculated the values of the thermodynamic properties for temperatures up to 1000°K and pressures up to 1400 bar. In the US National Bureau of Standards [2076] tables of thermodynamic properties for temperatures up to 1500°K at pressures up to 100 atm were compiled, on the basis of the most reliable p-V-T data. An analogous work was carried out by Din [1350] in the interval from 193-423°K for pressures from 0.5 to 3000 atm. Chen [1094] calculated the thermodynamic properties of gaseous CO_2 for a wide pressure and temperature interval on the basis of the Beattie-Bridgman equation.

CS_2 (gas). Casado, Masie and Whytlaw-Gray [1068] obtained the value of $-658 \text{ cm}^3/\text{mole}$ for the second virial coefficient of carbon bisulfide at 295.15°K . Bottomley and Remington [874] determined the values of B for carbon bisulfide equal to -646 and $-582 \text{ cm}^3/\text{mole}$ for temperatures of 295.15 and 308.15°K , respectively. It is, however, evident that the measurements carried out by Bottomley and Reeves [873] who determined the values of the second virial coefficient at three temperatures: -849 at 295.15°K , -748 at 308.15°K and $-661 \text{ cm}^3/\text{mole}$ at 323.15°K , are more reliable. When determining the virial coefficients Bottomley and Reeves applied the method of differential compressibility which is considerably less sensitive to small impurities of the substance under investigation compared to the method of determining B by measuring the density as it was applied by Casado, Masie and Whytlaw-Gray, and Bottomley and Remington. The latter authors attest to the possibility that CS_2 could be contaminated by small quantities of CO , a phenomenon that could give rise to an inaccuracy in the values of B obtained for hydrogen sulfide.

O'Brien and Alford [3107] tabulated the thermodynamic properties of carbon bisulfide in the interval from 273 - 1088°K for pressures up to 69 atm by making use of the Beattie-Bridgman equation of state.

CH_4 (gas). The most comprehensive survey of works on the investigation of p-V-T data of methane up to 1948 was published by Keyes [2385]. In their works, Keyes and collaborators [2387, 2388] determined the p-V-T data in the interval from 273 - 473°K at pressures up to 300 atm , and an equation of state of methane was proposed for the mentioned temperature and pressure interval. Kvalnes and Gaddy [2506] measured the compressibility of methane for pressures up to 1000 atm in the interval from 203 - 473°K . Dinglinger [1351] determined the p-V-T data of CH_4 in the interval from 223 - 400°K for pressures up to 230 atm .

Thomaes and Steenwinkel [3963a] measured the second virial coefficient of CH_4 in the temperature interval from 108-250°K by the method of measuring the difference in gas expansion compared to hydrogen. The results are in good agreement with the earlier measurements carried out by Michels and Nederbragt [2889].

Schamp, Mason, Richardson and Altman [3606] measured the compressibility of methane at temperatures from 273-423°K and pressures from 20 to 230 atm with an accuracy of 0.01%, making use of the method of constant volumes. The authors of the work [3606] explain the small deviation from Michels' [2897] data by the presence of small quantities of ethane in the methane applied by Michels. From the data on the compressibility of CH_4 Schamp, Mason, Richardson and Altman determined the values of the virial coefficients in the mentioned temperature interval. Beattie and Stockmayer [707] calculated the values of the second virial coefficient in the interval from 423-573°K by means of the Beattie-Bridgman equation. Zaalishvili [191] also calculated the values of B in the interval from 203-477°K by using experimental p-V-T data of various research workers that are in good agreement.

On the basis of the experimental p-V-T data Michels and Nederbragt [2889] determined the force constants of the intermolecular Lennard-Jones potential: $\epsilon/k = 148.2^\circ\text{K}$ and $b = 70.16 \text{ cm}^3/\text{mole}$. Schamp, Mason, Richardson and Altman [3606] calculated force constants that are in good agreement with data obtained by Michels and Nederbragt, from the values of the second virial coefficient. These values of the force constants are more reliable than the values $\epsilon/k = 148^\circ\text{K}$ and $b_0 = 67.06 \text{ cm}^3/\text{mole}$ obtained by Corner [1180] on the basis of crystallographic data. Spencer and Flannagan [3820] proposed an empirical equation for the specific heat of methane on the basis of the experimental data. Sledjeski [3765] drew diagrams showing the specific heat of methane for the

interval from 297-1019°K and for pressures from 34 to 680 atm, making use of the data in literature and of the equation of state obtained by Benedict. Masie [2798] published a survey of works carried out on the determination of the specific heat of methane.

On the basis of the experimental p-V-T data, Keyes, Taylor and Smith [2389] tabulated the thermodynamic properties of methane. Corcoran, Bowles, Sage and Lacey [1175] tabulated the thermodynamic properties of methane at low temperatures in the interval from 128-294°K for pressures up to 95 atm.

CH_3F (gas). Cawood and Patterson [1075] measured the compressibility of CH_3F at low pressures. Michels and Visser [2891] determined the p-V-T data of methyl fluoride in the interval from 273-423°K for pressures up to 150 atm. Using data on compressibility Cawood and Patterson determined the second virial coefficient of methyl fluoride and proposed an empirical equation for B

$$\ln(-B) = 7,596 - 7,6 \cdot 10^{-3} T.$$

Hamann and Pearse [1940] calculated the values of B of methyl fluoride after having measured the compressibility at low pressures of the order of 0.5 atm in the interval from 293-353°K. On the basis of the experimental p-V-T data Michels and Visser [2891] determined the values of the second virial coefficient in the interval from 273-423°K and calculated the force constants of the intermolecular Stockmayer potential. The same authors tabulated the thermodynamic properties of methyl fluoride in the interval from 273-423°K for pressures up to 150 atm.

CH_3Cl (gas). Bodareu [846] measured the compressibility of methyl chloride for pressures up to 4 atm. Ramaswamy [3355] determined the compressibility of CH_3Cl in the interval from 298-368°K at pressures below 1 atm. Hirschfelder, McClure and Weeks [2084] calculated the values of the second virial coefficient from experimental p-V-T data in

the interval from 238-450°K and proposed an empirical equation for B

$$\ln(-B) = 8,363 - 7,61 \cdot 10^{-3} T.$$

Hamann and Pearse [1940] determined the values of B in the interval from 293-353°K. Rowlinson [3542] calculated the force constants of the intermolecular Stockmayer potential, on the basis of the experimental values of B.

On the basis of the experimental data Tanner, Benning and Mathewson [3943] and Kobe and Long [2450] tabulated the thermodynamic properties of methyl chloride.

CH₃Br (gas). Ramaswamy [3355] measured the compressibility of CH₃Br in the interval from 298-368°K for a pressure below 1 atm. Curtiss and Hirschfelder [1237] calculated the compressibility of methyl bromide by making use of the heat of vaporization and the pressure of the vapors.

Hamann and Pearse [1940] determined the values of the second virial coefficient in the interval from 293-323°K by means of data on compressibility. Fogg, Hanks and Lambert [1571] measured the values of B in the interval from 273-373°K. The experimental values of B are well described by the equation

$$\ln(-B) = 8,148 - 6,49 \cdot 10^{-3} T.$$

On the basis of the experimental values of B Baybuz [80a] calculated the force constants of the intermolecular Stockmayer potential.

CH₃J (gas). The compressibility of CH₃J in the interval from 298-368°K at a pressure below 1 atm was measured by Ramaswamy [3355]. Fogg, Hanks and Lambert [1571] determined the second virial coefficient of CH₃J in the interval from 273-373°K and proposed an empirical equation for B

$$\ln(-B) = 8,366 - 6,08 \cdot 10^{-3} T.$$

On the basis of these experimental values Baybuz [80a] calculated the

force constants of the intermolecular Stockmayer potential.

CH_2Cl_2 (gas). Sugawara and Nozawa [3896] investigated the thermodynamic properties of methylene chloride in the interval from 248-328°K at pressures from 0.04 to 2 atm. Masia and Pena [2799] determined the values of the second virial coefficient in the interval from 323-423°K. Dzung [1439] and Kobe and Long [2450] tabulated the thermodynamic properties of CH_2Cl_2 on the basis of the experimental data.

CHF (gas). Ramaswamy [3355] determined the compressibility of CHF_3 in the interval from 298-368°K for pressures up to 1 atm. Lambert and collaborators [2550] determined the p-V-T data in the interval from 313-402°K at pressures up to 1 atm. Fogg, Hanks and Lambert [1571] determined the values of the second virial coefficient of CHF_3 . The experimental values of B in the interval from 273-373°K are well reproduced by the equation

$$\ln(-B) = 14,636 - 2,93 \cdot 10^{-3} T.$$

CHF_2Cl (gas). Benning and McHarness [744] published a survey of works undertaken to determine the p-V-T data of CHF_2Cl . The same authors proposed an equation of state and compiled tables of the thermodynamic properties.

CHFCl_2 (gas). In 1939, surveys [744, 3961] of works on the determination of the p-V-T data of CHFCl_2 were published. In the same surveys the thermodynamic properties of CHFCl_2 are given.

Hirschfelder, McClure and Weeks [2084] determined the values of the second virial coefficient in the interval from 238-450°K on the basis of data on compressibility. Making use of these experimental values of B Rowlinson [3542] calculated the force constants of the intermolecular Stockmayer potential.

CHCl_3 (gas). The compressibility of CHCl_3 in the interval from 298-368°K at pressures below 1 atm was investigated by Ramaswamy

[3355]. Lambert, Roberts, Rowlinson and Wilkinson [2550] calculated the values of the second virial coefficient of CHCl_3 in the interval from 323-383°K, making use of their own p-V-T measurements. On the basis of these values of B the authors calculated the force constants of the intermolecular Stockmayer potential. Francis and McGlashan [1591] measured the values of B in the interval from 316-398°K. The values of B obtained by Francis and McGlashan are in good agreement with the data of the preceding authors and can be calculated from the equation

$$B = 83 - 1.12 \frac{10^6}{T} \text{ cm}^3/\text{mole}.$$

On the basis of the experimental data Kobe and Long [2450] calculated the thermodynamic properties of CHCl_3 .

CF_4 (gas). McCormack and Schneider [2692, 2693] measured the compressibility of carbon tetrafluoride in the interval from 273-673°K for pressures up to 50 atm and determined the values of B in this temperature interval. Douslin, Harrison, Moore and McCullough [1390a] also measured the compressibility of CH_4 in the interval from 273-623°K for pressures up to 394 atm. Making use of the results of the compressibility measurements in this work [1390a] the values of the second, third and fourth virial coefficients were determined. The values of the second virial coefficient obtained in the works [2692, 2693] and [1390a] are in good agreement with each other. McCormack and Schneider [2693] determined the values of the force constants of the intermolecular Lennard-Jones potential: $\epsilon/k = 153^\circ\text{K}$ and $b_0 = 130.97 \text{ cm}^3/\text{mole}$, which practically coincide with the values of the force constants recommended in the work [1390a] and accepted in this Handbook.

CF_3Cl (gas). Benning and McHarness [745] determined the p-V-T data of CF_3Cl . The authors proposed an equation of state and calculated the thermodynamic properties of CF_3Cl .

CF_2Cl_2 (gas). Buffington and Gilkey [1009] determined the p-V-T data and proposed an equation of state for CF_2Cl_2 . On the basis of the obtained p-V-T data Benning and McHarness [745] calculated the thermodynamic properties of CF_2Cl_2 . Hirschfelder, McClure and Weeks [2084] calculated the values of the second virial coefficient in the interval from 238-311°K by making use of the most reliable p-V-T data. The values of B obtained by Masie [2796] agree with the data of the preceding authors. On the basis of the equation of state $pV = RT + B'p + C'p^2$ Masie [2796] tabulated the thermodynamic properties of CF_2Cl_2 in the interval from 200-1500°K.

CFCl_3 (gas). Using their own p-V-T data Benning and McHarness [744, 745, 746, 747] determined the thermodynamic properties of CFCl_3 . Pavlova [329] carried out an experimental investigation of the basic thermodynamic properties of CFCl_3 .

Making use of the most accurate p-V-T data [3962] Hirschfelder, McClure and Weeks [2084] determined the values of B in the interval from 238-450°K. On the basis of these values of B Baybuz [80a] calculated the force constants of the intermolecular Lennard-Jones potential.

CCl_4 (gas). Ramaswamy [3355] determined the compressibility of carbon tetrachloride in the interval from 298-368°K for pressures up to 1 atm. Casado, Masie and Whytlaw-Gray [1068] determined the values of the second virial coefficient of CCl_4 at 295.15°K equal to $-1107 \text{ cm}^3/\text{mole}$. Applying a more perfect equipment, Bottomley and Remington [874] obtained $B = -1283 \text{ cm}^3/\text{mole}$ for the same temperature. Lambert and collaborators [2550] measured the compressibility of CCl_4 in the interval from 313-403°K for pressures up to 1 atm. These authors calculated the values of the second virial coefficient for the interval from 313-353°K from Berthelot's equation. Francis and McGlashan [1591] deter-

mined the second virial coefficient of CCl_4 in the interval from 316-343°K. The experimental values of B obtained by Francis and McGlashan in the mentioned temperature interval may be calculated from the equation

$$B = 575 - \frac{2 \cdot 10^6}{T^2} \text{ cm}^3/\text{mole}$$

and are in good agreement with the results of the work by Lambert and collaborators. These results, however, differ considerably from the data obtained by Eucken and Meyer [1502], especially in the upper part of the investigated temperature interval. Lambert and collaborators [2550] established that at a temperature above 353°K CCl_4 reacts vigorously with the mercury of the manometer, a fact that forbids the measurement of B and may be the reason for the error in the measured values of B also for temperatures below 353°K.

CBr_4 (gas). Marshall and Staveley [2784] investigated the isothermal compressibility of CBr_4 in the interval from 300-356°K at pressures up to 50 atm.

C_2H_4 (gas). Comprehensive surveys of works carried out in order to determine the p-V-T data of ethylene were published by Keyes [2385] and Nelson, Obert and Evanston [3038]. The most reliable data on the compressibility of C_2H_4 were obtained in a work by Michels and Geldermans [2877] who determined the compressibility of ethylene in the interval from 273-423°K at pressures up to 3000 atm with an accuracy of 0.01%. Making use of the data on the compressibility of ethylene at low pressures Roper [3479] determined the second virial coefficient in the interval from 199-343°K. Hirschfelder, Bird and Spatz [2081] used the p-V-T data of C_2H_4 taken from the work by Michels and Geldermans and determined the second and third virial coefficients of ethylene in the interval from 273-423°K and calculated the force constants of the in-

termolecular Lennard-Jones potential. On the basis of the experimental p-V-T data Din and Hadley [1350] tabulated the thermodynamic properties of ethylene in the interval from 273-423°K at pressures up to 250 atm.

C_2H_3F (gas), C_2H_3Cl (gas). Fogg and Lambert [1572] proposed empirical equations for the second virial coefficient of the monohalogen-substitution products of ethylene:

$$C_2H_3F: \ln(-B) = 8,538 - 9,67 \cdot 10^{-3} T,$$
$$C_2H_3Cl: \ln(-B) = 7,218 - 4,01 \cdot 10^{-3} T.$$

$C_2H_2F_2$ (gas). Mears and collaborators [2826] determined the constants in the Beattie-Bridgman equation of state for 1,1-difluoro ethylene on the basis of an experimental investigation of the p-V-T properties of $C_2H_2F_2$ in the interval from 233-353°K and tabulated the thermodynamic properties of saturated $C_2H_2F_2$ vapor for the interval from 233-298°K. Fogg and Lambert [1572] proposed an empirical equation for the second virial coefficient of 1,1-difluoro ethylene

$$\ln(-B) = 8,435 - 10,41 \cdot 10^{-3} T.$$

$C_2H_2Cl_2$ (gas). Sugawara and Nozawa [3897] published tables of the thermodynamic properties of $C_2H_2Cl_2$ that are based on their own experimental investigations. Fogg and Lambert [1572] proposed empirical equations for the second virial coefficient of cis dichloro ethylene

$$\ln(-B) = 8,830 - 6,02 \cdot 10^{-3} T$$

and trans dichloro ethylene

$$\ln(-B) = 9,221 - 7,43 \cdot 10^{-3} T.$$

C_2HF_2Cl (gas). Mears and collaborators [2826] investigated the p-V-T properties of 2-chloro-1,1-difluoro ethylene. In the same work the constants of the Beattie-Bridgman equation are determined and tables of the thermodynamic properties of C_2HF_2Cl for the interval from 233-393°K are given.

C_2HCl_3 (gas), C_2F_4 (gas). Fogg and Lambert [1572] proposed empiri-

cal equations for the second virial coefficients of trichloro ethylene

$$\ln(-B) = 9,186 - 5,93 \cdot 10^{-3} T$$

and tetrafluoro ethylene

$$\ln(-B) = 9,255 - 12,82 \cdot 10^{-3} T.$$

C_2F_3Cl (gas). Oliver, Grisard and Cunningham [3130] investigated the p-V-T properties of chloro-trifluoro ethylene in the interval from 298 to 379°K at low pressures.

C_2H_2 (gas). The most accurate measurements of the compressibility of acetylene at 273 and 298°K at pressures up to 12 atm were carried out by Sameshima [3576]. Vestin [4104] measured the compressibility at 298°K for pressures up to 1.5 atm. Schaefer [3604] determined the second virial coefficient of acetylene in the interval from 200-273°K. Bottomley, Reeves and Seiflow [873] investigated the p-V-T properties in the interval from 273-313°K at a pressure of 0.2 atm and determined the values of the second virial coefficient in the mentioned temperature interval. On the basis of the data obtained by Schaefer, Vestin and Bottomley, Reeves and Seiflow, the authors of the Handbook calculated the force constants of the intermolecular Lennard-Jones potential. Using the experimental data on p-V-T properties Din [1350] tabulated the thermodynamic properties of acetylene in the interval from 160-320°K for pressures from 0.1 to 100 atm. Wieber [4191] tabulated the thermodynamic properties of C_2H_2 in the interval from 273-523°K for pressures up to 140 atm..

