TEMPERATURE DEPENDENCE OF THE SATURATED VAPOR PRESSURE OF TELLURIUM

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Data available in the literature on vaporization of tellurium are analyzed. The saturated vapor pressure of tellurium at temperatures below the melting point is measured and the equilibrium constant of the reaction $Te_2 = Te + Te$ is calculated.

In practice, it is often necessary to have exact data on the pressure and composition of the saturated vapor of tellurium over a wide temperature range.

The temperature dependence of the total vapor pressure of tellurium P_{total} above $T_{melt} = 723°K$ was studied in [1-6]. Analysis of these papers shows that there is good agreement between data obtained by different researchers and leads to the conclusion that the most reliable work is that in [1, 4], wherein almost identical equations were obtained experimentally:

$$\begin{gathered} \lg P_{total}(\text{torr}) = (7.5999 \pm 0.0136) - (5960 \pm 14.5)/T \ [1]; \\ \lg P_{total}(\text{torr}) = 7.5992 - 5960.5/T \ [4]. \end{gathered} \tag{1}$$

The vaporization of tellurium below T_{melt} was investigated experimentally in [7, 8]. It is evident from Table 1 that there is good agreement between the results in [7, 8], but extrapolation of the data in these papers and in [1, 4] to T_{melt} gives quite different magnitudes of the vapor pressure of tellurium at this temperature.

The experimental work in (9) was carried out in the temperature range including T_{melt} and a theoretical estimate is given in [10]. As follows from Table 1, there is good agreement between the data in [9] and in [1, 4, 7, 8] for temperatures chosen from overlapping regions and there is appreciable disagreement with the data in [10]. Thus, [9] is the only work which relates the results of investigations obtained in the temperature range above and below T_{melt} for tellurium.

Brebrick [9] measured the optical density of tellurium vapor at the absorption bands of tellurium with wavelengths 1995 Å, 2025 Å, 2056 Å, and 4357 Å and obtained the linear de-

TABLE 1.* Vapor Pressure of Tellurium from Data in Different Papers

Reference	Experimental temperature range, °K	P_{total}, torr, for T, °K					
		573	627	648	723, T_{melt}	798	998
[1]	785—1105	—	—	—	0,2272	1,353	42,46
[4]	919—1263	—	—	—	0,2265	1,349	42,36
[7]	593—683	$3{,}08\cdot10^{-4}$	$4{,}41\cdot10^{-3}$	$1{,}10\cdot10^{-2}$	0,1878	—	—
[8]	481—700	$3{,}16\cdot10^{-4}$	$4{,}38\cdot10^{-3}$	$1{,}08\cdot10^{-2}$	0,1777	—	—
[9]	643—888	$2{,}87\cdot10^{-4}$	$4{,}57\cdot10^{-3}$	$1{,}18\cdot10^{-2}$	0,2270	1,352	42,44
[10]	Theoretical	$1{,}94\cdot10^{-4}$	$3{,}46\cdot10^{-3}$	$0{,}93\cdot10^{-2}$	0,2020	1,300	45,0
Present paper, Eq. (11)	656—763	$2{,}83\cdot10^{-4}$	$4{,}54\cdot10^{-3}$	$1{,}18\cdot10^{-2}$	0,2272	—	—
Present paper, Eq. (13)	—	$2{,}64\cdot10^{-4}$	$4{,}54\cdot10^{-3}$	—	—	—	—

*The results in the table were obtained with the help of analytical equations, presented in the papers cited.

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pendence log D = f(1/T), whose slope coincided with the slope of the dependence (1) from [1] for temperatures above T_{melt}. Based on this, equations are obtained in [9], by comparing the results of the experiment therein with the data in [1], that express the temperature dependence of the vapor pressure of tellurium for temperature ranges above and below T_{melt}, respectively:

$$\lg P_{total}(\text{torr}) = 10.421 - 8001/T; \quad \lg P'_{total}(\text{torr}) = 7.600 - 5960.2/T. \tag{2}$$

Reference [9] has two questionable aspects. The absorption measurements were conducted on absorption bands, of which, according to the latest spectroscopic data for Te_2 molecules [11, 12], an exact transition is known only for 4357 Å, $A0_u^+(v' = 27) - X0_g^+(v'' = 0)$. This band, according to data in [9], was almost ten times less sensitive in absorption in tellurium vapors than the remaining three short-wavelength bands with unknown transitions. In the experiments in [9], the temperature of the cell containing the tellurium vapors was 1028°K. Nevertheless, starting from the magntiude of the dissociation energy of the Te_2 molecule, Brebrick proposed that tellurium vapor consists only of Te_2 molecules.

