PHYSICAL PROPERTIES OF WELL SEASONED MOLYBDENUM AND TANTALUM AS A FUNCTION OF TEMPERATURE

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Abstract

Spectral emissivity, spectral reflectivity, resistivity, radiation intensity, and thermal expansion measurements as a function of temperature have been made for well seasoned molybdenum and tantalum. From the data thus obtained values for many other physical quantities as functions of temperature have been computed. As such physical quantities, *average visible emissivity, color emissivity, total emissivity, brightness temperature, color temperature, radiation temperature, normal brightness, luminous efficiency,* and certain temperature variation coefficients have been included. Comparison measurements on moderatly pure and very pure molybdenum showed no appreciable variation in radiation properties, though appreciable variations appeared in their resistivities. The results for very pure molybdenum and moderately pure tantalum are incorporated in tables giving values where known for every 200 degrees between room temperature and the melting points.

Molybdenum

Introduction. Once given the temperature scale[1] for molybdenum, a determination of some of the physical properties naturally follows. Herewith are presented measurements of resistance, total radiation and thermal expansion. From these, with the aid of the temperature scale, various physical properties as a function of temperature have been derived.

The data have been obtained from two lamps. Both lamps had long cylindrical filaments bent into squared U-forms with fine pick-off leads spot welded to the main filaments for the elimination of end loss effects. Both lamps were well evacuated by means of a condensation pump operating through a liquid air trap. The familiar coil test showed high vacua in both cases.

During their lives, both lamps were operated at such high temperatures that on account of vaporization they had to be opened up three or four times to remove the deposit on the inside of the bulb. Finally, after having found the highest temperatures for safe operation, a series of measurements was obtained on each, the lamps opened and the filaments measured. The dimensions obtained were, therefore, strictly applicable to the filaments as used in obtaining the data to be reported.

[1] Worthing, Phys. Rev., **25**, 846, 1925.

Mechanical data relating to the filaments are given in Table I. The filament of 2-W-37 was supplied by the Cleveland Wire Division of the National Lamp Works. It was drawn from the most nearly

TABLE I

Mechanical data relating to molybdenum filaments in the lamps studied.

Lamp	Radius	Overall length	Length between pot. leads	Surface	Maximum end loss correction	Filament purity
2–W–46	0.262mm	39.8cm	31.6cm	5.20cm²	0.3%	Commercial
2–W–37	0.120	41.2	30.1	2.27	0.0	Very pure

pure slug that that division had been able to obtain. Results on the temperature coefficient of resistance, as will be reported, bear out this claim for purity.

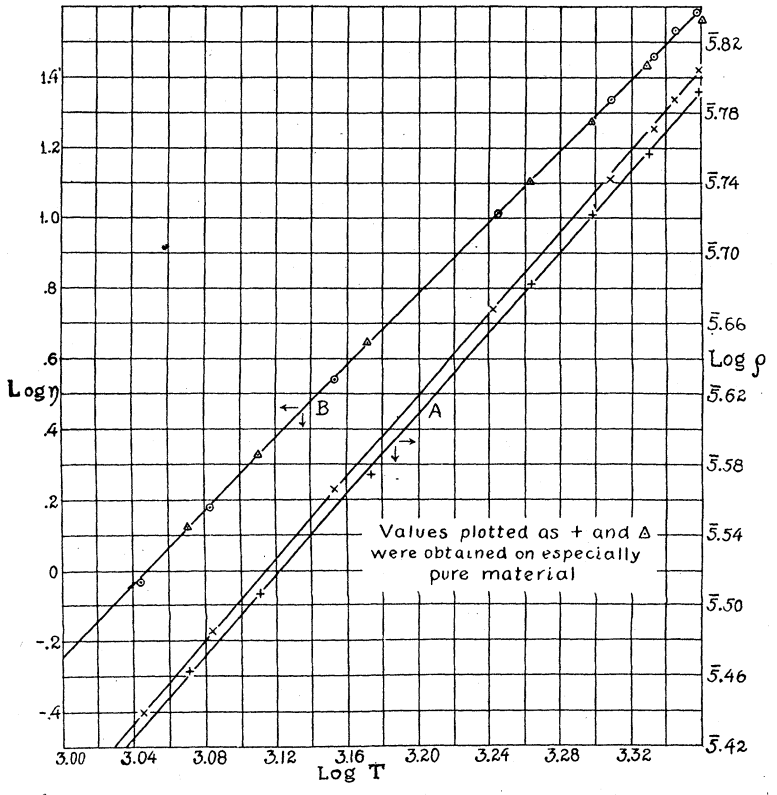

Fig. 1. Variation of resistivity ρ in ohm cms (Curve A) and radiation intensity η in watts/cm² (Curve B) with temperature T in °K for well seasoned molybdenum.