C_2N_2 (gas). Hamann, McManamey and Pearse [1939] determined the values of the second virial coefficient of cyanogen gas in the interval from 308-423°K with an accuracy of ± 4 cm³/mole. In the same work reliable values of the force constants of the intermolecular Lennard-Jones potential are calculated.

SiF_4 (gas). On the basis of experimental p-V-T data Moles [2936]

determined the volatility of SiF_4 . Hamann, McManamey and Pearse [1939] determined the values of the second virial coefficient in the interval from 293-353°K with an accuracy of $\pm 4 \text{ cm}^3/\text{mole}$. In the same work the force constants of the intermolecular Lennard-Jones potential are calculated; they are sufficiently reliable since they were verified in a later work by Hamann and Lambert [1937].

BF_3 (gas). Brooks and Raw [1973, 3403a] verified experimentally that the values of the second virial coefficient of BF_3 may be calculated exactly enough by means of the values of the force constants of the intermolecular Lennard-Jones potential obtained from measuring the viscosity of boron trifluoride [2695].

BCl_3 (gas). Howes, Mackenzie and Raw [1977] determined the force constants of the intermolecular Lennard-Jones potential, making use of viscosity measurements of BCl_3 . In analogy to BF_3 these values of the force constants evidently may be used to calculate the virial coefficients.

Hg (gas). Epstein and Powers [1486] determined the force constants of the intermolecular Lennard-Jones potential on the basis of an investigation of the viscosity of mercury vapors in the interval from 491-883°K. These authors also calculated the values of the second virial coefficient of Hg vapors and proposed an equation for B

$$B = 56,4 - 43,82 e^{\frac{685}{T}} \text{ cm}^3/\text{mole}$$

Eck [1446] compiled tables and diagrams of the thermodynamic properties of mercury vapors on the basis of the experimental data.

He (gas). The most comprehensive survey of works on the investigation of the p-V-T data of helium is given in the work by Gratch [1842]. Bridgman [936] determined the compressibility of helium in the temperature interval from 303-368°K at pressures from 3000 to 15,000 atm.

Tanner and Maccon [3942] investigated the p-V-T properties of He in the interval from 298-448°K at pressures from 30 to 125 atm. Wiebe, Gaddy and Heins [4256] measured the compressibility of helium at temperatures from 203 to 473°K and at pressures up to 1000 atm. The compressibility of He at high temperatures (up to 873°K) was measured by Schneider and Duffie [3646].

In Zaalishvili's work [193] a sufficiently comprehensive survey of works on the determination of the experimental values of the virial coefficients of helium is given. The most reliable values of the second virial coefficient of helium in the interval of 273-1473°K are obtained in the works by Schneider and Duffie [3646] and Yntema and Schneider [4356]. The accuracy of the measurements of B in these works attains approximately 0.5% at 273°K and 3.5% at 1473°K.

White, Rubin, Camky and Johnston [4239a] investigated the compressibility of helium in the temperature interval from 20-300°K at pressures from 1-33 atm. By means of the obtained results the authors calculated the values of the second, third and fourth virial coefficients in the mentioned interval. Using the experimental values of B obtained at low temperatures Michels and Wouters [2896] calculated the values of the force constants of the intermolecular Lennard-Jones potential and the quantum-mechanical parameter λ^* . Simmons [3726] tabulated the thermodynamic properties of helium on the basis of the experimental p-V-T data.

Ne (gas). Crommelin, Martiner and Onnes [1223] determined the p-V-T data of neon in the interval from 56-293°K for pressures up to 93 atm. Making use of the experimental data on compressibility Maslan and Littman [2800] worked out diagrams showing the compressibility of neon in the interval from 36-666°K and for pressures up to 3500 atm. Nickolson and Schneider [3068] measured the compressibility of neon at tem-

peratures from 273 to 973°K and at pressures from 10 to 80 atm.

Keesom and Lammeren [2347] calculated the values of second virial coefficient in the interval from 30-80°K from data on sonic speed. In this work they used the values of B calculated by Crommelin, Martiner and Onnes [1223] from data on compressibility, for higher temperatures. On the basis of data on the compressibility of neon Nickolson and Schneider [3068] calculated the values of B in the interval from 273-973°K. Holborn and Otto [2103] calculated the force constants of the intermolecular Lennard-Jones potential, making use of the experimental values of B: $\epsilon/k = 34.9^\circ\text{K}$, $b_0 = 27.1 \text{ cm}^3/\text{mole}$. Corner [1183] obtained the values of the force constants $\epsilon/k = 36.3^\circ\text{K}$, $b_0 = 28.14 \text{ cm}^3/\text{mole}$, from data on the crystal structure of Ne and from the experimental values of B. Obviously, the most accurate values of the force constants were obtained by Buckingham [1003] from a careful evaluation of the experimental data on the second virial coefficient.

Ar (gas). The first accurate measurements of the compressibility of argon were carried out by Onnes and Crommelin [3142] for the low-temperature interval from 123-293°K and for pressures from 1 to 62 atm. Later on, Cragoe [1209] obtained more accurate data for the aforementioned low-temperature interval. The data obtained by Holborn and collaborators [2101, 2102, 2103, 2104] comprise a temperature interval from 73-673°K for pressures up to 105 atm. In a work by Noury and collaborators [3105] the compressibility of argon from the critical temperature up to 523°K for pressures up to 1200 atm was determined on the basis of ultrasonic speed measurements. Tanner and Maccon [3942] measured the compressibility of argon between 298 and 448°K for pressures from 30 to 125 atm. The compressibility of argon at low pressures in the interval from 273-373°K was measured by Oishi [3121]. Bridgman [938, 939] determined the p-V-T data of argon at high pressures up to

15,000 atm in the interval from 100-328°K. The most accurate p-V-T data were obtained by Michels and collaborators [2894] in the interval from 273-423°K at pressures up to 2900 atm. In a later work, Michels, Levelt and De Graaff [2882] determined the compressibility of argon at temperatures from 118-248°K at pressures up to 1050 atm. Whalley, Lupien and Schneider [4229, 4230] measured the compressibility of argon in the interval from 273-873°K at pressures from 10-80 atm.

Van Itterbeck and Paemel [4050] calculated the values of the second virial coefficient of argon in the interval from 80-273°K, from sonic speed measurements. In a work by Kerr [2373] more reliable values of B of argon for low temperatures are given. Bird and Spatz [821] experimentally determined the values of the second virial coefficient in the interval from 100-323°K.

Lecocq [2579a] carried out an experimental determination of the equation of state of argon up to 1273°K and 1000 atm. From these data the author determined the second, third and fourth virial coefficients and compared them with the virial coefficients calculated on the basis of the different intermolecular potentials of argon. The best agreement was obtained for the exp-6 potential, where the repulsion is represented by an exponential function, and the attraction is a sixth-power one, as in the case of the Lennard-Jones potential. But there is also a satisfactory agreement for the Lennard-Jones potential.

Michels and collaborators [2892] determined the values of B in the interval from 273-423°K making use of the p-V-T data. From the data on compressibility, Whalley, Lupien and Schneider [4229, 4230] calculated the values of the second and third virial coefficients in the interval from 173-873°K. Making use of crystallographic data and the experimental values of B, Corner [183] calculated the force constants of the intermolecular Lennard-Jones potential: $\epsilon/k = 119.3^\circ\text{K}$ and $b_0 = 51.71$

cm³/mole. Michels and collaborators [2893] suggested the values $\epsilon/k = 119.8^\circ\text{K}$ and $b_0 = 49.8 \text{ cm}^3/\text{mole}$ for the force constants, on the basis of the p-V-T data. Whalley and Schneider [4232] calculated the values of the force constants $\epsilon/k = 119.49 - 0.33^\circ\text{K}$ and $b_0 = 49.92 \pm 0.35 \text{ cm}^3/\text{mole}$, on the basis of the experimental values of B obtained in the interval from 173-873°K. Thomas [3966] compared the calculated and experimental values of B and showed that the values of the force constants suggested by Whalley and Schneider are the most accurate.

Bahadur and Madan [607a] calculated the values of the force constants of the intermolecular potential of argon making use of the results of determining the values of the third virial coefficient well-known in literature. In view of the fact that the third virial coefficient is very sensitive to the form of the intermolecular potential the values of the force constants obtained from those of the third virial coefficient may be very reliable. But since the accuracy of the values of the third virial coefficient used by the authors was not high we have, at present, no reason for assuming that the values of the force constants of argon $\epsilon/k = 120.5^\circ\text{K}$ and $b_0 = 50.42 \text{ cm}^3/\text{mole}$ recommended in the work by Bahadur and Madan are more reliable than those proposed by Whalley and Schneider.

Michels and collaborators [2883, 2895] tabulated the thermodynamic properties of argon in the interval from 273-423°K for pressures up to 2900 atm, on the basis of the p-V-T data. Whalley [4229] tabulated the thermodynamic properties of Ar in the temperature interval from 173-873°K and for pressures up to 2900 atm, making use of experimental data on compressibility. The tables of the thermodynamic properties of argon compiled in the US National Bureau of Standards [2076] on the basis of the experimental data on the p-V-T properties, and the equations of state with virial coefficients comprise a temperature interval from

1-100 atm. Din [1351] tabulated the thermodynamic properties of Ar in the interval from 273-423°K for pressures up to 3000 atm, making use of the most accurate experimental data on the compressibility of argon.

Kr (gas). Beattie, Brierley and Barriault [704] investigated the compressibility of krypton in the interval from 273-573°K at pressures from 45 to 470 atm. At higher temperatures from 273-873°K and pressures from 10 to 80 atm the compressibility of krypton was measured by Whalley and Schneider [4231].

On the basis of the experimental data on compressibility Beattie, Brierley and Barriault [705] as well as Whalley and Schneider [4231] determined the values of the second virial coefficient of krypton in the mentioned temperature intervals. On the basis of crystallographic data, Corner [1180] calculated the values of the force constants of the intermolecular Lennard-Jones potential: $\epsilon/k = 159^\circ\text{K}$, $b_0 = 58.8 \text{ cm}^3/\text{mole}$. Beattie, Brierley and Barriault [705] obtained $\epsilon/k = 172.7^\circ\text{K}$ and $b_0 = 58.4 \text{ cm}^3/\text{mole}$, on the basis of the values of the second virial coefficient. As was shown by Thomas [3966], Whalley and Schneider [4232] obtained the most accurate values of the force constants $\epsilon/k = 166.67 \pm 0.47^\circ\text{K}$ and $b_0 = 62.92 \pm 0.44 \text{ cm}^3/\text{mole}$.

The values of the force constants $\epsilon/k = 193.7^\circ\text{K}$ and $b_0 = 64.87 \text{ cm}^3/\text{mole}$ obtained in the work by Bahadur and Madan [607a] on the basis of the well-known values of the third virial coefficient of triton, cannot be considered sufficiently reliable since even the values of the third virial coefficient were obtained with low accuracy.

Xe (gas). The compressibility of xenon was investigated by Beattie, Barriault and Brierley [705] in the interval from 17-300°K at pressures up to 300 atm. On the basis of the compressibility measurements these authors determined the values of the second and third virial coefficients in this temperature interval. The p-V-T data obtained by Michels,

Wassenar and Lauwerse [2892] comprise an interval from 273-423°K at pressures up to 2800 atm. Whalley, Lupien and Schneider [4230] measured the compressibility of xenon at temperatures from 273-973°K and pressures from 8 to 50 atm. Besides, Michels, Wassenar and Lauwerse as well as Whalley, Lupien and Schneider determined the second and third virial coefficients.

Corner [1180] determined the values of the force constants of the intermolecular Lennard-Jones potential: $\epsilon/k = 228^\circ\text{K}$ and $b_0 = 79.11 \text{ cm}^3/\text{mole}$ on the basis of crystallographic data. Newitt [3062] obtained $\epsilon/k = 217^\circ\text{K}$ and $b_0 = 78.5 \text{ cm}^3/\text{mole}$. On the basis of the experimental data of B, Beattie, Barriault and Brierley [705] found $\epsilon/k = 224.5^\circ\text{K}$ and $b_0 = 84.65 \text{ cm}^3/\text{mole}$. The values of the force constants $\epsilon/k = 212.8^\circ\text{K}$ and $b_0 = 94.8 \text{ cm}^3/\text{mole}$ obtained by Michels, Wassenar and Lauwerse [2892] differ considerably from the data obtained by other authors. As was shown by Thomas [3966], the most accurate values of the force constants of xenon $\epsilon/k = 225.3 \pm 1.1^\circ\text{K}$ and $b_0 = 84.96 \pm 0.84 \text{ cm}^3/\text{mole}$ were obtained by Whalley and Schneider [4232] on the basis of crystallographic data and the experimental values of B.

On the basis of the experimental data on the compressibility of xenon Michels, Wassenar, Wolkers and Dawson [2893] tabulated the thermodynamic properties of Xe in the interval from 273-423°K for pressures up to 2800 atm.

- 2025 When employing this method we must keep in mind that any empirical equation of state, even when it covers the p-V-T-data on a certain gas very accurately in a wide range of pressure and temperature, may lead to inexact or even wrong values of entropy and specific heat capacity of this gas because for the calculation of these quantities one has to find the first and second derivatives of volume with respect to p and T; even with an equation of state that is exact in the above sense, inexact values of these derivatives might be obtained.
- 2035 Some calculations of the virial coefficients have been made for Lennard-Jones potentials with a repulsion exponent unequal to 12. Since it has been found in numerous investigations that for many substances a power of twelve leads to a better agreement with the experimental values of the second virial coefficient and the Joule-Thomson coefficient, only the results for a Lennard-Jones (6-12) potential are given in this Appendix. However, for helium, mercury, and some other substances a Lennard-Jones (6-9) potential may turn out to be somewhat better. Calculations of the second virial coefficient by the punched-card method for a Lennard-Jones (6-9) potential with kT/ϵ ranging from 0.5 to 100 have been carried out in a paper by Epstein and Hibbert [1485a]. The third virial coefficient for a (6-9) potential has been calculated in the paper by Kihara [2397].
- 2060 The attempt to calculate the force constants of the intermolecular Lennard-Jones on the basis of these experimental values of B proved to be unsuccessful. The reason for this lack of success has not become clear, as yet.

Manu-
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Page
No.

[Transliterated Symbols]

2028 ид = id = idel'nyy = ideal
2033 станд = stand = standartnyy = standard
2034 кр = kr = kriticheskiy = critical
2035 полн = poln = polnyy = full
2044 кв = kv = kvantovoy = quantum

Appendix 6

CRITICAL CONSTANTS

Table 321 shows the adopted values of the critical constants of some substances considered in the Handbook. Some of these values were adopted according to data in literature, some of them were estimated by means of a method set forth below.

At present, experimental data on the critical constants are missing for the majority of compounds with a boiling point above 500°K. Since the experimental determination of the critical constants of substances with high boiling points is a very complex problem, several empirical relations were proposed in order to estimate the values of these quantities. In a work by Kobe and Lynn [2451] a detailed review of the relations proposed by several authors up to 1953 is given. Nevertheless, these relations that were obtained on the basis of experimental data for substances with low boiling points and their application to an estimation of the critical constants of substances with high boiling points have neither a theoretical nor an experimental foundation.

Prudhomme [3341, 3342, 3343] suggested a simple, but inaccurate relation between the critical temperature and the boiling and melting temperatures

$$T_{cp} = T_{bp} + T_{mp}, \quad (P6.1)$$

where T_{kip} is the boiling point at 760 mm Hg. The possibility of applying Prudhomme's formula to an estimation of the critical temperatures of liquids with high boiling points is very problematical since even

for substances with low boiling points this formula has very many exceptions. Phosphorus, sulfur and mercury are the substances with the highest boiling points for which more or less reliable values of the critical constants are available, at present. The estimation of the values of T_{kr} for these substances according to the Prudhomme formula leads to values which differ considerably from the experimental ones. Thus, e.g., for mercury the value of $T_{kr} = 865^{\circ}\text{K}$ is obtained whereas the experimental value is $T_{kr} = 1733^{\circ}\text{K}$.

Guldberg [1884] found that for many substances the relation

$$T_{kp} = 1,55 T_{km} \quad (\text{P6.2})$$

holds true. Several authors [1968, 3562] applied this formula to an estimation of the critical temperatures of alkali and alkali earth metals. It is, however, difficult to estimate the accuracy of the obtained values of the critical temperatures. Besides, we have to note that the ratio T_{kr}/T_{kip} is, respectively, equal to 1.75, 1.83 and 2.75 for phosphorus, sulfur and mercury.

With the help of the principle of the corresponding states and Guldberg's equation Lorenz and Herz [2655] proposed the relations

$$T_{kp} = 2,273 T_{ms} \quad (\text{P6.3})$$

and

$$V_{kp} = 3,121 V_{ms} \quad (\text{P6.4})$$

for alkali metal salts, where V_{kr} is the critical volume, and V_{pl} is the specific volume at the melting point.

We must note that Guldberg's formula is correct only for some low-boiling compounds and its application to an estimate of the critical temperatures of high-boiling substances cannot be proved in any serious way.