In the present work, we determined the dependence of the partial pressure of Te_2 and the total saturation vapor pressure of tellurium on the temperature of solid tellurium using a pulsed photolysis setup [13] calibrated according to the absolute concentration of Te_2 molecules. For this, using the procedure proposed in [9], the characteristics log D_λ = f(1/T) were measured for the tellurium band with wavelength 3999.7 Å. This band corresponds to the transition $X0_g^+(v'' = 0) - B0_g^+(v' = 19)$, [11, 12] and is situated between the 0-18 and 0-20 bands with wavelengths 4021 Å and 3979 Å, respectively.

For this work, we used the setup described in detail in [13], consisting of a cell with tellurium, a monochrometer, and a photoelectric detection system. An intensity-stabilized tungsten halide KGM 30-300 incandescent lamp was used instead of the high-frequency electrodeless lamp. A parallel beam of light passed through an interference filter with transmission maximum at the wavelength 4000 Å and then passed through the cell with the tellurium vapor at a temperature of 1000°K with a length 30 cm and a diameter 2.2 cm. The cell had an extension with length 12 cm, in which T-V 3 brand tellurium with an impurity content not exceeding 0.001%, redistilled in a vacuum of 10^{-6} torr, was placed. The cell, together with the extension, was separated from the rest of the vacuum system by a special valve after evacuation: The temperature of the cell throughout its entire length was the same to within ±10°K. The drift in temperature did not exceed 1°K. The temperature of the extension with tellurium was measured to within ±0.3°K and varied in the range (656-763)°K. The SPM-2 monochromater, adjusted to a wavelength of 3999.7 Å to within ±0.5 Å, had slits with a width of 0.04 mm, which corresponded to a transmission band of width 2.8 Å. The signal separated out by the photomultiplier was fitted into a microammeter through an intermediate amplifier.

The experimental results are shown by the points in Fig. 1 and were obtained in several series of measurements for both increased and decreased temperatures in the tellurium cell. The experiments showed that in order to establish the vapor equilibrium in the cell, corresponding to a fixed temperature in the extension with the tellurium, 20-30 min are sufficient.

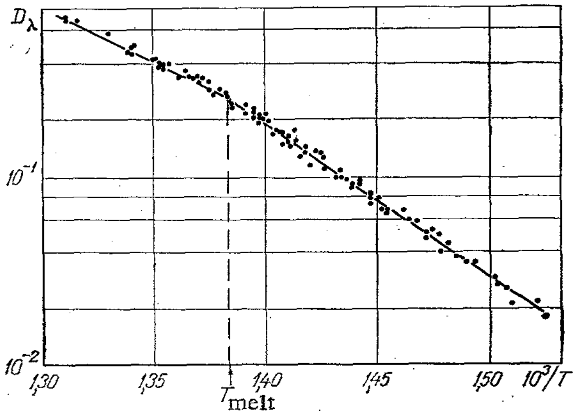

Fig. 1. The dependence of the optical density of tellurium vapor (3999.7 Å) on the tellurium temperature.

TABLE 2. Thermal Equilibrium Constant K_e at 1000°K for the Reaction $Te_2 = Te + Te$ according to Data in Different Papers

Reference	[10]	[15]	[16]	[17]
K_e, torr	0,00198	0,0006	0,0054	$0{,}76 \cdot 10^{-6}$

Reference	[18]	[19]	Present work
K_e, torr	$0{,}73 \cdot 10^{-9}$	$0{,}75 \cdot 10^{-5}$	$(1{,}04 \pm 0{,}07) \cdot 10^{-5}$

Analysis of the experimental results using the method of least squares yields linear equations, whose accuracy is determined by the mean-square deviations of the coefficients:

$$\lg D_\lambda = (10.6 \pm 0{,}1) - (8080 \pm 71)/T, \quad T < 723 \text{ K}, \tag{3}$$

$$\lg D_\lambda = (7{,}67 \pm 0.29) - (6012 \pm 220)/T, \quad T > 723 \text{ K}. \tag{4}$$

Going from Eqs. (3) and (4) to equations for the temperature dependence of the partial pressure of Te_2 molecules, it is necessary to compare (3) and (4) with the known magntiude of P_{Te_2} for one temperature of the extension with the tellurium. For this, it is convenient to choose T_{melt}, at which P_{total} of tellurium is given with good accuracy by Eqs. (1) in [1, 4]. In this case, in order to obtain the magnitude of P_{Te_2}, it is sufficient to know the equilibrium reaction constant K_e for thermal decomposition $Te_2 = Te + Te$ at the temperature of the cell containing the tellurium vapor (T_{cell} = 1000°K), since, according to [14], the fraction of tellurium molecules with a number of atoms exceeding 2 under the experimental conditions is much less than 1%.