Resistivity, radiation intensity, and thermal expansion measurements. Measurements of resistance and of the total radiation rate at high

temperatures were made simultaneously. Temperatures were obtained with a disappearing filament pyrometer. Settings were made at several points along the filaments. The observed resistivities and radiation intensities are shown in Fig. 1. The individual data on radiation intensity for the two lamps fall very closely on the same curve. The resistivity curves for the two specimens of wire differ, however. While the resistivities at incandescence show the same variation with temperature, there is a definite difference at any one temperature, the resistivity of the pure material being less by about 2 percent. As with tungsten, the plot showing $\log\rho$ as a function of $\log T$ is a straight

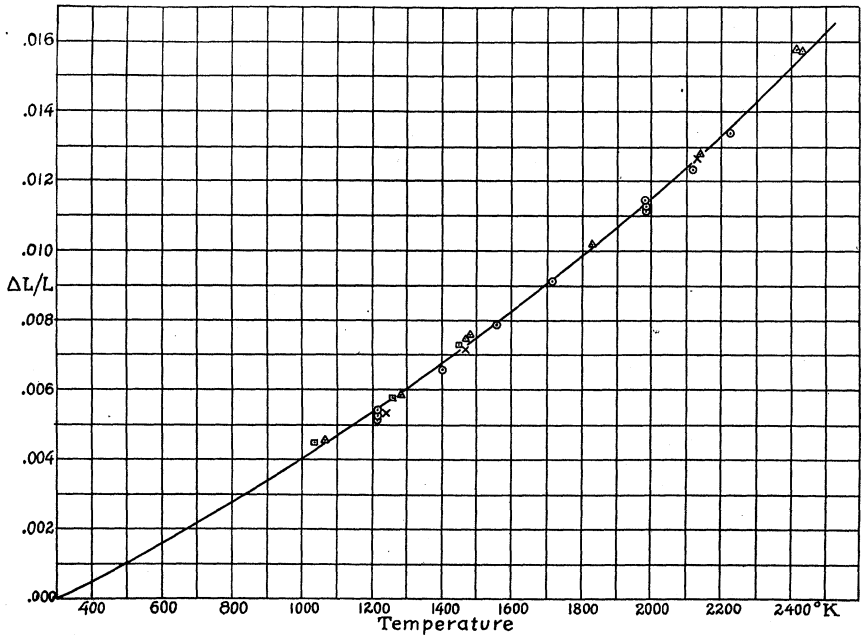

Fig. 2. Thermal expansion of molybdenum.

line. Its slope, which gives the percentage change in resistance due to one percent change in temperature, is less for molybdenum, however, being about 1.145, against 1.200 for tungsten.

Thermal expansion measurements were made with the aid of a position microscope and a micrometer microscope. Small scratches made on the filaments before mounting were used as reference marks. The data on thermal expansion (see Fig. 2) are the results of four runs. In each set some low incandescent temperature was selected as a condition to which references were frequently made to assure freedom from instrumental variations. In each set at the beginning, one meas-

urement of expansion between room temperature and this reference temperature was taken. Because of the long time required by a filament in vacuo to reach temperature equilibrium at room temperature after being once heated to incandescence, the usual check back could not be carried out for this step. The equation obtained for the curve of Fig. 2 is

$$L/L_{300}=5.00\times10^{-6}(T-300)+10.5\times10^{-10}(T-300)^2.$$

The first coefficient is greater than that obtained for tungsten, namely 4.44×10^{-6} deg.$^{-1}$. It is less, however, than the 5.08×10^{-6} deg.$^{-1}$ which is obtained from the equation giving the carefully determined results of Schad and Hidnert,[2] though well within the range of the recently determined coefficients of Hidnert and Gero[3] for various specimens having different coarseness of grains. The check is quite satisfactory.

Measurements of resistance at low temperatures were made by a Kelvin double bridge method with the filament immersed in a high grade lubricating oil and heated externally. Temperatures were determined with mercury in glass thermometers which were calibrated in melting ice and in steam and compared elsewhere with the indications of a standard calibrated Pt, Pt+10 percent Rh thermocouple.