Guareschi [1870] suggested the following relation in order to estimate the critical temperatures of metals

TABLE 321

Adopted Values of Critical Constants

Вещество 1	$T_{кр}, ^\circ K$ 2	Ссылка 3	$\rho_{кр}, \text{г/мл}$ 4	5 Ссылка	$V_{кр}, \text{см}^3/\text{моль}$ 6	7 Ссылка
O ₂	154,77	[2097]	50,14	[2097]	78	[2451]
O ₃	261,05	[2224]	54,6	[2224]	89,4	[3442]
H ₂	33,23	[4235]	12,797	[4235]	64,99	[2805]
H ₂ O	647,4	[2451]	218,3	[2451]	56	[2451]
H ₂ O ₂	730	[3599]	214	[3599]	—	—
D ₂	38,35	[1616, 1617]	16,432	[1616, 1617]	60,3	[1616, 1617]
DH	35,91	[570, 2098]	14,64	[570, 2098]	62,8	[570, 2098]
D ₂ O	644,2	[3129]	215,7	[3129]	54,8	[1447]
T ₂	43,7	[1942]	20,8	[1942]	53,7	[1942]
HT	38,3	[1616]	16,4	[1616]	60,3	[1616]
DT	39,5	[1616]	17,3	[1616]	58,5	[1616]
He	5,25	[2451]	2,26	[2451]	57,8	[2451]
Ne	44,5	[2451]	26,9	[2451]	41,7	[2451]
Ar	151	[2451]	48,0	[2451]	75,2	[2451]
Kr	209,4	[2451]	54,3	[2451]	92,2	[2451]
Xe	289,74	[4195]	57,64	[1917]	119,5	[1917]
F ₂	144	[1042]	55	[1042]	—	—
F ₂ O	215	[553]	48,9	[553]	97,6	[553]
HF	461	[1592]	64,1	[1592]	69	[1592]
Cl ₂	417	[3214]	76,1	[3214]	124	[3214]
HCl	324,6	[2451]	81,5	[2451]	48	[2451]
Br ₂	584	[3613]	102	[3613]	135	[2180]
HBr	363,2	[2451]	84,0	[2451]	98	[2451]
I ₂	826	[3402]	—	—	—	—
HI	423	[2451]	81,0	[2451]	117	[2451]
S	1313	[2451]	116	[2451]	80	[2451]
SO ₂	430,7	[2451]	77,8	[2451]	122	[2451]
SO ₂	491,4	[799]	83,8	[799]	126	[799]
H ₂ S	373,6	[2451]	88,9	[2451]	97,7	[2451]
SF ₄	347	[985]	—	—	—	—
SF ₆	318,7	[2694]	37,113	[2694]	194,3	[585]
N ₂	126,25	[4236]	33,54	[4236]	90,1	[2807]
NO	180	[2451]	64	[2451]	58	[2451]
NO ₂	431	[2451]	100	[2451]	82	[2451]
N ₂ O	309,7	[2451]	71,7	[2451]	96,3	[2451]
NH ₃	405,5	[2451]	111,3	[2451]	72,48	[2451]
N ₂ H ₄	653	[999]	145	[999]	—	—
NF ₃	233,9	[2218]	44,72	[2218]	—	—
P	968	[3799]	824	[3799]	—	—
PF ₃	271,1	[866, 867]	42,69	[866, 867]	—	—
C	> 20 000	Оценка 8	—	—	—	—
CO	132,91	[2803, 2804]	34,529	[2803, 2804]	93,06	[2803, 2804]
CO ₂	304,19	[2874]	72,85	[2874]	94,04	[2874]
CH ₄	190,55	[2056a]	45,41	[2056a]	99	[2451]
CF ₄	227,7	[2451]	36,9	[2451]	136	[2451]
CHF ₃	317,8	[2451]	58,0	[2451]	113	[2451]
CHF ₃	356	[470]	—	—	—	—
CHF ₃	300	[470]	—	—	—	—

TABLE 321 (continued)

Вещество 1	$T_{кр.} \text{ } ^\circ\text{K}$ 2	Ссылка 3	$P_{кр.} \text{ атм}$ 4	Ссылка 5	$V_{кр.} \text{ см}^3/\text{моль}$ 6	Ссылка 7
OCl_2	556,4	[2451]	45	[2451]	275	[2451]
Cl_2CO	445	[2451]	50	[2451]	190	[2451]
CH_2Cl_2	416,3	[2451]	65,9	[2451]	143	[2451]
CH_2Cl_2	510	[2451]	60	[2451]	—	—
CHCl_3	530,6	[2451]	54	[2451]	240	[2451]
CF_2Cl_2	302	[2451]	39	[2451]	218	[2451]
CF_2Cl_2	384,7	[2451]	39,6	[2451]	218	[2451]
CFCl_3	471,2	[2451]	43,2	[2451]	248	[2451]
CH_2FCl	427	[470]	—	—	—	—
CBr_4	715	[470]	—	—	—	—
CH_2Br_2	464	[470]	—	—	—	—
CHBr_3	662	[470]	—	—	—	—
CF_2Br_2	333	[470]	—	—	—	—
CF_2Br_2	464	[470]	—	—	—	—
CFBr_3	591	[470]	—	—	—	—
CH_2FBr	440	[470]	—	—	—	—
CHF_2Br	409	[470]	—	—	—	—
CH_2FBr_2	532	[470]	—	—	—	—
OCl_2Br	600	[470]	—	—	—	—
OCl_2Br_2	653	[470]	—	—	—	—
OClBr_3	700	[470]	—	—	—	—
CH_2ClBr	575	[470]	—	—	—	—
CHCl_2Br	582	[470]	—	—	—	—
CHClBr_2	627	[470]	—	—	—	—
CF_2ClBr	430	[470]	—	—	—	—
CFCl_2Br	520	[470]	—	—	—	—
CFClBr_2	570	[470]	—	—	—	—
CH_2J_2	528	[470]	—	—	—	—
CH_2J_2	608	[470]	—	—	—	—
CHJ_3	690	[470]	—	—	—	—
CF_2J_2	359	[470]	—	—	—	—
CF_2J_2	477	[470]	—	—	—	—
CHF_2J	459	[470]	—	—	—	—
COS	375,4	[3194]	62	[3867]	—	—
CS_2	552	[2451]	78	[2451]	170	[2451]
HCN	456,65	[914]	53,2	[914]	138,6	[914]
C_2H_2	309,65	[2708]	61,6	[2708]	113	[2451]
C_2H_4	282,42	[2336]	50,6	[2336]	123,6	[2336]
$\text{C}_2\text{H}_2\text{F}_2$	303,2	[2826]	43,8	[2826]	154	[2626]
C_2F_4	306,45	[3422]	38,9	[3422]	173	[3422]
$\text{C}_2\text{F}_2\text{Cl}_2$	379,0	[3130]	40,1	[3130]	211	[3130]
$\text{C}_2\text{HF}_2\text{Cl}_2$	400,6	[2826]	44,0	[2826]	197	[2826]
C_2N_2	400	[2451]	59	[2451]	—	—
Si	>6000	Оценка	—	—	—	—
SiO_2	>6000	8	—	—	—	—
SiF_4	259,0	[869]	36,66	[869]	—	—
SiCl_4	506,8	[3189]	—	—	—	—
SiC	—	—	Разлагается ниже $T_{кр}$	9	—	—
Pb	4500	Оценка 8	500	Оценка 8	—	—
PbO	5300	8	3250	8	—	—

TABLE 321 (continued)

Вещество ¹	T_{kr} , °K ²	3 Ссылка	P_{kr} , атм ⁴	5 Ссылка	V_{kr} , ⁶ см ³ /моль	7 Ссылка
PbF ₂	3150	8 Оценка	215	Оценка 8	—	—
Hg	1733	[816]	1640	[816]	48	[1063]
Zr	>6000	8 Оценка	—	—	—	—
ZrO ₂	>6000	"	—	—	—	—
B	>6000	"	—	—	—	—
B ₂ O ₃	>6000	"	—	—	—	—
BF ₃	280,9	[869]	49,2	[869]	—	—
BCl ₃	452	[3189]	38,2	[3892]	148	[1977]
Al	>6000	8 Оценка	—	—	—	—
Al ₂ O ₃	>6000	"	—	—	—	—
AlF ₃	4700	"	3800	Оценка	—	—
AlCl ₃	1350	"	150	"	—	—
AlN						
Be	>6000	8 Оценка	9 Разлагается ниже T_{kr}			
BeO	>6000	"	—	—	—	—
BeF ₂	5000	"	5400	8 Оценка	—	—
BeCl ₂	2800	"	8000	"	—	—
Mg	3000	"	445	"	—	—
MgO	>6000	"	—	—	—	—
MgF ₂	5000	"	140	8 Оценка	—	—
MgCl ₂	2900	"	62	"	—	—
Ca	3400	"	150	"	—	—
CaO	>6000	"	—	—	—	—
Sr	3150	"	150	8 Оценка	—	—
SrO	>6000	"	—	—	—	—
Ba	3750	"	200	8 Оценка	—	—
BaO	>6000	"	—	—	—	—
Li	3500	"	390	8 Оценка	—	—
Li ₂ O	>6000	"	—	—	—	—
LiH	3450	"	110	8 Оценка	—	—
LiOH	4000	"	160	"	—	—
LiF	4700	"	700	"	—	—
LiCl	3500	"	235	"	—	—
Na	2200	"	125	"	—	—
NaF	4300	"	350	"	—	—
NaCl	3450	"	150	"	—	—
K	1700	"	43	"	—	—
KF	3800	"	250	"	—	—
KCl	3350	"	205	"	—	—
Rb	1600	"	38	"	—	—
Cs	1550	"	32	"	—	—

1) Substance; 2) T_{kr} , °K; 3) reference; 4) P_{kr} , atm; 5) reference; 6) V_{kr} ; cm³/mole; 7) reference; 8) estimate; 9) is decomposed below T_{kr} .

$$T_{\text{cp}} = 3 T_{\text{кр}}. \quad (\text{P6.5})$$

This formula yields a more or less correct value of $T_{\text{кр}}$ only for mercury, and it is hardly possible to prove seriously if it can be applied to estimate $T_{\text{кр}}$ of the rest of the metals.

Waldschmidt [4130] proposed an empirical relation on the basis of the values of critical temperatures obtained experimentally for a rather considerable number of compounds

$$T_{\text{cp}} = (1,2 + 0,125 \sqrt[3]{M}) T_{\text{кр}}. \quad (\text{P6.6})$$

where M is the mean molecular weight at the critical point. The author of the work [4131] used this formula to estimate the critical temperatures of metals. In order to clarify the considerable discrepancy between the experimental values of $T_{\text{кр}}$ and those calculated from Formula (P6.6) Waldschmidt was forced to introduce the assumption that some substances are polymerized at the critical point. He assumed that at the critical point methane exists in the form $(\text{CH}_4)_4$, water in the form $(\text{H}_2\text{O})_4$, sulfur in the form S_4 , mercury in the form Hg_9 , etc. In most cases these assumptions were not verified experimentally, a fact that forbids us to consider the values of $T_{\text{кр}}$ suggested by Waldschmidt to be reliable to any extent.

On the basis of the statistical theory of surface tension Zadumkin [194a] proposed a formula to be used to estimate approximately the critical temperatures of metals with cubic structure

$$T_{\text{cp}} = \frac{\sqrt{0,481 + \frac{4}{3} m \Delta H_s \left(\frac{\alpha a \theta_D}{h}\right)^2} - 0,693}{2mk \left(\frac{\alpha a \theta_D}{h}\right)}. \quad (\text{P6.7})$$

where θ_D is the characteristic Debye temperature, k is the Boltzmann constant, m is the atomic mass, α is the thermal coefficient of linear expansion of metals, a is the half period of the crystal lattice, ΔH_s is the heat of sublimation at $T = 0^\circ\text{K}$ (relative to one atom), $h = h/2\pi$

(h is Planck's constant).

From this formula Zadumkin calculated the values of the critical temperatures of several metals. As shown by the same author, however, the values of the critical temperatures as calculated from the proposed formula are too high since in the calculation of T_{kr} a value obtained for melting at a temperature close to the melting point of the corresponding metal was chosen for the magnitude of α . Unfortunately, the fact that there are no experimental values of α does not permit carrying out more accurate calculations.

Gates and Thodos [1657a] considered the connection between the critical temperature T_{kr} and the boiling point T_{kip} for 16 simple substances, making use of a survey of the experimental data given in literature. The authors found that the best representation of this connection is given by the equation

$$T_{kr} = 1,4732 T_{kip}^{1.0219}. \quad (P6.8)$$

On the basis of Eq. (P6.8) Gates and Thodos calculated the critical constants of several metals. We must, however, mention that although Eq. (P6.8) was recommended by Gates and Thodos for estimating the critical temperatures of simple substances, the values of T_{kr} calculated from Eq. (P6.8) for such simple substances as helium and mercury differ essentially from the experimental values of T_{kr} .

Since Eqs. (P6.1)-(P6.8) do not enable us to obtain reliable values of the critical temperatures of high-boiling substances, a new relation which could be applied to an estimation of the critical temperatures of both high and low-boiling substances was obtained when the Handbook was prepared (see [80v]).

As is well known, the pressure of saturated vapor can be found from the equation

$$\lg p = \int \frac{\Delta H_s}{4,5758 T^2} dT + \text{const.} \quad (\text{P6.9})$$

On the basis of the works [2463a, 3829a] the dependence of the heat of sublimation ΔH_s on temperature may be represented by the empirical equation

$$\Delta H_s = (n + 1) A^{n+1} \frac{T^2}{T_{\text{kp}}} \left(1 - \frac{T}{T_{\text{kp}}}\right)^n, \quad (\text{P6.10})$$

where the values of n and A are determined from the experimentally obtained values of ΔH_s and T_{kr} . For most of the nonassociated liquids we have $n = 0.5$ and $A = 0.284$. Substituting the value of ΔH_s from (P6.10) into Eq. (P6.9) and integrating (P6.9) from T_{kip} to T_{kr} , we obtain

$$\lg p_{\text{kp}} = 0,1514 \left(1 - \frac{T_{\text{ккк}}}{T_{\text{kp}}}\right)^{1/2}. \quad (\text{P6.11})$$

Hence

$$T_{\text{kp}} = \frac{T_{\text{ккк}}}{1 - 0,284 (\lg p_{\text{kp}})^{1/2}}. \quad (\text{P6.12})$$

Since Eq. (P6.10) holds true for nonassociated liquids, strictly speaking, also Eq. (P6.12) can only be used to estimate the critical temperatures of nonassociated liquids. For an associated liquid the exponent n in the relation (P6.10) must be greater than 0.5. Even for strongly associated liquids, however, the calculated values do not exceed the experimental ones by more than 15-18% (see [80a]). This fact permitted the use of Eq. (P6.12) to estimate the values of the critical constants of substances with high boiling points. Table 321 shows the values of the critical constants for such compounds.

The calculation of the critical constants was carried out by the method of successive approximations. To estimate the values of T_{kr} and p_{kr} in the first approximation the tables of pressure of saturated vapors calculated in the present Handbook from the equation

$$4,57584 \lg p = -\frac{\Delta H_{\text{so}}}{T} + \Phi_{\text{гас}}^{\circ} - \Phi_{\text{конд}}^{\circ}. \quad (\text{P6.13})$$

were used.

The value of T at which T and p satisfy Eq. (P6.12) was determined by the method of selection, by means of the tables of pressure of saturated vapors. These values of T and p were also chosen to be the first approximation of T_{kr} and p_{kr} . On the basis of these values of the critical constants the nonideality corrections to Φ_{gaz}^* were calculated:

$$\delta\Phi_{gaz}^* = 0,13973 \frac{p}{p_{kr}} \frac{T_{kr}}{T} \left(6 \frac{T_{kr}^2}{T^2} - 1 \right). \quad (P6.14)$$

The correction in Φ_{kond}^* was not taken into account in the given approximation. Tables of the pressure of saturated vapors were, then, compiled on the basis of the equation

$$4,57584 \lg p = -\frac{\Delta H_{20}}{T} + \Phi_{gaz}^* + \delta_1 \Phi_{gaz}^* - \Phi_{kond}^*. \quad (P6.15)$$

With the help of the tables compiled by means of Eq. (P6.15) the values of T_{kr} and p_{kr} were determined in the second approximation, etc. The calculations were carried out until the difference of two successively obtained values of T_{kr} was not greater than 50° .

The estimation of the values of critical temperatures was limited by 6000°K , i.e., the temperature up to which the values of the thermodynamic properties of substances are given in this Handbook. We must note that at high temperatures (above $2000-3000^\circ\text{K}$) the concept "critical state" in some cases, obviously, cannot be understood in the sense in which it is used at lower temperatures. The point is that at high temperatures the composition of the vaporization products of the substance is very complex in many cases since besides the molecules of the evaporating substance, associated molecules and dissociation products are present in the vapors. On the other hand, heating the substance up to high temperatures is in some cases accompanied by a change of the composition of the condensed phase (e.g., $2\text{LiOH}(\text{krist.}) \rightarrow \text{Li}_2\text{O}(\text{krist.}) + \text{H}_2\text{O}(\text{gaz})$).

Consequently, the problem whether the application of the concept "critical state" to processes essentially representing a complex chemical reaction rather than an evaporation in the usual sense of this term is justified remains unsolved.

Manu-
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Page
No.