The reaction $Te_2 = Te + Te$ was studied in a number of papers, whose results for T = 1000°K are presented in Table 2, which shows that these data are very contradictory and there is no basis for choosing any one of the works as being the most reliable. Together with this, it is known that the methods of statistical thermodynamics [20, 21] make it possible to calculate K_e for the reaction $A_2 = A + A$ when exact molecular constants are available for the ground and lowest excited electronic states. Application of such methods for halogen molecules leads to agreement with experiment to within 1% [21]. The availability of exact molecular constants for Te_2, obtained in recent years, permits carrying out a similar calculation of the thermal equilibrium constants for atoms and molecules in tellurium vapor.

According to [20, 21], the equilibrium constant is related to the partition functions for atoms q_A and molecules q_{A_2} by the relation

$$K_e = \frac{(q_A/N_A)^2}{q_{A_2}/N_A} \exp(-D_0^0/RT) = \frac{P_A P_A}{P_{A_2}} . \tag{5}$$

Substituting expressions for q_A and q_{A_2} from [21] into (5), we obtain

$$K_e = \frac{[g_A (2\pi m_A kT/h^3)^{3/2} RT/N_A]^2 \exp(-D_0^0/RT)}{g_{A_2} (2\pi m_{A_2} kT/h^3)^{3/2} (RT/N_A)(8\pi^2 IkT/\sigma h^2)[1 - \exp(-\theta_v/T)]^{-1}} . \tag{6}$$

Simplifying and substituting the numerical constants into expression (6), we obtain finally

$$K_e(\text{torr}) = \left[4{,}737 \cdot 10^{-14} \left(\frac{m_A m_A}{m_{A_2}}\right)^{3/2} \frac{\sigma}{\mu_{A_2} r_e^2} \frac{g_A^2}{g_{A_2}}\right] T^{3/2} \times$$
$$\times \left\{1 - \exp\left[-\frac{(\omega_e - 2x_e\omega_e) \cdot 1{,}43884}{T}\right]\right\} \exp\left(-\frac{D_0^0 \cdot 1{,}43884}{T}\right) . \tag{7}$$

In this work, in calculating the equilibrium constant using Eq. (7), the following parameters, characterizing tellurium atoms and molecules, were used: $m_A = 128$ and $m_{A_2} = 256$; $\sigma = 2$; $I = \mu r_e^2$; $r_e^2 = 2.548$ Å [12]. The values 247.036 cm^{-1} and 0.5169 cm^{-1} were obtained in [12] for w_e and $x_e w_e$, respectively. It is shown in [12] that due to the presence of a c-type bond, according to Hund, the ground state of the Te_2 molecule is split into a 0_g^+ state and a 1_g state, of which the last state has a higher energy by 1975 cm^{-1}. In this case, in calculating the statistical weight of Te_2 molecule, for Eq. (7), the following should be used:

$$g_{A_2} = \sum_{i=1}^{n} (2j_i + 1) \exp E/kT \text{ [20]},$$

or in particular, for Te_2

$$g_{Te_2} = (2j_i + 1) + (2j_i + 1) \exp(1975 \cdot 1.4388/T).$$

Similarly, for g_A of tellurium atoms, in Eq. (7), it is necessary to take into account the total statistical weight of the three ground-state triplet states $Te^3P_{0,1,2}$, whose energy splitting is taken from [22]:

$$g_{Te} = (2j_i + 1) + (2j_i + 1) \exp(4707 \cdot 1.4388/T) +$$
$$+ (2j_i + 1) \exp(4751 \cdot 1.4388/T).$$

It is evident from Eq. (7) that the exponential factor containing the dissociation energy of the molecule $D_0^0(Te_2)$ has the largest effect on the quantity K_e. There are several papers in the literature in which $D_0^0(Te_2)$ is determined by different methods and the same results are obtained. Since there is no reason to prefer any one of these works, the best variant appears to be the mean weighted value, obtained from a set of values of the dissociation energy (in cm^{-1}): 21595 ± 65, 21455 ± 400, 21230 ± 670, 21365 ± 100, 21540 ± 285, 21330 ± 200, proposed in [11, 17, 23, 24, 25, and 26], respetively. Calculation of the mean weighted value leads to the quantity $D_0^0(Te_2) = (21370 \pm 45)$ cm^{-1}, where the magnitude of the error is one mean-square deviation.