Purity of the molybdenum in lamp 2-W-37. Giess and van Liempt[4] showed from a study of alloys of tungsten and molybdenum that the average temperature coefficient of resistance between 0°C and 100°C, α, for any specimen gave a very good index of its purity. In a plot of α as a function of atomic percent of Mo, they found $\alpha\times10^5$ in deg.$^{-1}$, starting with 100 percent W and 0 percent Mo, to have a value of 482, to decrease rapidly with increasing Mo content to a minimum of 290 at 35 percent Mo and thereafter to increase at first slowly and then more rapidly to 457 for 100 percent Mo. Later, in a personal letter, Dr. van Liempt states that he has obtained still purer Mo with $\alpha\times10^5$ equal to 463. Apparently for nearly pure Mo a change of 1 percent in the tungsten content means approximately a change of 10 in this quantity.

The writer, in his measurements made previous to Giess and van Liempt's publication, obtained three values at different temperatures which can be used in checking purity. Using the formula

$$\alpha=\frac{R_1-R_2}{R_2t_1-R_1t_2} \tag{1}$$

[2] Schad and Hidnert, Bull. Bur. Stds., **15,** 31, 1919.

[3] Hidnert and Gero, Bull. Bur. Stds., p. 429, 1924.

[4] Giess and van Liempt, Z. für Anorg. u. Allg. Chem., **128,** 355, 1923.

where α refers to the temperature range 0° to 100°C and R_1 and R_2 the resistances of the specimen at two temperatures, t_1 and t_2, expressed in °C, he obtained three values for $\alpha \times 10^5$ in deg.$^{-1}$, namely, 463, 464, and 464. The check with van Lempt's later result for pure Mo is excellent and indicative of the supposed purity of the specimen. The foregoing indicates that for Mo for $0°C < t < 100°C$

$$R = R_0(1 + 0.00464t) \ . \tag{2}$$

For the range $0°C < t < 200°C$ additional readings (see Table II) indicated the best value for α should be decreased by about 2 in the

TABLE II

Resistance temperature data for a pure molybdenum specimen at low temperatures.

t	R	.3417 (1+.000464t)	.3418 (1+.000462t)
30.80°C	.3905 ohms	.3905 ohms	.3905 ohms
52.10	.4242	.4243	.4241
100.85	.5015	.5014	.5011
155.7	.5872	.5887	.5876
181.7	.6287	.6299	.6287
206.5	.6681	.6692	.6679

last place. This may only mean that the temperature measurements are more uncertain in the range $100°C < t < 200°C$, or that the departure from the linear relation is more evident over the longer range.

Unfortunately, the variation in resistance with temperature at low temperatures was not determined for lamp 2-W-46. From a careful comparison of the variation between low incandescence and room temperature, for the two lamps, it is certain that, at low temperatures, the temperature coefficient of 2-W-46 was much lower than for the pure material of 2-W-37. Whereas, as shown in Fig. 1, at incandescent temperatures the resistivity of the pure specimen was about 2 percent less than for the other specimen, at room temperature it was about 11 percent less. These values are in agreement with what is expected from a consideration of Giess and van Liempt's results and the known purities of the wires used.

Collected data. From the data presented in the paper on the temperature scale, together with that presented above, it is possible to compute values for various other properties. In Table III at equal temperature intervals from room temperature, values for the observed and computed properties are given. The meaning of some of the terms and the manner of their obtaining follow.

Color emissivity, e_c, and color temperature, T_c, have significance for a source only when its light may be matched with that from a black

TABLE III

Properties of well seasoned molybdenum for various temperatures
($c_2 = 14330\mu$ deg., $T_{au} = 1336°K$)