[Transliterated Symbols]

2091	кип = kip = kipeniye = boiling
2091	кр = kr = kriticheskiy = critical
2091	пл = pl = plavleniye = melting
2099	газ = gaz = gas
2099	конд = kond = kondensirovanny = condensed
2100	крист = krist = kristallicheskiy = crystalline

SYMBOLS AND DENOTATIONS ADOPTED IN THE HANDBOOK

- a_0, a_1, \dots are the constants in Dunham's equation for the potential energy of a diatomic molecule.
- a, A are the symbols of the first excited electronic state of a diatomic molecule.
- A is the interaction constant of the orbital and spin angular momenta of electron motion.
- A is the affinity to an electron.
- $A_{00\dots 0}$ is the totational constant of a polyatomic molecule in the ground state of vibration.
- A_e is the rotational constant of a polyatomic molecule in the equilibrium state.
- A_S is the constant term in the equation used to calculate S_{post}^*
- $A_{v_1 v_2 \dots v_n}$ is the rotational constant of a polyatomic molecule in the vibrational state (v_1, v_2, \dots, v_n) .
- A_ϕ is the constant term in the equation used to calculate ϕ_{post}^*
- b_0 is the force constant of the intermolecular potential.
- b, B are symbols for the second excited electronic state of a diatomic molecule.
- B is the second virial coefficient.
- B^* is the reduced second virial coefficient.
- B_0 is the rotational constant of a diatomic molecule in the ground state of vibration.

- $B_{00\dots 0}$ is the rotational constant of a polyatomic molecule in the ground state of vibration.
- B_e is the rotational constant of a diatomic or polyatomic molecule in the equilibrium state.
- B_v is the rotational constant of a diatomic molecule in the vibrational state v .
- B_{v_1, v_2, \dots, v_n} is the rotational constant of a polyatomic molecule in the vibrational state (v_1, v_2, \dots, v_n) .
- c is the velocity of light in empty space.
- c, C are the symbols of the third excited electronic state of a diatomic molecule.
- C is the third virial coefficient.
- C^* is the reduced third virial coefficient.
- $C_{00\dots 0}$ is the rotational constant of a polyatomic molecule in the ground state of vibration.
- C_e is the rotational constant of a polyatomic molecule in the equilibrium state.
- C_p is the specific heat at constant pressure.
- $C_{p_{g.o}}$ is the component of the harmonic oscillator in C_p .
- C_S is the constant term in the equation used to calculate S_T° of diatomic gases or polyatomic gases with linear molecules.
- C_S^I is the constant term in the equation used to calculate S_T° of polyatomic gases with nonlinear molecules.
- C_v is the specific heat at constant volume.
- C_{v_1, v_2, \dots, v_n} is the rotational constant of a polyatomic molecule in the vibrational state (v_1, v_2, \dots, v_n) .
- C_Φ is the constant term in the equation used to calculate Φ_T^\ddagger of a diatomic gas and a polyatomic gas with linear

- molecules.
- C_{ϕ}' is the constant term in the equation used to calculate ϕ_T^* of a polyatomic gas with nonlinear molecules.
- d is the electronic state with quantum number $l = 2$.
- d_1 is the degree of degeneracy of the i th vibration of a polyatomic molecule.
- d_0/T is the coefficient in the equations used to calculate $S_{kol.vr.}$ and $\phi_{kol.vr.}^*$.
- D is the atomic state with the quantum number $L = 2$.
- D_0 is the rotational constant (constant of centrifugal stretching) of a diatomic molecule in the vibrational ground state.
- D_0 is the energy of dissociation of a molecule into atoms.
- D_e is the rotational constant (constant of centrifugal stretching) of a diatomic molecule in the equilibrium state.
- D_e is the energy of dissociation of a diatomic molecule in the given electronic state, related to the minimum of the potential curve of this state.
- D_J, D_{JK}, D_K are the constants of the centrifugal stretching of a polyatomic molecule.
- D_v is the rotational constant (constant of the centrifugal stretching) of the diatomic molecule in the vibrational state v .
- e is the electronic charge.
- E is the internal energy.
- $E^{(1)}$ is the internal energy of a molecule in the i th electronic state.
- f is the volatility (fugacity).

- f is the electronic state with the quantum number $l = 3$.
- f_0/T^2 is the coefficient in the equations used to calculate $\Phi_{kol.vr}^*$ and $S_{kol.vr}$.
- $f(K, J - K)$ is the component of the multiplet splitting for the Hund case b.
- f_α is the force constant of deformation.
- $f_{\alpha\alpha}$ is the force constant of the interaction of two neighboring angles.
- $f_{\alpha\alpha'}$ is the force constant of the interaction of two noncontact angles.
- f_d is the valence force constant of bond.
- $f_{d\alpha}$ is the force constant of the interaction between the bond and the angle adjacent to it.
- $f_{d\alpha'}$ is the force constant of the interaction between the bond and an angle not adjacent to it.
- f_{dd} is the force constant of the interaction between two bonds.
- F is the isochoric (isochoric (isochoro-isothermal) thermodynamic potential.
- F is the atomic state with the quantum number $L = 3$.
- $F_v(J)$ is the rotational energy of a diatomic molecule in the vibrational state v.
- $F_v(J, K)$ is the rotational energy of a molecule of the symmetric-gyroscope type.
- $F_v(J_\tau)$ is the rotational energy of molecules of the asymmetric-gyroscope type.
- $F_v^{(1)}(J)$ is the rotational energy of a molecule in the vibrational state v of the ith electronic state.
- S_{11} is the constant of the vibrational momentum for the ith

degenerate vibration of a linear polyatomic molecule.

G is the isobaric (isobaro-isothermal) thermodynamic potential.

G is the atomic state with the quantum number $L = 4$.

$G(0)$ is the zero vibrational energy of a diatomic molecule.

$G(0,0,\dots)$ is the zero vibrational energy of a polyatomic molecule.

$\Delta G_{1/2}$ the difference between the vibrational energies of a diatomic molecule in the states $V = 0$ and $V = 1$.

$\Delta G_{v+1/2}$ is the difference of the vibrational energies in the states $v + 1$ and v .

$G(v_1, v_2, \dots)$ is the vibrational energy of a polyatomic molecule related to the minimum of potential energy.

$G^{(i)}(v)$ is the vibrational energy of a diatomic molecule in the i th electronic state related to the minimum of potential energy.

$G_0(v_1, v_2, \dots)$ is the vibrational energy of a polyatomic molecule, related to the ground state of vibration.

$G_0^{(i)}(v)$ is the vibrational energy of a diatomic molecule in the i th electronic state, related to the ground state of vibration.

h is Planck's constant.

H is the atomic state with the quantum number $L = 5$.

ΔH is the thermal effect of a process at constant pressure (heat of reaction or phase transition).

H is the enthalpy.

H_0 is the rotational constant of a diatomic molecule in the vibrational ground state.

ΔH_c is the heat of combustion.

H_c is the rotational constant of a diatomic molecule in the

equilibrium state.

ΔH^*f is the heat of formation of a substance from the elements in the normal states.

ΔH_m is the heat of fusion.

ΔH_s is the heat of sublimation.

$H_T^\circ - H_0^\circ$ is the variation of the enthalpy of the substance in normal state from 0° to $T^\circ K$.

$H_T^\circ - H_{T_1}^\circ$ is the variation of the enthalpy of the substance in the temperature interval from T_1 to T .

$(H_T^\circ - H_0^\circ)g.o$ is the component of the harmonic oscillator in $H_T^\circ - H_0^\circ$.

ΔH_{tr} is the heat of transformation.

ΔH_v is the heat of evaporation.

H_v is the coefficient with $J^3(J+1)^3$ in the expression for $F_v(J)$.

i is the index of the inverse electronic state.

I is the moment of inertia.

I is the ionization potential.

I is the nuclear spin.

I_0 is the moment of inertia of the molecule in the vibrational ground state.

I_{pr} is the reduced moment of inertia of the molecule in the case of an intramolecular rotation.

$I_{A_0}, I_{B_0}, I_{C_0}$ are the principal moments of inertia of a polyatomic molecule in the vibrational ground state.

$I_{A_e}, I_{B_e}, I_{C_e}$ are the principal moments of inertia of a polyatomic molecule in the equilibrium configuration.

$I_{A_{v_1}}, I_{B_{v_1}}, I_{C_{v_1}}$ are the principal moments of inertia of a polyatomic molecule in the vibrational state (v_1, v_2, \dots, v_n)

- I_e is the moment of inertia of diatomic and polyatomic molecules (linear ones and those of the symmetric-gyroscope type) in the equilibrium state.
- I_T is the total enthalpy of the substance at a temperature of $T^\circ\text{K}$.
- J is the quantum number of the total angular momentum of the electron.
- J is the total angular momentum of the electron.
- J is the rotational quantum number of the molecule.
- J is the quantum number of the total angular momentum of the electrons in the atom.
- J is the total angular momentum of the electrons in the atom.
- J_{max} is the maximum value of J in a given vibrational state.
- J_r is the rotational quantum number of a molecule of the asymmetric-gyroscope type.
- k is the Boltzmann constant.
- k_e is the force constant of a diatomic molecule.
- K is the rotational quantum number.
- K_p is the equilibrium constant of reaction at constant pressure.
- l is the quantum number of the orbital angular momentum of the electron.
- l is the orbital angular momentum of the electron.
- l_n is the quantum number of the vibrational momentum relative to a straight line on which the atoms of a linear polyatomic molecule are located.
- L is the quantum number of the total orbital angular momentum of the electrons in an atom.

- L is the resulting orbital angular momentum of the electrons in a molecule.
- L is the total orbital angular momentum of the electrons in an atom.
- L_e is the rotational constant of a diatomic molecule in the equilibrium state.
- L_v is the coefficient with $J_4(J + 1)^4$ in the expression for $F_v(J)$.
- m_i is the mass of the ith atom in the molecule.
- m_l is the magnetic quantum number of the electron.
- m_e is the electronic mass.
- m_j is the quantum number of the component j in the direction of the magnetic field.
- m_s is the quantum number of the electron spin.
- M is the molecular weight.
- M_L is the projection of L on the internuclear axis.
- n is the principal quantum number of the electron.
- n is the number of the vibrational degrees of freedom of a polyatomic molecule.
- n is the number of maxima on the potential curve of internal rotation.
- n_{\max} is the maximum value of the principal quantum number n.
- N is the Avogadro number.
- N is the angular momentum of the nuclei in a molecule.
- O-vetv' is the line series in a band corresponding to the transitions $\Delta J = 2$.
- p is the pressure.
- p is the electronic state with the quantum number $l = 1$.
- p_0 is the statistical weight of the electronic ground state

of the atom.

P_{kr} is the critical pressure.

P_i is the statistical weight of the i th electronic state of the atom.

P is the atomic state with the quantum number $L = 1$.

$P\text{-vetv}'$ is the series of lines in a band corresponding to the transition $\Delta J = 1$.

q_l is the constant of l -splitting.

q_0/T is the coefficient in the equations used to calculate $\Phi_{kol.vr}^*$ and $S_{kol.vr}$.

Q is the statistical sum over the atomic or molecular states of the gas.

$Q\text{-vetv}'$ is the series of lines in a band corresponding to the transition $\Delta J = 0$.

Q_{vn} is the statistical sum over the intramolecular states.

$Q_{g.o}$ is the statistical sum over the states of a harmonic oscillator.

$Q_{zh.r}$ is the statistical sum over the states of a rigid rotator.

$Q_{zh.r.g.o}$ is the statistical sum over the states of a rigid rotator and harmonic oscillator.

Q_{post} is the statistical sum corresponding to translatory degrees of freedom.

Q_{el} is the statistical sum over the electronic states of an atom.

$Q_{ya.s}$ is the component of the nuclear spins in the statistical sum over the states.

$Q_{kol.vr}^{(i)}$ is the statistical sum over the rotational and vibrational states in the i th electronic state.

$Q_{\text{kol. vr}}^{(1)\infty}$	is the statistical sum over the rotational and vibrational states in the <u>i</u> th electronic state at $J_{\text{max}} = \infty$.
$Q_{\text{v, vr}}^{(1)}$	is the statistical sum over the rotational states of a molecule in the <u>i</u> th electronic state and in the vibrational state <u>v</u> .
$Q_{\text{v, vr}}^{(1)\infty}$	is the statistical sum over the rotational states of a molecule in the <u>i</u> th electronic state and in the vibrational state <u>v</u> at $J_{\text{max}} = \infty$.
r	is the index of the normal (regular) electronic state.
r	is the interatomic distance.
r_0	is the effective interatomic distance at $v = 0$.
r_e	is the equilibrium interatomic distance.
R	is the Rydberg constant.
R	is the universal gas constant.
$R\text{-vetv}'$	is the series of lines in a band, corresponding to the transition $\Delta J = +1$.
s	is the electronic state with the quantum number $l = 0$.
s	is the electron spin.
S	is the quantum number of the total electron spin of a molecule.
S	is the atomic state with the quantum number $L = 0$.
S	is the quantum number of the total electron spin of the atom.
S	is the entropy.
S	is the total electron spin of the molecule.
S	is the total electron spin of the atom.
$S\text{-vetv}'$	is the series of lines in a band corresponding to the transition $\Delta J = +2$.
S_{vn}	is the intramolecular component of S_{T}° .

$S_{g.o}$ is the component of the harmonic oscillator in S_T° .
 $S_{zh.r}$ is the component of the rigid rotator in S_T° .
 $S_{zat.vr}$ is the component of the retarded internal rotation in S_T° .
 $S_{kol.vr}$ is the vibrational-rotational component of S_T° .
 S_{post}° is the forward component of S_T° .
 $S_{sv.vr}$ is the component of the free internal rotation in S_T° .
 $S_{sm.iz} (= \Phi_{sm.iz}^*)$ is the entropy of isotope mixing.
 $S_{sim} (= \Phi_{sim}^*)$ is the correction in S_T° (and, accordingly, in Φ_T^*) allowing for the difference in the symmetry numbers of the different isotopic modifications of the molecules.
 S_{el} is the electronic component of S_T° .
 ΔS_{el} is the correction in S_T° , allowing for the existence of gas molecules in excited electronic states.
 $S_{yad} (= \Phi_{yad}^*)$ is the nuclear component of the thermodynamic functions.
 $S_{ya.s} (= \Phi_{ya.s}^*)$ is the entropy of nuclear spin.
 S_T° is the entropy of a substance in normal state at $T^\circ K$.
 S^{id} is the entropy of an ideal gas in a state different from the normal one.
 t^* is the force constant of the intermolecular potential.
 T is the temperature.
 T^* is the reduced temperature.
 T_0 is the difference between the energies of the excited and the ground state of an electron at v' and $v'' = 0$.
 T_{kr} is the critical temperature.
 T_e is the difference between the energies of the excited and the ground state of an electron in the minima of the potential curves.
 u, g are the indices of the odd and even electronic states,

respectively.

U is the intermolecular potential.

v is the vibrational quantum number.

v_{\max} is the maximum value of v in a given electronic state.

V is the volume.

V_0 is the height of the potential barrier of the retarded internal rotation.

V_0 is the volume of 1 mole of gas.

V_{kr} is the critical volume.

$V(r)$ is the potential energy of the molecule.

W is the constant of the anharmonic resonance of polyatomic molecules.

x is the ratio of the anharmonicity constant to the oscillation frequency.

x_1 is the molar fraction.

x_{1k} are the anharmonicity constants of polyatomic molecules.

X is the symbol of the electronic ground state of a diatomic molecule.

Y_{1k} are the coefficients in the Dunham equation for the vibrational-rotational energy of a diatomic molecule.

Z is the number of valence electrons in the atom.

α is the polarizability.

$\alpha_1, \alpha_2, \dots$ are the coefficients of an expansion of B_v into a power series of $v + 1/2$ (interaction constants of vibration and rotation).

$\alpha_1^A, \alpha_1^B, \alpha_1^C$ are the interaction constants of rotation and vibration of polyatomic molecules.

β_1, β_2 are the coefficients with v and v^2 in the expansion of $1/B_v$ into a power series of v .

β_1, β_2, \dots are the coefficients of an expansion of D_v into a power series of $v + 1/2$.

γ is the constant of spin-orbit interaction in the Hund case b.

Δ is the electronic state of a molecule with $\Lambda = 2$.

Δ_M is the correction in the expression for $Q_{kol.vr}$ of diatomic molecules, allowing for the splitting of rotational levels in multiplet electronic states.

ϵ is the force constant of the intermolecular potential.

ϵ_0 is the zero energy.

$\epsilon_{1,vn}$ is the energy of the ith state of the molecule connected with intramolecular degrees of freedom.

$\epsilon_{1,post}$ is the energy of the ith state of the molecule connected with translatory degrees of freedom.

θ_1 is the characteristic temperature of the ith vibration.

θ_D is the characteristic Debye temperature.

Λ is the quantum number of the projection of L on the internuclear axis.

μ is the dipole moment of the molecule.

μ is the constant in the equation for the rotational levels of diatomic molecules in the $^3\Sigma$ state.

μ is the reduced mass of a diatomic molecule.

ν is the fundamental frequency of a diatomic molecule.

ν is the mole number of a substance.

$\Delta\nu$ is the change in the mole number of a gas in a reaction.

ν_0 is the wave number of the band origin.

ν_{00} is the wave number of the origin of a band system.

ν_n is the fundamental frequency of the nth vibration of a polyatomic molecule.

- ν_1 is the energy of the 1th state of an atom or molecule in inverse centimeters.
- $\delta\nu_n$ is the interval between unifiable electronic states of atoms.
- $\nu_n(2)$ is the fundamental frequency of a doubly degenerate vibration.
- $\nu_n(3)$ is the fundamental frequency of a threefold degenerate vibration.
- Π is the electronic state of a molecule with $\Lambda = 1$.
- ρ_1, ρ_2 are constants with T and T^2 in the formulas used to calculate the correction to the centrifugal stretching in the thermodynamic functions of polyatomic gases.
- ρ^2 is the ratio of the reduced masses of the isotopic molecules.
- σ is the force constant of the intermolecular potential.
- σ is the symmetry number of a molecule.
- σ_1 is the symmetry number of the gyroscope relative to the molecular frame.
- Σ is the quantum number of the component of S along the axis of the molecule.
- Σ is the electronic state of the molecule with $\Lambda = 0$.
- Σ is the correction to the anharmonicity of vibrations, the interaction of rotation and vibrations and the centrifugal stretching in the expression for $Q_{\text{kol.vr}}$ of perfect gases.
- φ is the deflection angle of the gyroscope relative to the frame for internal rotation.
- Φ is the electronic state of a molecule with $\Lambda = 3$.
- Φ^* is the reduced thermodynamic (isobaric) potential.