Substituting into (7) the parameters listed and analyzed above, we obtain the expression $K_e = f(T)$ for the Te_2 molecule:

$$K_e\,(\text{torr}) = 1167 T^{3/2} \exp[-(30747 \pm 45)/T]\,[1 - \exp(-354/T)] \times$$
$$\times \frac{[5 + 3\exp(-6835/T) + \exp(-6772/T)]^2}{1 + 3\exp(-2842/T)}. \tag{8}$$

$K_e(Te_2)$, calculated in Eq. (8) taking into account the uncertainty in the determination of $D_0^0(Te_2)$, agrees satisfactorily only with the results of [19] (Table 2), where the following equation, obtained by the methods of thermodynamics, was used to find $K_e(Te_2)$:

$$\lg K_e\,(\text{torr}) = -1349/T + 1{,}59 \lg(1/T) - 0{,}9592 \cdot 10^{-3}/T -$$
$$- 2355\,(1/T)^2 + 0{,}10046 \cdot 10^{-4} T^{3/2} + 4{,}237. \tag{9}$$

In [19], the quantity $D_0^0(Te_2)$ from [11] was used to obtain Eq. (9). If in Eqs. (7) and (8), we substitute this magnitude of the dissociation energy, then in the calculations we obtain K_e coinciding in magnitude with the results of a calculation using Eq. (9), which indicates that both methods give identical results.

Now, using the dependences (5) and (8) and $P_{total} = P_{Te_2} + P_{Te}$, it is possible to calculate the partial fraction of atoms and tellurium molecules at $T_{melt} = 723$°K and the temperature of the cell with the vapor $T_{cell} = 1000$°K. The calculation, carried out taking into account errors in determining K_e and P_{total}, gives: $P_{total} = (0.2272 \pm 0.0024)$ torr [1]; $P_{Te_2} = (0.2257 \pm 0.0024)$ torr, $P_{Te} = (1.53 \pm 0.052) \cdot 10^{-3}$ torr. Comparing the value of P_{Te_2} obtained and the dependence (3), we obtain the equation for the dependence of the partial pressure of Te_2 molecules on the temperature of solid tellurium in the saturated vapor at 1000°K:

$$\lg P_{Te_2} = (10.5292 \pm 0.1) - (8080 \pm 71)/T, \ T < 723 \text{ K}^*. \qquad (10)$$

Using $P_{total} = P_{Te} + P_{Te_2}$, (5), and (10) in the value of K_e (1000°K), it is possible to calculate the dependences $P_{total} = f(T)$ and $P_{Te} = f(T)$. Applying the method of least squares to the set of computed values at separate temperatures leads to the equations:

$$\lg P_{total}(\text{torr}) = (10.45 \pm 0.10) - (8021 \pm 71)/T, \ T < 723 \text{ K}; \qquad (11)$$

$$\lg P_{Te}(\text{torr}) = 2.752 - 4026/T, \ T < 723 \text{ K}, \ T_{cell} = 1000 \text{ K}. \qquad (12)$$

The mean-square deviation in determining P_{Te_2} and P_{total} from Eqs. (10) and (11) is less than 2%, while that in determining P_{Te} using Eq. (12) does not exceed 3.5%. These estimates of errors were obtained based on the experimental material, presented in Fig. 1, using the technique proposed in [27].

Equation (11) obtained in this paper describes the equilibrium between the saturated vapor pressure of tellurium and the temperature of the solid phase of tellurium and does not depend on the temperature of the cell which contains the vapor. Calculation using Eq. (11) gives values of P_{total}, which agree well with the results in [9], as well as with the results in [7, 8] in the temperature range of the experiments carried out in these works (see Table 1). The partial pressures P_{Te_2} and P_{Te} are related by Eq. (5) for K_e and depend on the vapor temperature. For this reason, the dependences (10) and (12) are only applicable for saturated tellurium vapor heated to 1000°K. For other temperatures, it is necessary to carry out the calculation anew with the help of dependences (5), (8), and (11). Such calculations for experimental conditions such that $T = T_{melt} = T_{cell}$ give $K_e = 7 \cdot 10^{-11}$ torr and $P_{Te} = 4 \cdot 10^{-6}$ torr, which is a very small quantity: 0.0017% of the total vapor pressure of tellurium at 723°K.

Thus, from an analysis of the data in the literature and the results of the present work, it follows that the temperature dependence of the saturated vapor pressure of tellurium in a temperature range above T_{melt} is described with good accuracy by Eqs. (1) in [1, 4], while in the temperature range below T_{melt}, Eq. (11) above is applicable, as well as Eq. (2) in [9] (the agreement between the data is about 1%). The temperatures of the experiments of these works cover the interval (643-1263)°K, but Eqs. (2) and (11), evidently, are valid up to a temperature of 627°K, which, according to data in [28], is the phase transition point in crystalline tellurium. The range of temperatures examined corresponds to a saturated vapor pressure of tellurium (0.0045-760) torr.

Above this temperature range, we can recommend the results of [5], Eq. (13), wherein the saturated vapor pressure of tellurium was studied up to 21 atm (temperature range 981-1780°K) and whose data in the temperature range near 1263°K agree well with the data in [1, 4]:

$$\lg P(\text{torr}) = 11.0755 - 6447/T - \lg T. \qquad (13)$$

The work described in [7, 8] was carried out in the temperature range including the phase transition point T = 627°K. According to [29, 30], in using equations of the type (2) and (11) in the temperature range below the phase transition point, it is necessary to take into account the heat of the phase transition ΔH_{pt}. In this case, Eq. (11) above for temperatures below 627°K has the form

$$\lg P_{total}(\text{torr}) = 10.8 - 8239/T \qquad (14)$$

and its accuracy is determined by the accuracy of the quantity $\Delta H_{pt} = 1$ kcal/mole for tellurium [28]. Calculation using Eq. (14) does not agree very well with the data in [7, 8] (see Table 1). The results of the present work show that it is possible to check the validity of Eq. (14), as well as the data in [7, 8], by measuring the dependence $\log D_\lambda = f(1/T)$ on a setup permitting measuring D_λ in the range 0.001-0.02.

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*Using (4), an analogous dependence can be obtained for T > 723°K. This was not done due to the availability of very accurate Eqs. (1) and significant errors in the coefficients in Eq. (4).

NOTATION

P_{total}, total saturated vapor pressure of tellurium; P_{Te} and P_{Te_2}, partial pressures of tellurium atoms and molecules in the saturated vapor; T, T_{cell}, and T_{melt}, temperature of the condensed phase of tellurium, temperature of the cell containing the tellurium vapor, and melting temperature of tellurium, respectively; D_λ, optical density of tellurium vapor at a definite wavelength; K_e, equilibrium constant; q_A and q_{A_2}, partition functions of atoms and diatomic molecules, respectively; N_A, Avogadro's number; D_0^0, dissociation energy of the molecules; R, universal gas constant; M_A and M_{A_2}, atomic and molecular weights in gram-atoms; σ, symmetry factor of the diatomic molecules; I, moment of inertia of the diatomic molecules; μ, reduced mass; r_e, internuclear separation of the diatomic molecules; k, Boltzmann's constant; h, Planck's constant; θ, characteristic temperature; w_e and $x_e w_e$, vibrational and anharmonicity constants of the diatomic molecules; g_A and g_{A_2}, statistical weights of the atom and diatomic molecule; j_i, quantum number of the resulting angular momentum of the i-th state of the atom or molecule; E_i, energy of the i-th state; and ΔH_{pt}, heat of the phase transition.

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DRYING OF GRANULATED THERMOLABILE PLASTICS IN A VIBROROTATING LAYER

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Features of drying and storing of thermoplastics were determined on an installation of industrial dimensions.

At present the technology of molding plastic products, with ever more stringent requirements as to the quality of the products, poses the problem of thorough dehydration (to hundredths of a percent) of the raw material before molding. Initial experiments with thorough dehydration of thermoplastics showed that it is very promising to use for this purpose apparatus with a vibrorotating layer [1]. After intensive mixing in combination with evacuation, certain peculiarities of the drying of these materials arise. Some of these peculiarities will be examined in the present work.

A diagram of the installation is shown in Fig. 1. The inner diameter of the chamber is 0.5 m, its height is 0.7 m. The material was fluidized by the rotation of the rotor consist-

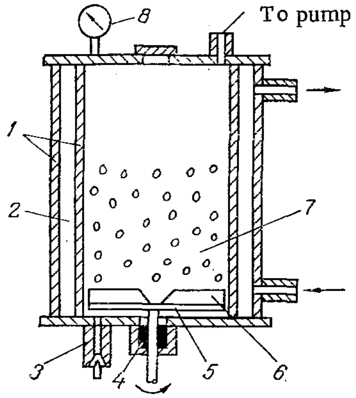

Fig. 1. Diagram of the experimental installation: 1) chamber with double walls; 2) water; 3) flow regulator; 4) vacuum seal; 5) disk; 6) blades; 7) material; 8) manometer.

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