- Φ_{vn}^* is the intramolecular component of Φ_T^* .
- $\Phi_{g.o}^*$ is the component of the harmonic oscillator in Φ_T^* .
- $\Phi_{zh.r}^*$ is the component of the rigid rotator in Φ_T^* .
- $\Phi_{zat.vr}^*$ is the component of the retarded internal rotation in Φ_T^* .
- $\Phi_{kol.vr}^*$ is the vibrational-rotational component of Φ_T^* .
- Φ_{post}^* is the forward component of Φ_T^* .
- $\Phi_{sv.vr}^*$ is the component of the free internal rotation in Φ_T^* .
- Φ_{el}^* is the electronic component in Φ_T^* .
- $\Delta\Phi_{el}^*$ is a correction in Φ_T^* allowing for the existence of gas molecules in excited electronic states.
- Φ_T^* is the reduced isobaric potential of a substance in the normal state at $T^\circ K$.
- $\omega_0, \omega_0^x, \omega_0^y, \omega_0^z, \dots$ are the vibrational constants of a diatomic molecule in the equation for $G_0(v)$.
- ω_e is the frequency of oscillation of a diatomic molecule.
- $\omega_e^x, \omega_e^y, \omega_e^z, \dots$ are the anharmonicity constants of the vibrations of diatomic molecules.
- ω_n is the frequency of the nth normal vibration of a polyatomic molecule.
- ω_n^0 is the vibrational constant of the nth vibration of a polyatomic molecule.
- Ω is the quantum number of the projection of the total angular momentum on the internuclear axis.
- $" , '$ are primes denoting the lower and upper states, respectively.
- $+ , -$ are indices of Σ -states with symmetric and antisymmetric wave functions.

[Footnotes]

- 2102 пост = post = postupatel'nyy = translatory
- 2103 г.о = g.o = garmonicheskiy ostsillyator = harmonic oscillator
- 2104 кол.вр. = kol. vr. - kolebatel'nyy, vrashchatel'nyy =
vibrational, rotational
- 2107 пр = pr = irivedenny = reduced
- 2109 ветвь = vetv' = branch
- 2110 кр = kr = kriticheskiy = critical
- 2110 вн = vn = vnutrimolekulyarnyy = intramolecular
- 2110 ж.р = zh. r = zhestkiy rotator - rigid rotat r
- 2110 эл = el = elektronnyy - electronic
- 2110 я.с = ya. s = yadernyy spin - nuclear spin
- 2111 вр = vr = vrashchatel'nyy = rotational
- 2112 зат.вр = zat. vr = zatormozhennoye vrashcheniye = retarded
rotation
- 2112 св. вр = sv. vr = svobodnoye vrashcheniye = free rotation
- 2112 см. из = sm. iz = smesheniye, izotop = mixing, isotope
(isotope mixing entropy)
- 2112 сим = sim = simmetriya = symmetry
- 2112 яд = yad = yadernyy = nuclear
- 2112 ид = id = ideal'nyy = ideal

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- Vestn. Mosk. un-ta - Vestnik Moskovskogo universiteta (SSSR) [Herald of the Moscow University].
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- Zh. russk. fiz.-khim. ob-va - Zhurnal russkogo fiziko-khimicheskogo obshchestva (SSSR) [Journal of the Russian Physico-Chemical Society].
- Zh. strukturn. khimii - Zhurnal strukturnoy khimii (SSSR) [Journal of Structural Chemistry].
- Zh. tekhn. fiz. - Zhurnal tekhnicheskoy fiziki (SSSR) [Journal of Technical Physics].
- Zh. fiz. khimii - Zhurnal fizicheskoy khimii (SSSR) [Journal of Physical Chemistry].
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- Arch. Eisenhuettenwesen - Archiv fuer das Eisenhuettenwesen (FRG) [Archive of Iron Metallurgy].
- Arkiv. mat. astr. fys. A. - Arkiv. mat. astr. fys. A. - Arkiv foer matematik, astronomi und fysik (Sweden) [Archive for Mathematics, Astronomy and Physics].
- Arkiv fys. - Arkiv foer fysik (Sweden) [Archive for Physics].
- Astron. J. - The Astronomical Journal (USA).
- Astrophys. J. - The Astrophysical Journal (USA).
- Astrophys. J. Suppl. Ser. I. - The Astrophysical Journal Supplement Series (USA).
- Atti Accad. liqure sci. e lettere (Pavia) - Atti della Accademia liqure di scienze e lettere (Italy) [Transactions of the Liqurian Academy of Sciences and Letters].
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- Atti Congr. intern. chim. - atti del congresso internazionale di chimica (Italy) [Proceedings of the International Congress on Chemistry].
- Austral. J. Chem. - Australian Journal of Chemistry (Australia).
- Austral. J. Phys. - Australian Journal of Physics (Australia).

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 schaftliche Klasse (GDR) [Reports on the Transactions of the
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- Boll. scient. Fac. chim. industr. Bologna - Bollettino scientifico
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 Bologna].
- Brennstoff-Waerme-Kraft - Brennstoff-Waerme-Kraft (FRG) [Fuel-Heat-
 Power].
- Brown Boveri Rev. - Brown Boveri Review (Switzerland).
- Bull. Amer. Phys. Soc. - Bulletin of the American Physical Society
 (USA).
- Bull. Chem. Soc. Japan - Bulletin of the chemical Society of Japan
 (Japan).
- Bull. Chem. Thermod. - Bulletin of Chemical Thermodynamics. Commission
 on Thermodynamics, Sub-commission on Experimental Thermo-
 chemistry, Subcommission on Experimental Thermodynamics
 (England).
- Bull. cl. sci. Acad. roy. Belgique - Bulletin de la classe des sciences
 Academie royale de Belgique (Belgium) [Bulletin of the Class
 of Sciences of the Royal Academy of Belgium].
- Bull. inst. internat. froid - Bulletin de l'Institut international du
 froid (France) [Bulletin of the International Institute for
 Refrigeration].
- Bull. Internat. Acad. Polonaise - Bulletin International de l'Academie
 Polonaise des Sciences et de Lettres (Poland) [International

Bulletin of the Polish Academy of Sciences and Letters].
Bull. Soc. chim. belges - Bulletin des Societes chimiques belges
(France) [Bulletin of the Belgian Chemical Societies].
Bull. Soc. chim. France - Bulletin de la Societe chimique de France
(France) [Bulletin of the Chemical Society of France].
Bull. Soc. roy. sci. Liege - Bulletin de la Societe royale des sciences
de Liege (Belgium) [Bulletin of the Royal Scientific Society
of Liege].
C. r. Acad. sci. - Comptes rendus hebdomadaires des seances de l'Acad-
emie des sciences (France) [Weekly Reports on the Sessions of
the Academy of Sciences].
Cahiers phys. - Cahiers de physique (France) [Physical Papers].
Canad. J. Chem. - Canadian Journal of Chemistry (Canada).
Canad. J. Phys. - Canadian Journal of Physics (Canada).
Canad. J. Res. - Canadian Journal of Research (Canada).
Canad. J. Technol. - Canadian Journal of Technology (Canada).
Chem. Ber. - Chemische Berichte (GDR) [Chemical Reports].
Chem. Engng. - Chemical Engineering (USA).
Chem. Engng. Progr. - Chemical Engineering Progress (USA).
Chem. Engng. Sci. - Chemical Engineering Science (England).
Chem.-Ingr.-techn. - Chemie-Ingenieur-Technik (FRG) [The Technology of
the Chemical Engineer].
Chem. listy - Chemické listy (Czechoslovakia) [Chemical Papers].
Chem. News - Chemical News (Japan).
Chemical Revs. - Chemical Reviews (USA).
Chemiker-Ztg. - Chemiker-Zeitung (FRG) [The Chemist's Journal].
Chemistry and Industry - Chemistry and Industry (Japan).
Collection Czechoslov. Chem. Com. - Collection of Czechoslovak Chemical
Communications (Czechoslovakia).

Combustion and Flame - Combustion and Flame (England).

Communs. Kamerlingh Onnes Lab. Univ. Leiden - Communications from the
Kamerlingh Onnes Laboratory of the University of Leiden
(Netherlands).

Communs. Phys. Lab. Univ., Leiden - Communications from the Physics
Laboratory of the University of Leiden (Netherlands).

Compt. rend. Acad. Bulgare sci. - Comptes rendues de l'Academie Bulgare
des sciences (Bulgaria) [Reports of the Bulgarian Academy of
Sciences].

Current Sci. - Current Science (India).

Denkschr. Oesterr. Akad. Wiss. - Denkschriften der Oesterreichischen
Akademie der Wissenschaften (Austria) [Memorials of the Aus-
trian Academy of Sciences].

Disc. Faraday Soc. - Discussions of the Faraday Society (England).

Dissert. Abstr. - Dissertation Abstracts (USA).

Euclides - Euclides (Spain).

Forsch. Geb Ingenieurwesens - Forschung auf dem Gebiet des Ingenieur-
wesens (FRG) [Investigations in the Field of Engineering].

Gazz. chim. ital. - Gazzetta chimica italiana (Italy) [Italian Chemical
Journal].

Glastechn. Ber. - Glastechnische Berichte (FRG) [Glass Engineering Re-
ports].

Heat., Piping and Air Condit. - Heating, Piping and Air Conditioning
(USA).

Helv. chim. acta - Helvetica chimica acta (Switzerland).

Helv. phys. acta - Helvetica physica acta (Switzerland).

Illinois Inst. Technol., Research Bulletin - Illinois Institute of
Technology, Research Bulletin (USA).

Indian J. Phys. - Indian Journal of Physics and Proceedings of the

Indian Association for the Cultivation of Science (India).
Ingr.-Arch. - Ingenieur-Archiv (FRG) [Archive for Engineering].
Industr. and Engng. Chem. - Industrial and Engineering Chemistry (USA).
J. Amer. Ceram. Soc. - Journal of the American Ceramic Society (USA).
J. Amer. Chem. Soc. - Journal of the American Chemical Society (USA).
J. Appl. Chem. - The Journal of Applied Chemistry (England).
J. Appl. Phys. - Journal of Applied Physics (USA).
J. Atmos. and Terr. Phys. - Journal of Atmospheric and Terrestrial
Physics (England).
J. Chem. Educ. - Journal of Chemical Education (USA).
J. Chem. Phys. - The Journal of Chemical Physics (USA).
J. Chem. Soc. - Journal of the Chemical Society (England).
J. Chem. Soc. Japan - Journal of the Chemical Society Japan (Japan).
J. chim. phys. et phys.-chim. biol. - Journal de chimie physique et de
physico-chimie biologique (France) [Journal of Chemical Phys-
ics and Biological Physicochemistry].
J. Chinese Chem. Soc. - Journal of the Chinese Chemical Society (China).
Electronics - Journal of Electronics (England).
J. Franklin Inst. - Journal of the Franklin Institute (USA).
J. Inorg. and Nucl. Chem. - Journal of Inorganic and Nuclear Chemistry
(England).
Iron and Steel Inst. - Journal of the Iron and Steel Institute (Eng-
land).
Japan Ceram. Assoc. - Journal of the Japanese Ceramic Association
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J. Less-Common Metals - Journal of the Less-Common Metals. Internation-
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J. Math. and Phys. - Journal of Mathematics and Physics (USA).
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J. Molecul. Spectrosc. -- Journal of Molecular Spectroscopy (USA).
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J. Phys. Chem. - The Journal of Physical Chemistry (USA).
J. Phys. and Chem. Solids - Journal of the Physics and chemistry of
Solids (England).
J. Phys. and Colloid Chem. - Journal of Physical and Colloid Chemistry
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J. phys. et radium - Journal de physique et le radium (France) [Journal
of Physics and Radium].
J. Phys. Soc. Japan - Journal of the Physical Society of Japan (Japan).
J. prakt. Chem. - Journal fuer praktische Chemie (GDR) [Journal for
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Res. NBS - Journal of Research of the National Bureau of Standards
(USA).
J. Sci. Hiroshima Univ. - Journal of Science of the Hiroshima Univer-
sity. Series A., Mathematics, Physics, Chemistry (Japan).
J. Scient. Res. Banaras Hindu Univ. - The Journal of Scientific Re-
search of the Banaras Hindu University (India).
Scient. Res. Inst. - Journal of the Scientific Research Institute
(Japan).
Soc. Chem. Ind. (London) - Journal of the Society of chemical industry
(England).
J. South African Chem. Inst. - Journal of the South African Chemical
Institute (Union of South Africa).
Japan. J. Phys. - The Japanese Journal of Physics (Japan).
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skabernes selskab. Det matematisk-fysiske meddelelser (Den-
mark) [Royal Danish Scientific Society. Mathematical and

Physical Communications].

Kgl. norske Vid. selskabs skr. - Det Kongelige norske videnskabers
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Scientific Society].

Kolloid-Z. - Kolloid-Zeitschrift (FRG) [Colloid Journal].

Low Temperat. Sci. A. - Low Temperature Science A. (Japan).

Magyar fiz. folyoirat - Magyar fizikai folyoirat (Hungary) [Hungarian
Physical Journal].

Mech. Engng. - Mechanical Engineering (USA).

Mem. Coll. Sci. Univ. Kyoto A. - Memoirs of the College of Science.
University of Kyoto. Series A., Mathematics (Japan).

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ence, Agriculture Taihoku University (Japan).

Mem. Soc. roy. sci. Liege - Memoires de la Societe royale des sciences
de Liege (Belgium) [Communications of the Royal Scientific
Society of Liege].

Metall - Metall (Zeitschrift fuer Technik, Industrie und Handel) (FRG)
[Metal (Journal for Technology, Industry and Commerce)].

Metall und Erz - Metall und Erz (Germany) [Metal and Ore].

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Mitt. K.W.I. Eisenforsch. Duesseldorf - Mitteilungen des Kaiser Wilhelm
Instituts fuer Eisenforschung, Duesseldorf (Germany) [Com-
munications of the Kaiser-Wilhelm Institute for Iron Re-
search].

Monatsh. Chemie - Monatshefte fuer Chemie (Austria) [Monthly Journal on
Chemistry].

Molecular Phys. - Molecular Physics (England).

Monthly Notices Roy. Astron. Soc. - Monthly notices of the Royal As-
tronomical Society (England)

Nachr. Akad. Wiss. Goettingen. Math.-phys. Kl. - Nachrichten der Akademie der Wissenschaften zu Goettingen. Mathematisch-physikalische Klasse IIa. Mathematisch-physikalisch-chemische Abteilung (FRG) [Communications of the Academy of Sciences at Goettingen. Mathematical-Physical Department IIa. Mathematical-Physical-Chemical Department].

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Nature - Nature (England)

Naturwissenschaften - Die Naturwissenschaften (FRG) [Science].

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Nucl. Sci. Abstrs. - Nuclear Science Abstracts (USA).

Nuovo cimento - Il nuovo cimento (Italy) [The New Venture].

Petrol. Process. - Petroleum Processing Including Petrochemical Processing Section (USA).

Petrol. Refiner - Petroleum Refiner (USA).

Petroleum - Petroleum (England).

Philips Res. Repts. - Philips Research Reports (Netherlands).

Philos. Mag. - The Philosophical Magazine (England).

Philos. Trans. Roy. Soc. (L) A. - Philosophical Transactions of the Royal Society of London. Series A. (England).

- Phys. of Fluids - Physics of Fluids (USA).
- Phys. Rev. - The Physical Review (USA).
- Phys. Z. - Physikalische Zeitschrift (Germany) [Physical Journal].
- Proc. Acad. Sci. Amsterdam - Proceedings of the Royal Academy of Sciences of Amsterdam (Netherlands).
- Proc. Amer. Acad. Arts and Sci. - Proceedings of the American Academy of Arts and Sciences (USA).
- Proc. Chem. Soc. - Proceedings of the Chemical Society (England).
- Proc. Indian Acad. Sci. A. - Proceedings of the Indian Academy of Sciences. Section A. (India).
- Proc. Nat. Acad. Sci. U.S.A. - Proceedings of the National Academy of Sciences of the United States of America (USA).
- Proc. Nat. Inst. Sci. India - Proceedings of the National Institute of Sciences of India (India).
- Proc. Phys. Soc. A. - The Proceedings of the Physical Society. Section A. (England).
- Proc. Phys. Soc. Japan - Proceedings of the Physical Society, Japan (Japan).
- Proc. Roy. Acad. Sci. Amst. - Proceedings Koninklijke Academie van Wetenschappen Amsterdam (Netherlands).
- Proc. Roy. Irish Acad. A. - Proceedings of the Royal Irish Academy. Section A., Mathematical, Astronomical and Physical Sciences (England).
- Proc. Roy. Soc. A. - Proceedings of the Royal Society. Series A. Mathematical and Physical Sciences (England).
- Proc. Univ. Durham. Philos. Soc. - Proceedings of the University of Durham Philosophical Society (England).
- Progr. Theoret. Phys. - Progress of Theoretical Physics (Japan).
- Publs. Astron. Soc. Pacif. - Publications of the Astronomical Society

of the Pacific (USA).

Publs. Domin. Astrophys. Observ. - Publications of the Dominion Astrophysical Observatory (Canada).

Quart. Rev. - Quarterly Reviews (Chemical Society) (England).

RCA Rev. - RCA Review Technical Journal (England).

Recueil trav. chim. - Recueil des travaux chimiques des Pays-Bas (Netherlands) [Collection of Chemical Works of the Netherlands].

Refrig. Engng. - Refrigerating Engineering (USA).

Rend. Acad. Linc. - Atti della Accademia Nazionale dei Lincei. Rendiconti (Italy) [Transactions of the National Lincei Academy. Reports].

Research - Research. Science and its Application in Industry (England).

Rev. acad. cienc. exact. fis. - quim. y nat. Zaragoza. - Revista de la academia de ciencias exactas fisicoquimicas y naturales de Zaragoza (Spain) [Review of the Academy of Exact Physicochemical and Natural Sciences of Zaragoza].

Rev. Inst. franc. petrole - Revue de l'Institut francais du petrole et Annales des combustibles liquides (France) [Review of the French Petroleum Institute and Annals for Combustible Liquids].

Rev. optique - Revue d'optique theorique et instrumentale (France) [Review of Theoretical and Instrumental Optics].

Rev. Phys. Chem. Japan - The Review of Physical Chemistry of Japan (Japan).

Rev. Scient. Instrum. - The review of Scientific Instruments (USA).

Rev. univ. mines - Revue universelle des mines, de la metallurgie, des travaux publics, des sciences et des arts appliquees a l'industrie (Belgium) [General Review of Mines, Metallurgy, Public Works, Science and Art Applied to Industry].

Revs. Mod. Phys. - Reviews of Modern Physics (USA).

Revs. Pure and Appl. Chem. - Reviews of Pure and Applied Chemistry
(Australia).

Ricerca sci. - Ricerca scientifica ed il progresso tecnico nell' ec-
onomia nazionale (Italy) [Scientific Research and Technical
Progress in Political Economy].

Roczn. chem. - Roczniki chemii (Poland) [Chemical Dictionaries].

Schweiz. Arch. angew. Wiss. und Techn. - Schweizer Archiv fuer ange-
wandte Wissenschaft und Technik (Switzerland) [Swiss Archive
for Applied Science and Technology].

Sci. Abstr. A. - Science Abstracts. Section A. Physics Abstracts
(England).

Sci. of Light - Science of Light (Japan).

Sci. Papers Inst. Phys. Chem. Res. Tokyo - Scientific Papers of the
Institute of Physico-Chemistry Research, Tokyo (Japan).

Sci. Repts. Tohoku Univ. - The Science Reports of the Tohoku University.
First Series-Physics, Chemistry, Astronomy (Japan).

Science - Science (USA).

Science and Culture - Science and Culture (India).

Sitzber. kgl. preuss. Akad. Wiss. - Sitzungsberichte der koeniglichen
preussischen Akademie der Wissenschaften (Germany) [Proceed-
ings of the Royal Prussian Academy of Sciences].

Sitzungsber. Dtsch. Akad. Wiss. Berlin. Kl. Math. und allgem. Natur-
wiss - Sitzungsberichte der Deutschen Akademie der Wissen-
schaften zu Berlin. Klasse fuer Mathematik und Allgemeine
Naturwissenschaften (GDR) [Proceedings of the German Academy
of Sciences in Berlin. Department of Mathematics and General
Science].

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Soc. Sci. Fennica, Commentationes Phys. Math. - Societas Scientiarum Fennica Commentationes physico-mathematicae (Finland) [Finnish Scientific Society, Physical and Mathematical Reports].

South African J. Sci. - South African Journal of Science (Union of South Africa).

Spectrochim. acta - Spectrochimica acta (England).

Studii si cercetari stiint ser. stiinte chim. - Studii si cercetari stiinte Serie stiinte chimie (Roumania) [Scientific Studies and Investigations, Series on Chemistry].

Suomen kem. - Suomen kemistilehti (Finland) [Finnish Chemical Journal].

Thermochem. Bull. - Thermochemical Bulletin (England).

Termotecnica - La termotecnica (Italy) [Heat Engineering].

Tetrahedron - Tetrahedron (England).

Tidsskr. kjemi, bergves. og metallurgi - Tidsskrift for kjemi, bergvesen og metallurgi (Norway) [Journal for Chemistry, Industrial Mining and Metallurgy].

Tonind.-Ztg. - Tonindustrie-Zeitung und Keramische Rundschau (FRG) [Journal for Clay Industry and Ceramical Review].

Trans. Amer. Inst. Chem. Engng. - Transactions of the American Institute of Chemical Engineers (USA).

Trans. ASME - Transactions of the ASME (USA).

Trans. Amer. Soc. Metals - Transactions of American Society for Metals (USA).

Trans. Electrochem. Soc. - Transactions of the Electrochemical Society
(England).

Trans. Faraday Soc. - Transactions of the Faraday Society (England).

Trans. Roy. Soc. Canada. Sec. III - Transactions of the Royal Society
of Canada. Section III. Chemical, Mathematical and Physical
Sciences (Canada).

Trans. Roy. Soc. (Dublin) - Transactions of the Royal Society (Ireland).

Trans. Soc. Mech. Engng. Japan - Transaction of the Japan Society of
Mechanical Engineers (Japan).

Verslag Akad. Wetenschappen Amsterdam - Verslag koninlijke mederlandse
Akademie van Wetenschappen (Netherlands) [Proceedings of the
Royal Dutch Academy of Sciences].

Virginia J. Sci. - The Virginia Journal of Science (USA).

Wien Ber. - Wiener Berichte (Austria) [Vienna Reports].

Wiss. Natuurk. Tydschr. - Naturwetenschappelijk Tijdschrift (Denmark)
[Scientific Journal].

Z. angew. Chemie - Zeitschrift fuer angewandte Chemie (Germany) [Jour-
nal for Applied Chemistry].

Z. angew. Mineralogie - Zeitschrift fuer angewandte Mineralogie (Ger-
many) [Journal for Applied Mineralogy].

Z. angew. Phys. - Zeitschrift fuer angewandte Physik (FRG) [Journal
for Applied Physics].

Z. anorg. und allgem. Chem. - Zeitschrift fuer anorganische und all-
gemeine Chemie (GDR) [Journal for Inorganic and General Chem-
istry].

Elektrochem. - Zeitschrift fuer Elektrochemie. Berichte der Bunsenge-
sellschaft fuer physikalische Chemie (FRG) [Journal for Elec-
trochemistry. Reports of the Bunsen Society for Physical
Chemistry].

- Z. Erzbergbau und Metallhuettenwesen - Zeitschrift fuer Erzbergbau und Metallhuettenwesen (FRG) [Journal for Ore Mining and Metallurgical Engineering].
- Z. ges. Kaelte -- Ind. Beih. - Zeitschrift fuer die gesamte Kaelte - Industrie Beihefte zur (Germany) [Journal for the Whole Field of Refrigeration - Industrial Supplements to].
- Z. Krist. - Zeitschrift fuer Kristallographie (Germany) [Journal for Crystallography].
- Z. Naturforschung. - Zeitschrift fuer Naturforschung. Ser. a, Astrophysik, Physik und physikalische Chemie (FRG) [Journal for Natural Science. Series a, Astrophysics, Physics and Physical Chemistry].
- Z. Phys. - Zeitschrift fuer Physik (FRG) [Journal of Physics].
- Z. phys. Chem. BDR - Zeitschrift fuer physikalische Chemie (FRG) [Journal for Physical Chemistry].
- Z. phys. Chem. DDR - Zeitschrift fuer physikalische Chemie (GDR) [Journal for Physical Chemistry].
- Z. techn. Phys. - Zeitschrift fuer technische Physik (Germany) [Journal for Technical Physics].
- Z. wiss. Photogr. - Zeitschrift fuer wissenschaftliche Photographie, Photophysik und Photochemie (GDR) [Journal for Scientific Photography, Photophysics and Photochemistry].

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- of the Chemical Faculty of the Moscow State University], Moscow, 1958.
2. Akishin, P.A., Gorokhov, L.N., Nikitin, O.T., Khodeyev, Yu.S. et al., Mass-spektrometicheskoye issledovaniye sostava parov i teplot sublimatsii galogenidov shchelochnykh metallov i B_2O_3 [Mass Spectrometric Investigation of the Composition and Heat of Sublimation of Alkali Metal Halides and B_2O_3]. Otchet khim. fak-ta MGU, Moscow, 1957.
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 - 4a. Akishin, P.A., Naumov, V.A., Elektronograficheskoye issledovaniye stroyeniya molekuly Ga_2O [Electron Diffraction Investigation of Ga_2O Molecule Structure] Otchet khim. fak-ta MGU, Moscow, 1958.
 5. Akishin, P.A., Naumov, V.A., Rambidi, N.G., Spiridonov, V.P., Opisaniye elektronografa MGU i metodik issledovaniya trudnoletuchikh soyedineniy [Description of the Electron Diffraction Camera of the Moscow State University and the Methods of Investigating Compounds Not Easily Volatilized], Otchet khim.

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 9. Akishin, P.A., Spiridonov, V.P., Rambidi, N.G. et al., Opre-deleniye geometricheskikh parametrov molekul NaF , LiF , Li_2O , $MgCl_2$, $BeCl_2$, BeF_2 [Determination of the Geometrical Parameters of the NaF , LiF , Li_2O , $MgCl_2$, $BeCl_2$ and BeF_2 Molecules], Otchet khim. fak-ta MGU, Moscow, 1955.
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FORMULA INDEX FOR VOLUMES 1 AND 2.

Вещество 1	2 I том				3 II том		
	4 Молекулярные постоянные	Термодинамическая функция 5	Термодинамические величины 6	Свойства реальных газов 7	Таблица термодинамических свойств 8	Интерполяционные формулы 9	Таблица виральных коэффициентов 10
Al (т., ж.)	—	767—769	772—773, 785	—	638—639	—	—
Al (газ)	751—752	762—763	772—773, 785	1025	640—641	841, 855, 869	—
Al ⁺ (газ)	752	762—763	774, 785	—	642—643	841, 855, 869, 882	—
Al ₂ (газ)	753, 755	763	774, 785	—	644—645	841, 855, 869, 882	—
AlCl (газ)	755, 757—758	763—765	781—782, 785	—	662—663	841, 855, 869, 882	—
AlCl ₂ (газ)	759—760	766—767	782—783, 785	—	664—665	841, 855, 869, 882	—
AlCl ₃ (т., ж.)	—	768, 771	783, 785	—	666	—	—
AlCl ₃ (газ)	759, 761—762	766—767	783—785	1025	668—669	842, 855, 869, 882	—
AlF (газ)	755—757	763—765	778—780, 785	—	654—655	841, 855, 869, 882	—
AlF ₂ (газ)	759—760	766—767	780, 785	—	656—657	841, 855, 869, 882	—
AlF ₃ (т., ж.)	—	768, 770—771	780, 785	—	658	—	—
AlF ₃ (газ)	759—761	766—767	780—781, 785	1025	660—661	841, 855, 869, 882	—
AlH (газ)	754—756	763—764	778, 785	—	652—653	841, 855, 869, 882	—
AlN (т., ж.)	—	768, 771—772	784—785	—	670—671	—	—
AlN (газ)	755, 758	763	784—785	1025	672—673	842, 855, 869, 882	—
AlO (газ)	753—755	763—764	774—775, 785	—	646—647	841, 855, 869, 882	—
Al ₂ O (газ)	758—760	766	775—776, 785	—	648—649	841, 855, 869, 882	—
Al ₂ O ₃ (т., ж.)	—	768—770	776—777, 785	—	650—651	—	—
Al ₂ O ₃ (газ)	—	—	777—778, 785	1025	—	—	—
Ar (газ)	945	945	—	1006, 1020, 1021, 1023	116—117	831, 845, 859	903
B (т., ж.)	—	727—729	731—732, 748	—	596—597	—	—
B (газ)	699	718	731—733, 748	1025	598—599	840, 854, 868	—
B ₂ (газ)	699—701	718—720	733, 748	—	600—601	840, 854, 868, 881	—
BCl (газ)	700, 704—705	719—721	746—748	—	628—629	841, 855, 869, 882	—
BCl ₂ (газ)	714—715	722, 726	747—748	—	630—631	841, 855, 869, 882	—
BCl ₃ (газ)	715, 717	722, 726—727	747—748	1006, 1019, 1025	632—633	841, 855, 869, 882	916

Вещество	I том				II том		
	Молекулярные постоянные	Термодинамические функции	Термодинамические величины	Свойства реальных газов	Таблица термодинамических свойств	Интерполяционные формулы	Таблица виральных коэффициентов
BF (газ)	700,703—704	719—721	745, 748	—	620—621	841, 855, 869, 882	—
BF ₂ (газ)	714—715	722, 726	745—746, 748	—	622—623	841, 855, 869, 882	—
BF ₃ (газ)	715—717	722, 726—727	746, 748	1006, 1019, 1025	624—625	841, 855, 869, 889	916
BH (газ)	700,702—703	719—720	741—742, 748	—	612—613	841, 854, 868, 881	—
BN (т., ж.)	—	727, 730	748—749	—	634—635	—	—
BN (газ)	700,705—706	719, 721—722	748—749	—	636—637	841, 855, 869, 882	—
BO (газ)	700—702	718—720	733—734, 748	—	602—603	840, 854, 868, 881	—
BO ₂ (газ)	706—707	722—723	734, 748	—	604—605	840, 854, 868, 881	—
B ₂ O ₃ (газ)	707—709	722—724	734—736, 748	—	606—607	840, 854, 868, 881	—
B ₂ O ₃ (т., ж.)	—	727, 729—730	737—739, 748	—	608—609	—	—
B ₂ O ₃ (газ)	709—712	722—724	739—741, 748	1025	610—611	841, 854, 868, 881	—
Ba (т., ж.)	—	845, 847	855, 858	—	744—745	—	—
Ba (газ)	835—838	842—843	855, 858	1025	746—747	843, 856, 870	—
Ba ⁺ (газ)	838	842—843	855, 858	—	748—749	843, 856, 870, 883	—
BaO (т., ж.)	—	845, 847—848	855, 858	—	750—751	—	—
BaO (газ)	839, 842	843—844	855—858	1025	752—753	843, 857, 871, 883	—
Be (т., ж.)	—	797—798	801—802, 808	—	674—675	—	—
Be (газ)	787	793	801—802, 808	1025	676—677	842, 855, 869	—
BeCl (газ)	788, 790—791	794—795	806, 808	—	690—691	842, 856, 870, 883	—
BeCl ₂ (т., ж.)	—	797, 800	806—808	—	692	—	—
BeCl ₂ (газ)	792—793	796	807—808	1025	694—695	842, 856, 870, 883	—
BeF (газ)	788—790	794—795	804—805, 808	—	684—685	842, 856, 870, 883	—
BeF ₂ (т., ж.)	—	797, 799—800	805, 808	—	698	—	—
BeF ₂ (газ)	791—792	796	805—806, 808	1025	688—689	842, 856, 870, 883	—
BeH (газ)	788—789	794	803, 808	—	682—683	842, 856, 870, 883	—
BeN (газ)	788, 791	794—796	807—808	—	696—697	842, 856, 870, 883	—
BeO (т., ж.)	—	797—799	802, 808	—	678—679	—	—
BeO (газ)	787—788	793—794	802—803, 808	1025	680—681	842, 856, 870, 882	—
Br (газ)	267	271	275, 278	—	160—161	832, 846, 860	—
Br ⁻ (газ)	267—268	272	275, 278	—	162—163	832, 846, 860, 873	—
Br ₂ (ж.)	—	—	275—276, 278	—	—	—	—

Вещество	I том				II том		
	Молекулярные востановные	Термодинамические функции	Термодинамические величины	Свойства реальных газов	Таблица термодинамических свойств	Интерполяционные формулы	Таблица виральных коэффициентов
Br ₂ (газ)	268—270	272—273	276, 278	1023	164—165	832, 846, 860, 873	—
BrCl (газ)	269—271	274	277—278	—	176—177	832, 846, 860, 874	—
BrF (газ)	269—270	273—274	276—278	—	174—175	832, 846, 860, 874	—
BrO (газ)	269—270	273	276, 278	—	166—167	832, 846, 860, 873	—
C (алмаз)	—	477—481	481, 489	—	—	—	—
C (графит, ж.)	—	477—481	481, 484, 489	—	314—315	—	—
C (газ)	439—440	467	481—484, 489	1023	316—319	835, 849, 863	—
C ⁺ (газ)	440—441	468	484, 489	—	320—323	835, 849, 863, 876	—
C ₂ (газ)	441—444	468—469	484, 489	—	324—325	835, 849, 863, 876	—
C ₃ (газ)	451—453	473—474	484—485, 489	—	326—327	835, 849, 863, 876	—
CBr ₄ (газ)	493, 496	524, 528	555	1017, 1024	410—411	837, 851, 865, 878	—
CCl (газ)	608—610	623—625	633—635	—	378—379	836, 850, 864, 877	—
CClBr ₃ (газ)	497, 508—509	528, 533—534	555	1024	434—435	837, 851, 865, 878	—
CCl ₂ (газ)	613, 615—616	624—626	634—635	—	380—381	836, 850, 864, 877	—
CCl ₂ Br ₂ (газ)	510, 514	534, 539	555	1024	432—433	837, 851, 865, 878	—
CCl ₃ (газ)	619—620	624—627	634—635	—	382—383	836, 850, 864, 877	—
CCl ₃ Br (газ)	497, 508	528, 533—534	553—554	1024	430—431	837, 851, 865, 878	—
CCl ₄ (газ)	493, 495—496	524, 526—528	553—554	1017, 1024	384—385	836, 850, 864, 877	—
CF (газ)	607—608	623—624	630—631, 635	—	352—353	836, 849, 863, 877	—
CFBr ₃ (газ)	497, 508	528, 533	554	1024	422—423	837, 851, 865, 878	—
CFCBr ₂ (газ)	515, 521	540, 543	554	1024	446—447	838, 851, 865, 879	—
CFCI ₂ Br (газ)	515—521	540, 543	554	1024	444—445	837, 851, 865, 879	—
CFCI ₃ (газ)	497, 507—508	528, 532—533	552—554	1006, 1017, 1024	400—401	837, 850, 864, 878	912
CF ₂ (газ)	613—615	624—626	631—632, 635	—	354—355	836, 849, 863, 877	—
CF ₂ BrJ (газ)	515, 521	540—541	554	—	470—471	838, 852, 866, 879	—
CF ₂ Br ₂ (газ)	510, 513	534, 538	554	1024	420—421	837, 851, 865, 878	—
CF ₂ ClBr (газ)	515, 520—521	540—541	554	1024	442—443	837, 851, 865, 879	—
CF ₂ ClJ (газ)	515, 521	540—541	554	—	468—469	838, 852, 866, 879	—

Вещество	I том				II том		
	Молекулярные постоянные	Термодинамические функции	Термохимические величины	Свойства реальных газов	Таблица термодинамических свойств	Интерполяционные формулы	Таблица виральных коэффициентов
CF ₂ Cl ₂ (газ)	510, 512—513	534, 537—538	552, 554	1006, 1017, 1024	398—399	837, 850, 864, 878	911
CF ₂ J ₂ (газ)	510, 513—514	534, 539	554	1024	458—459	838, 852, 866, 879	—
CF ₂ (газ)	619—620	624—627	632—633, 635	—	356—357	836, 850, 864, 877	—
CF ₂ Br (газ)	497, 506	528, 533	551, 554	1024	418—419	837, 851, 865, 878	—
CF ₂ Cl (газ)	497, 505—508	528, 532—533	550—551, 554	1017, 1024	396—397	836, 850, 864, 878	—
CF ₂ J (газ)	497, 506—507	528—533	551—552, 554	1024	456—457	838, 852, 866, 879	—
CF ₂ (газ)	493—495	524—526	549—550, 554	1006, 1017, 1023	358—359	836, 850, 864, 877	900
CH (газ)	606—608	623—624	627, 635	—	340—341	835, 849, 863, 876	—
CHBr ₂ (газ)	497, 503—504	528, 531—532	548—549, 554	1024	416—417	837, 851, 865, 878	—
CHClBr ₂ (газ)	515, 520	540, 543	554	1024	440—441	837, 851, 865, 879	—
CHCl ₂ Br (газ)	515, 519—520	540, 543	554	1024	438—439	837, 851, 865, 878	—
CHCl ₂ (газ)	497, 502—503	528, 531	548, 554	1006, 1016, 1017, 1024	394—395	836, 850, 864, 878	911
CHF (газ)	613, 615	624—626	633, 635	—	364—365	836, 850, 864, 877	—
CHFB ₂ (газ)	515, 519	540, 542—543	554	1024	428—429	837, 851, 865, 878	—
CHFCIBr (газ)	515, 518—519	540—541	554	—	448—449	838, 851, 865, 879	—
CHFCI ₂ (газ)	515, 518	540, 542	554	1006, 1016	408—409	837, 851, 865, 878	912
CHF ₂ (газ)	620—621	624—627	633, 635	—	370—371	836, 850, 864, 877	—
CHF ₂ Br (газ)	515, 517—518	540, 542—543	554	1024	428—427	837, 851, 865, 878	—
CHF ₂ Cl (газ)	515, 517	540, 542	554	—	406—407	837, 851, 865, 878	—
CHF ₂ J (газ)	515, 518	540, 541	554	1024	462—463	838, 852, 866, 879	—
CHF ₂ (газ)	497, 501—502	528, 531	547—548, 554	1016, 1023	374—375	836, 850, 864, 877	—
CHJ ₂ (газ)	497, 504—505	528, 532	549, 554	1024	454—455	838, 852, 866, 879	—
CH ₂ (газ)	610—613	624—625	627—629, 635	—	342—343	835, 849, 863, 876	—
CH ₂ BrJ (газ)	515—516	540—541	554	—	466—467	838, 852, 866, 879	—
CH ₂ Br ₂ (газ)	510—512	534—536	554	—	414—415	837, 851, 865, 878	—
CH ₂ ClBr (газ)	515—516	540—542	554	1024	436—437	837, 851, 865, 878	—
CH ₂ ClJ (газ)	515—516	540—541	554	—	464—465	838, 852, 866, 879	—

Вещество	I том				II том		
	Молекулярные возмущения	Термодинамические функции	Термохимические величины	Свойства реальных газов	Таблица термодинами- ческих свойств	Интерпо- ляционные формулы	Таблица вирьяль- ных коэф- фициентов
CH ₂ Cl ₂ (газ)	510—511	534—535	547, 554	1016, 1024	392—393	836, 850, 864, 878	—
CH ₂ F (газ)	620—621	624—627	632, 635	—	366—367	836, 850, 864, 877	—
CH ₂ FBr (газ)	515—516	540—541	554	1024	424—425	837, 851, 865, 878	—
CH ₂ FCl (газ)	514—515	540	554	1024	404—405	837, 851, 865, 878	—
CH ₂ FJ (газ)	515—516	540—541	554	—	460—461	838, 852, 866, 879	—
CH ₂ F ₂ (газ)	509—510	534—535	546—547, 554	1023	372—373	836, 850, 864, 877	—
CH ₂ J ₂ (газ)	510, 512	534, 536—537	547, 554	1024	452—453	838, 852, 866, 879	—
CH ₂ (газ)	616—620	624—627	629—630, 635	—	344—345	835, 849, 863, 877	—
CH ₂ Br (газ)	497, 499—500	528, 550	545—546, 554	1006, 1016, 1024	412—413	837, 851, 865, 878	913
CH ₂ Cl (газ)	497—499	528—530	545, 554	1006, 1015, 1016, 1024	390—391	836, 850, 864, 877	910
CH ₂ F (газ)	496—498	528—529	545, 554	1006, 1015, 1023	368—369	836, 850, 864, 877	910
CH ₂ J (газ)	497, 500—501	528, 530	546, 554	1006, 1016, 1024	450—451	838, 851, 865, 879	913
CH ₄ (газ)	493—494	524—525	544—545, 554	1006, 1014, 1015, 1023	346—347	835, 849, 863, 877	909
CN (газ)	637—639	649—651	657—660	—	478—479	838, 852, 866, 879	—
CO (газ)	444—450	469—470	485, 489	1006, 1013, 1023	328—331	835, 849, 863, 876	908
CO ⁺ (газ)	450—451	471	485, 489	—	332—335	835, 849, 863, 876	—
COS (газ)	642—643	652	656—657, 660	1024	476—477	838, 852, 866, 879	—
CO ₂ (газ)	453—456	471—474	486, 489	1006, 1013, 1014, 1023	336—337	835, 849, 863, 876	908
CP (газ)	637, 639—640	649, 651	660	—	484—485	838, 852, 866, 879	—
CS (газ)	636—638	649—650	654—655, 660	—	472—473	838, 852, 866, 879	—
CS ₂ (газ)	640—642	651—652	655—656, 660	1014, 1024	474—475	838, 852, 866, 879	—
C ₂ Cl ₄ (газ)	561, 564—565	581, 584	593—594	—	506—507	839, 853, 867, 880	—
C ₂ F (газ)	621—622	624, 627	633, 635	—	492—493	838, 852, 866, 880	—
C ₂ FCl ₂ (газ)	572, 580—581	586, 588	593—594	—	518—519	839, 853, 867, 880	—
C ₂ F ₂ (газ)	566—601	604	605	—	494—495	839, 852, 866, 880	—
C ₂ F ₂ Cl ₂ (газ)	566, 570—571	584—586	593—594	—	516—517	839, 853, 867, 880	—
C ₂ F ₂ Cl (газ)	572, 579—580	586, 588	593—594	1018, 1024	514—515	839, 853, 867, 880	—

Вещество	I том				II том		
	Молекулярные постоянные	Термодинамические функции	Термохимические величины	Свойства реальных газов	Таблица термодинамических свойств	Интерполяционные формулы	Таблица виральных коэффициентов
C ₂ F ₆ (газ)	561, 563—564	581, 583—584	592—594	1018, 1024	496—497	839, 852, 866, 880	—
C ₂ F ₄ (газ)	621—622	624, 625, 627	630, 635	—	496—497	839, 852, 866, 879	—
C ₂ HCl ₂ (газ)	572, 578—579	568, 588	591—592, 594	1018	512—513	839, 853, 867, 880	—
C ₂ HF (газ)	590—601	604	605	—	498—499	839, 852, 866, 880	—
C ₂ HFCl ₂ (газ)	572, 578	586—588	591, 594	—	524—525	839, 853, 867, 880	—
C ₂ HF ₂ Cl (газ)	572, 577—578	586—587	591, 594	1018, 1024	522—523	839, 853, 867, 880	—
C ₂ HF ₂ (газ)	572, 577	586—587	591, 594	—	504—505	839, 853, 867, 880	—
C ₂ H ₂ (газ)	595—598	601—604	604—605	1006, 1018, 1019, 1024	488—489	838, 852, 866, 880	914
C ₂ H ₂ Cl ₂ (газ)	566, 568	584—586	590—591, 594	1018	510—511	839, 853, 867, 880	—
C ₂ H ₂ FCI (газ)	572, 576	586—587	590, 594	—	520—521	839, 853, 867, 880	—
C ₂ H ₂ F ₂ (газ)	566—568	585	590, 594	1018, 1024	502—503	839, 853, 867, 880	—
C ₂ H ₂ Cl (газ)	572, 575—576	586—587	589, 594	—	508—509	839, 853, 867, 880	—
C ₂ H ₂ F (газ)	572—574	586	589, 594	1018	500—501	839, 853, 867, 880	—
C ₂ H ₄ (газ)	557—563	581—583	588—589, 594	1006, 1018, 1024	490—491	838, 852, 866, 880	914
C ₂ N ₂ (газ)	648—649	652—653	659—660	1006, 1019, 1024	526—527	839, 853, 867, 880	915
C ₂ O ₂ (газ)	456—458	474—475	486, 489	—	338—339	835, 849, 863, 876	—
Ca (т., ж.)	—	844—845	848, 858	—	724—725	—	—
Ca (газ)	830—832	842—843	848, 858	1025	726—727	842, 856, 870	—
Ca ⁺ (газ)	832—833	842—843	849, 858	—	728—729	842, 856, 870, 883	—
CaO (т., ж.)	—	845—846	849, 858	—	730—731	—	—
CaO (газ)	839—841	843—844	849—851, 858	1025	732—733	842, 856, 870, 883	—
Cl (газ)	249	258—260	263, 266	—	138—139	832, 845, 859	—
Cl ⁻ (газ)	249—250	259—260	263—264, 266	—	140—141	832, 845, 859, 873	—
Cl ₂ (газ)	250—253	259—260	264, 266	1009, 1023	142—143	832, 846, 860, 873	—
ClCO (газ)	461—462	474—476	488—489	—	386—387	836, 850, 864, 877	—
ClF (газ)	253—255	260—261	265—266	—	158—159	832, 846, 860, 873	—
ClO (газ)	253—254	260—262	264—266	—	144—145	832, 846, 860, 873	—
ClO ₂ (газ)	255—257	262	264—266	—	146—147	832, 846, 860, 873	—

Вещество	I том				II том		
	Молекулярные постоянные	Термодинамические функции	Термодинамические величины	Свойства реальных газов	Таблица термодинамических свойств	Интерполяционные формулы	Таблица виральных коэффициентов
Cl ₂ CO (газ)	459, 465—466	474, 476—477	488—489	1024	388—389	836, 850, 864, 877	—
Cl ₂ O (газ)	257—258	262—263	265—266	—	146—149	832, 846, 860, 873	—
Cs (т., ж.)	—	903, 906—907	914	—	818	—	—
Cs (газ)	895—896	900—901	914	1025	820—821	843, 857, 871	—
Cs ⁺ (газ)	896—897	900—901	914	—	822—823	843, 857, 871, 884	—
D (газ)	182	190—191	193, 195	—	80—81	830, 844, 858	—
D ₂ (газ)	186—188	190, 192—193	195	1007, 1023	82—83	830, 844, 858, 872	—
DBr (газ)	296, 299	301, 303—304	307—308	—	170—171	832, 846, 860, 874	—
DCl (газ)	296—298	301, 303	308, 308	—	152—153	832, 846, 860, 873	—
DF (газ)	295—296	301—302	306, 308	—	134—135	831, 845, 859, 873	—
DJ (газ)	296, 300	301, 304	307—308	—	188—189	833, 846, 860, 874	—
DT (газ)	188—189	190, 193	195	1023	102—103	831, 845, 859, 872	—
DTO (газ)	205—206	224—225	231, 237	—	110—111	831, 845, 859, 873	—
D ₂ O (газ)	204—205	224—225	231, 237	1023	88—89	831, 844, 858, 872	—
D ₂ O ₂ (газ)	208—210	226—227	232, 237	—	92—93	831, 844, 858, 872	—
F (газ)	238	241—242	244, 248	—	122—123	831, 845, 859	—
F ⁻ (газ)	238	242	244—245, 248	—	124—125	831, 845, 859, 873	—
F ₂ (газ)	238—240	242—243	245—248	1006, 1009, 1023	126—127	831, 845, 859, 873	904
FBO (газ)	707—708	722—724	746, 748	—	626—627	841, 855, 869, 882	—
FCN (газ)	645—647	654	659—660	—	482—483	838, 852, 866, 879	—
FCO (газ)	461—462	474—476	487, 489	—	360—361	836, 850, 864, 877	—
FCICO (газ)	462, 466	474, 476—477	489	—	402—403	837, 850, 864, 878	—
FNO (газ)	371, 373	388	400—401	—	272—273	834, 848, 862, 875	—
FO (газ)	240	243—244	247—248	—	128—129	831, 845, 859, 873	—
F ₂ CO (газ)	462—463	474, 476—477	488—489	—	362—363	836, 850, 864, 877	—
F ₂ O (газ)	241	243—244	247—248	1023	130—131	831, 845, 859, 873	—
H (газ)	—	189—190	193, 195	—	44—47	830, 844, 858	—
H ⁺ (газ)	181—182	190—191	—	—	78—79	830, 844, 858	—

Вещество	I том				II том		
	Молекулярные состояния	Термодинамические функции	Термодинамические величины	Свойства реальных газов	Таблица термодинамических свойств	Интерполяционные формулы	Таблица виральных коэффициентов
H ⁺ (газ)	182	190—191	194—195	—	48—51	830, 844, 858, 872	—
H ⁻ (газ)	182	190—191	194—195	—	52—55	830, 844, 858, 872	—
H ₂ (газ)	182—184	190—192	194—195	1006, 1007, 1023	56—59	830, 844, 858, 872	901
HBO (газ)	707—708	722—724	742, 748	—	614—615	841, 854, 868, 882	—
HBO ₂ (газ)	712	722, 725	742—744, 748	—	616—617	841, 855, 869, 882	—
HBr (газ)	296, 298—299	301, 303	306—308	1009, 1023	168—169	832, 846, 860, 874	—
HCN (газ)	643—645	653—654	659—660	1024	480—481	838, 852, 866, 879	—
HCO (газ)	458—461	474—476	486—487, 489	—	348—349	835, 849, 863, 877	—
HCl (газ)	295—296	301—303	306, 308	1009, 1023	150—151	832, 846, 860, 873	—
H ¹ D (газ)	185—186	190, 192—193	195	1023	84—85	830, 844, 858, 872	—
H ¹ DO (газ)	202—204	224—225	231, 237	—	90—91	831, 844, 858, 872	—
H ¹ DO ₂ (газ)	208, 210—211	226—227	232, 237	—	94—95	831, 845, 859, 872	—
HF (газ)	293—294, 296	301—302	305—306, 308	1009, 1023	132—133	831, 845, 859, 873	—
HFCO (газ)	462—465	474, 476—477	488—489	—	376—377	836, 850, 864, 877	—
HJ (газ)	296, 299—300	301, 304	307—308	1010, 1023	186—187	833, 846, 860, 874	—
HNO (газ)	371, 373	387—388	396—400	—	264—265	834, 848, 862, 875	—
HOCl (газ)	258	262—263	265—266	—	156—157	832, 846, 860, 873	—
HO ₂ (газ)	211—212	227	233, 237	—	70—71	830, 844, 858, 872	—
H ¹ T (газ)	188—189	190, 193	195	—	100—101	831, 845, 859, 872	—
H ¹ TO (газ)	205—206	224—225	231, 237	—	108—109	831, 845, 859, 873	—
H ₂ CO (газ)	459, 461—463	474, 476	487, 489	—	350—351	836, 849, 862, 877	—
H ₂ O (газ)	196—202	220—223	230—231, 237	1006, 1008, 1009, 1023	72—75	830, 844, 858, 872	901
H ₂ O ₂ (газ)	206—209	225—227	232, 237	1023	76—77	830, 844, 859, 872	—
H ₂ S (газ)	321—324	335—337	341—343	1010	212—213	833, 847, 861, 874	—
H ₂ BO ₂ (газ)	712—714	722, 725—726	744, 748	—	618—619	841, 853, 869, 882	—
He (газ)	945	945	—	1006, 1019, 1023	112—113	831, 845, 859	902
Hg (газ)	923—924	929—930	943—944	1019, 1025	582—583	840, 854, 868	—

Вещество	I том				II том		
	Молекулярные постоянные	Термодинамические функции	Термодинамические величины	Свойства реальных газов	Таблица термодинамических свойств	Интерполяционные формулы	Таблица виральных коэффициентов
HgF (газ)	926—927	930—932	943—944	—	584—585	840, 854, 868, 881	—
J (газ)	27 ¹⁾	284	288, 291	—	178—179	832, 846, 860	—
J ⁻ (газ)	270	284—285	288, 291	—	180—181	832, 846, 860, 874	—
J ₂ (газ)	279—282	285—286	288—289, 291	1009, 1023	182—183	832, 846, 860, 874	—
JBr (газ)	281, 283—284	285, 287	290—291	—	196—197	833, 847, 861, 874	—
JCl (газ)	261—283	285, 287	290—291	—	194—195	833, 847, 861, 874	—
JF (газ)	281—282	285—286	289—291	—	192—193	833, 847, 861, 874	—
JO (газ)	281—282	285—286	289, 291	—	184—185	833, 846, 860, 874	—
K (т., ж.)	—	903, 905	911, 914	—	798	—	—
K (газ)	893—894	900—901	911, 914	1025	800—801	843, 857, 871	—
K ⁺ (газ)	896—897	900—901	911, 914	—	802—803	843, 857, 871, 884	—
KCl (т., ж.)	—	903, 906	912—914	—	808—809	—	—
KCl (газ)	898—899	901—902	913—914	1025	810—811	843, 857, 871, 884	—
KF (т., ж.)	—	903, 906	911, 914	—	804—805	—	—
KF (газ)	897—899	901—902	911—912, 914	1025	806—807	843, 857, 871, 884	—
Kr (газ)	945	945	—	1006, 1021	118—119	831, 845, 859	903
Li (т., ж.)	—	874—876	879—880, 890	—	754	—	—
Li (газ)	859—860	869	879—880, 890	1025	756—757	843, 857, 871	—
Li ⁺ (газ)	860	869	880, 890	—	758—759	843, 857, 871, 883	—
Li ₂ (газ)	860—862	860—871	880, 890	—	760—761	843, 857, 871, 883	—
LiCl (т., ж.)	—	875, 878	888, 890	—	780—781	—	—
LiCl (газ)	862, 866—867	871, 873	889—890	1025	782—783	843, 857, 871, 884	—
LIF (т., ж.)	—	875, 877—878	885—886, 890	—	776—777	—	—
LIF (газ)	862—866	871, 873	886—888, 890	1025	778—779	843, 857, 871, 884	—
LiH (т., ж.)	—	875, 877	883, 890	—	768	—	—
LiH (газ)	862—863	870—871	883, 890	1025	770—771	843, 857, 871, 884	—
LiO (газ)	862, 867—868	871—872	880—881, 890	—	762—763	843, 857, 871, 883	—
LiOH (т., ж.)	—	875, 877	883—884, 890	—	772—773	—	—
LiOH (газ)	868	873—874	884—885, 890	1025	774—775	843, 857, 871, 884	—
Li ₂ O (т., ж.)	—	875—877	881, 890	—	764—765	—	—
Li ₂ O (газ)	868	873—874	881—883, 890	1025	766—767	843, 857, 871, 883	—

Вещество	I том				II том		
	Молекулярные постоянные	Термодинамические функции	Термодинамические величины	Свойства реальных газов	Таблица термодинамических свойств	Интерполяционные формулы	Таблица вициальных коэффициентов
Mg (т., ж.)	—	820—821	823—824, 829	—	698	—	—
Mg (газ)	809—810	816	823—824, 829	1025	700—701	842, 856, 870	—
Mg ⁺ (газ)	810	816	824, 829	—	702—703	842, 856, 870, 883	—
MgCl (газ)	812—814	817—818	827—829	—	716—717	842, 856, 870, 883	—
MgCl ₂ (т., ж.)	—	820, 822—823	828—829	—	718—719	—	—
MgCl ₂ (газ)	815—816	819	828—829	1025	720—721	842, 856, 870, 883	—
MgF (газ)	812—813	817—818	825—827, 829	—	710—711	842, 856, 870, 883	—
MgF ₂ (т., ж.)	—	820, 822	827, 829	—	712—713	—	—
MgF ₂ (газ)	814—815	819	827, 829	1025	714—715	842, 856, 870, 883	—
MgH (газ)	811—813	817—818	825, 829	—	708—709	842, 856, 870, 883	—
MgN (газ)	812, 814	817—819	828—829	—	722—723	842, 856, 870, 883	—
MgO (т., ж.)	—	820—822	824, 829	—	704—705	—	—
MgO (газ)	810—812	816—817	824—825, 829	1025	706—707	842, 856, 870, 883	—
N (газ)	346—348	379	393, 400	—	226—229	833, 847, 861	—
N ⁺ (газ)	348—349	379—380	393—394, 400	—	230—233	833, 847, 861, 875	—
N ₂ (газ)	349—353	380—382	394—395, 400	1006, 1010, 1011, 1023	234—237	833, 847, 861, 875	906
N ₂ ⁺ (газ)	353—358	382—383	395, 400	—	238—241	834, 847, 861, 875	—
N ₂ (газ)	366—367	387—388	395, 400	—	242—243	834, 847, 861, 875	—
NF (газ)	365	386—387	399—400	—	266—267	834, 848, 862, 875	—
NF ₂ (газ)	372	388	399—400	—	268—269	834, 848, 862, 875	—
NF ₃ (газ)	376	388—389	400	1013, 1023	270—271	834, 848, 862, 875	—
NH (газ)	363—365	385—386	397—398, 400	—	256—257	834, 848, 862, 875	—
NH ₂ (газ)	370—372	387—388	398, 400	—	258—259	834, 848, 862, 875	—
NH ₃ (газ)	373—376	390—392	398, 400	1006, 1012, 1013, 1023	260—261	834, 848, 862, 875	907
NO (газ)	357—362	383—384	395—396, 400	1006, 1011, 1023	244—247	834, 847, 861, 875	906
NO ⁺ (газ)	362—363	385	396—397, 400	—	248—251	834, 848, 862, 875	—
NO ₂ (газ)	367—369	388—389	397, 400	1023	252—253	834, 847, 862, 875	—
NS (газ)	365—366	386—387	400—401	—	274—275	834, 848, 862, 875	—
N ₂ H ₄ (газ)	376—378	392—393	398, 400	1023	262—263	834, 848, 862, 875	—

Вещество	I том				II том		
	Молекулярные постоянные	Термодинамические функции	Термодинамические величины	Свойства реальных газов	Таблица термодинамических свойств	Интерполяционные формулы	Таблица виральных коэффициентов
N ₂ O (газ)	369—370	388, 390	397, 400	1006, 1011, 1023	254—255	834, 848, 862, 875	917
Na (т., ж.)	—	903—904	907, 914	—	784	—	—
Na (газ)	892—893	900—901	907, 914	1025	786—787	843, 857, 871	—
Na ⁺ (газ)	896—897	900—901	907, 914	—	788—789	843, 857, 871, 884	—
NaCl (т., ж.)	—	903, 905	909, 914	—	794—795	—	—
NaCl (газ)	898—899	901—902	910—911, 914	1025	796—797	843, 857, 871, 884	—
NaF (т., ж.)	—	903—904	907—908, 914	—	790—791	—	—
NaF (газ)	897—899	901—902	908—909, 914	1025	792—793	843, 857, 871, 884	—
Ne (газ)	945	945	—	1006, 1019, 1020, 1023	114—115	831, 845, 859	912
O (газ)	163—165	174—175	178, 180	—	24—27	830, 844, 858	—
O ⁺ (газ)	166	175	178, 180	—	28—31	830, 844, 858, 872	—
O ⁻ (газ)	166	175	178—180	—	32—33	830, 844, 858, 872	—
O ₂ (газ)	166—171	175—177	179—180	1006, 1007, 1023	34—37	830, 844, 858, 872	900
O ₂ ⁺ (газ)	171—173	177	180	—	38—41	830, 844, 858, 872	—
O ₂ (газ)	173—174	177—178	180	1006, 1007, 1023	42—43	830, 844, 858, 872	900
OD (газ)	215—217	229	235, 237	—	86—87	830, 844, 858, 872	—
OH (газ)	212—215	227—229	233—235, 237	—	60—63	830, 844, 858, 872	—
OH ⁺ (газ)	217—219	230	235—237	—	64—67	830, 844, 858, 872	—
OH ⁻ (газ)	218—219	229—230	236—237	—	68—69	830, 844, 858, 872	—
OT (газ)	217	229	235, 237	—	104—105	831, 845, 859, 873	—
P (газ)	403	420	429, 436	1023	276—277	834, 848, 862	—
P ₂ (газ)	403—405	420—421	430, 436	—	278—279	834, 848, 862, 875	—
P ₄ (газ)	410—411	424—425	430, 436	—	280—281	834, 848, 862, 875	—
PCl (газ)	406—409	421—423	434—436	—	302—303	835, 849, 863, 876	—
PCl ₂ (газ)	414—416	424, 427—428	434—436	—	304—305	835, 849, 863, 876	—
PCl ₃ (газ)	417—419	424, 428	434—436	—	306—317	835, 849, 863, 876	—
PF (газ) ⇌	408—409	421—423	432—433, 436	—	292—293	835, 848, 862, 876	—
PF ₂ (газ)	414	424—426	433, 436	—	294—295	835, 848, 862, 876	—
PF ₃ (газ)	414—416	424, 426—427	433, 436	1023	296—297	835, 848, 862, 876	—

Вещество	I том				II том		
	Молекулярные востоякые	Термодинамические функции	Термохимические величины	Свойства реальных газов	Таблица термодинамических свойств	Интерполяционные формулы	Таблица эквивалентных коэффициентов
PF ₃ (газ)	417—419	424—427	433, 436	—	208—209	835, 849, 863, 876	—
PH (газ)	407—409	421—422	432, 436	—	290—291	834, 848, 862, 876	—
PN (газ)	408, 410	421, 423—424	435—436	—	312—313	835, 849, 863, 876	—
PO (газ)	404—407	421—422	430—431, 436	—	282—283	834, 848, 862, 875	—
POCl ₃ (газ)	414, 416—417	424, 428—429	435—436	—	308—309	835, 849, 863, 876	—
POF ₃ (газ)	414, 416—417	424, 427	433, 436	—	300—301	835, 849, 863, 876	—
PS (газ)	408—410	421, 423	435—436	—	310—311	835, 849, 863, 876	—
P ₂ O ₃ (газ)	411—412	424—425	431, 436	—	284—285	834, 848, 862, 876	—
P ₂ O ₅ (газ)	411—413	424—426	431—432, 436	—	286—287	834, 848, 862, 876	—
P ₂ O ₅ (газ)	411, 413—414	424, 426	432, 436	—	288—289	834, 848, 862, 876	—
Pb (т., ж.)	—	933—935	940, 944	—	568—569	—	—
Pb (газ)	923	929—930	940, 944	1024	570—571	840, 854, 868	—
PbF (газ)	925—927	930—932	942, 944	—	576—577	840, 854, 868, 881	—
PbF ₂ (т., ж.)	—	933, 936—937	942—944	—	578	—	—
PbF ₂ (газ)	928	932	943—944	1025	580—581	840, 854, 868—881	—
PbO (т., ж.)	—	933, 935—936	940—941, 944	—	572	—	—
PbO (газ)	925—926	930—931	941—942, 944	1024	574—576	840, 854, 868, 881	—
Rb (т., ж.)	—	903, 906—907	914	—	812	—	—
Rb (газ)	894—895	900—901	914	1025	814—815	843, 857, 871	—
Rb ⁺ (газ)	896—897	900—901	914	—	816—817	843, 857, 871, 884	—
S (газ)	310—311	329	339, 343	1010, 1023	198—199	833, 847, 861	—
S ₂ (газ)	312—314	329—331	339, 343	1010	200—201	833, 847, 861, 874	—
SF (газ)	312, 316	331, 333	342—343	—	214—215	833, 847, 861, 874	—
SF ₂ (газ)	324—325	337	342—343	—	216—217	833, 847, 861, 874	—
SF ₂ (газ)	324—326	337	343	1023	218—219	833, 847, 861, 874	—
SF ₆ (газ)	324, 326—327	337—338	343—344	1006, 1023	220—221	833, 847, 861, 875	905
SH (газ)	312, 315—316	331—333	341, 343	—	210—211	833, 847, 861, 874	—
SO (газ)	312, 314—315	331—332	339—340, 343	—	202—203	833, 847, 861, 874	—
SOF ₂ (газ)	324, 327	337—338	343—344	—	222—223	833, 847, 861, 875	—

Вещество	I том				II том		
	Молекулярные постоянные	Термодинамические функции	Термохимические значения	Свойства реальных газов	Таблица термодинамических свойств	Интерполяционные формулы	Таблица виральных коэффициентов
SO ₂ (газ)	315-318	333-334	340, 343	1006, 1010, 1023	204-205	833, 847, 861, 874	905
SO ₂ F ₂ (газ)	324, 327-328	337-338	340, 343	—	224-225	843, 847, 861, 875	—
SO ₃ (газ)	318-320	335, 337	343-344	1023	206-207	833, 847, 861, 874	—
S ₂ O (газ)	320-321	334-335, 337	341, 343	—	208-209	833, 847, 861, 874	—
Si (т., ж.)	—	681-683	685, 696	—	528-529	—	—
Si (газ)	662	675	685-687, 696	1024	530-531	839, 853, 867	—
Si ₂ (газ)	663-664	676-677	687, 696	—	532-533	839, 853, 867, 880	—
SiC (т.)	—	682, 684-685	694, 696	—	560-561	—	—
SiC (газ)	664, 668-669	676-677	694-696	1024	562-563	840, 854, 868, 881	—
SiC ₂ (газ)	669, 673-674	679, 681	695-696	—	564-565	840, 854, 868, 881	—
SiCl (газ)	662, 667	676, 678-679	692, 696	—	550-551	840, 853, 867, 881	—
SiCl ₂ (газ)	669-671	679-680	692-693, 696	—	552-553	840, 854, 868, 881	—
SiCl ₃ (газ)	669, 672	679-680	693, 696	—	554-555	840, 854, 868, 881	—
SiCl ₄ (газ)	669, 673	679-681	693, 696	1024	556-557	840, 854, 868, 881	—
SiF (газ)	664, 668-667	676, 678-679	690-691, 696	—	542-543	839, 853, 867, 881	—
SiF ₂ (газ)	669-670	679-680	691, 696	—	544-545	839, 853, 867, 881	—
SiF ₃ (газ)	669, 671	679-680	691, 696	—	546-547	840, 853, 867, 881	—
SiF ₄ (газ)	669, 672-673	679-681	691-692, 696	1006, 1019, 1024	548-549	840, 853, 867, 881	915
SiH (газ)	664-666	676-678	690, 696	—	540-541	839, 853, 867, 880	—
SiN (газ)	664, 667-668	676-677	693-694, 696	—	558-559	840, 854, 868, 881	—
SiO (газ)	663-665	676-677	687-688, 696	—	534-535	839, 853, 867, 880	—
SiO ₂ (т., ж.)	—	682-684	688-689, 696	—	536-537	—	—
SiO ₂ (газ)	669	679	689-690, 696	1024	538-539	839, 853, 867, 880	—
Si ₂ C (газ)	669, 674-675	679, 681	695-696	—	566-567	840, 854, 868, 881	—
Sr (т., ж.)	—	845-846	851, 858	—	734-735	—	—
Sr (газ)	833-835	842-843	851, 858	1025	736-737	842, 856, 870	—
Sr ⁺ (газ)	835	842-843	852, 858	—	738-739	842, 856, 870, 883	—
SrO (т., ж.)	—	845-846	852, 858	—	740-741	—	—
SrO (газ)	839, 841	843-844	852-855, 858	1025	742-743	842, 856, 870, 883	—

Вещество	I том				II том		
	Молекулярные постоянные	Термодинамические функции	Термодинамические величины	Свойства реальных газов	Таблица термодинамических свойств	Интерполяционные формулы	Таблица вириальных коэффициентов
T (газ)	182	190 - 191	193, 195	—	96 - 97	841, 845, 859	—
T ₂ (газ)	188 - 189	190, 193	195	102	98 - 99	841, 845, 859	—
TBr (газ)	296, 299	301, 303 - 304	306 - 308	—	172 - 173	841, 845, 860, 874	—
TCl (газ)	296, 298	301, 303	306, 308	—	154 - 155	832, 846, 860, 873	—
TF (газ)	295 - 296	301 - 302	306, 308	—	136 - 137	832, 845, 85, 873	—
TJ (газ)	296, 300	301, 304	307 - 308	—	190 - 191	833, 847, 861, 874	—
T ₂ O (газ)	205 - 206	224 - 225	231, 237	—	106 - 107	831, 845, 856, 873	—
Xe (газ)	945	945	—	1006, 1021	120 - 121	831, 845, 859	904
Zr (т., ж.)	—	933 - 934	937 - 938, 944	1025	586 - 587	—	—
Zr (газ)	916 - 923	928 - 929	937 - 938, 944	1025	588 - 589	840, 854, 868	—
ZrO (газ)	924 - 926	930	938, 944	—	590 - 591	840, 854, 868, 881	—
ZrO ₂ (т., ж.)	—	933 - 934	938 - 939, 944	—	592 - 593	—	—
ZrO ₂ (газ)	927 - 928	932	939 - 940, 944	1025	594 - 595	840, 854, 868, 881	—

- 1) Substance;
- 2) volume 1;
- 3) volume 2;
- 4) molecular constants;
- 5) thermodynamic functions;
- 6) thermodynamic quantities;

- 7) properties of real gases;
- 8) table of thermodynamic properties;
- 9) interpolation formulas;
- 10) table of virial coefficients;
- 11) solid body;
- 12) liquid;
- 13) gas.