Temperature T °K	Spectral Emissivity $e_{.665\mu}$	Spectral Emissivity $e_{.475\mu}$	Average Visible Emissivity e_v §	Color Emissivity e_c §	Total Emissivity e_t	Brightness Temperature $S_{.665\mu}$ °K	Color Temperature T_c °K	Radiation Temperature T_r °K	Relative Length L/L_o	Resistivity ρ μ-ohm cm. * ‡	$\frac{T}{\rho}\frac{d\rho}{dT}$ * ‡	Brightness normally B_n candles/cm²	$\frac{T}{B_n}\frac{dB_n}{dT}$	Radiation Intensity η watts/cm²	$\frac{T}{\eta}\frac{d\eta}{dT}$	Luminous Efficiency eff lumens/watt † §
273	.420	.425	....	....	....	....	.....	....	.....	5.14	1.265	.....	....	.....	....	.....
300	.419	.424	....	....	....	....	....	....	1.00000	5.78	1.235	.....	....	.....	....	.....
400	.415	.421	....	....	....	....	....	....	.....	8.15	1.170	.....	....	.....	....	.....
600	.406	.415	....	....	....	....	....	....	.....	.....	.....	.....	....	.....	....	.....
800	.398	.409	....	....	....	....	.....	....	.....	.....	.....	.....	....	.....	....	.....
1000	.390	.403	.393	.361	.096	958	1004	557	1.0041	23.9	1.145	.00010	22.3	.55	5.32	.....
1200	.382	.398	.386	.347	.121	1139	1207	708	1.0054	29.5	1.145	.0050	19.2	1.43	5.23	.....
1400	.375	.393	.379	.333	.145	1316	1411	864	1.0068	35.2	1.145	.089	16.9	3.18	5.16	.093
1600	.367	.388	.373	.321	.168	1489	1616	1024	1.0083	41.1	1.145	0.765	15.1	6.30	5.10	.40
1800	.360	.383	.367	.309	.189	1658	1823	1187	1.0099	47.0	1.145	4.13	13.6	11.3	5.04	1.22
2000	.353	.379	.362	.297	.210	1824	2032	1354	1.0115	53.1	1.145	15.9	12.4	19.2	4.99	2.75
2200	.347	.375	.357	.287	.230	1986	2244	1523	1.0133	59.2	1.145	48.5	11.3	30.7	4.94	5.28
2400	.341	.371	.352	.277	.248	2143	2456	1693	1.0151	65.5	1.145	123.	10.4	47.0	4.90	8.70
2600	.336	.368	.348	.268	.265	2297	2672	1866	.....	71.8	1.145	270.	9.6	69.5	4.86	13.0
2800	.331	.365	.344	.260	.281	2448	2891	2039	.....	78.2	1.145	540.	8.9	98.	4.83	18.4
2895¶	.328	.363	.342	.255	.290	2519	2997	2122	.....	81.4	1.145	730.	8.6	116.	4.81	.....

* Values given for ρ and $\frac{T}{\rho}\frac{d\rho}{dT}$ assume the dimensions possessed at 300°K. All other quantities have been corrected when necessary so as to be in strict accord with definitions.

‡ For the range 0°C < to < 200°C, the resistivity is given by the equation $\rho_t = \rho_o(1 + 0.00462t)$. The molybdenum wire used was especially pure.

† Luminous efficiencies were obtained by applying the relation $\left[\text{eff} = 1.06\pi\frac{B_n}{\eta}\right]$. The factor 1.06 was introduced to account for the deviation of molybdenum from Lambert's cosine law.

§ Values given for e_v, e_c, T_c, and eff presuppose an acurate color match between molybdenum at T and a black body at T_c.

¶ Melting point.

body. This is the case with molybdenum. Quantitatively for a source at a temperature, T, the color emissivity is the ratio of the brightness of the source to that of a black body at color match that is at T_c. Mathematically the relation is given by

$$ln\ e_c=\frac{c_2}{\lambda}\left(\frac{1}{T_c}-\frac{1}{S}\right)$$

where e and S are the spectral emissivity and brightness temperature for wave-length λ and c_2 the Wien-Planck constant.

Brightness may be computed in either one of two ways; either as the product of e_c and the brightness of a black body at temperature T_c, or as the product of the average visible emissivity, e_v, and the brightness of a black body at temperature T. In equation form,

$$ln\frac{e_v}{e_c}=\frac{c_2}{\lambda_0}\left(\frac{1}{T}-\frac{1}{T_c}\right)$$

where λ_0 is the Crova wave-length for black body radiation.

Total emissivity, e_t, and radiation temperature, T_r, corresponding to any temperature, T, have been computed from the defining equations

$$\eta=e_t\sigma T^4=\sigma T_r^4$$

where η is the radiation intensity expressed here in watts/cm^2, and σ the Stefan-Boltzmann constant.

Luminous efficiencies have been computed from the normal brightness and radiation intensities, with a correction of 6 per cent (to be published later) for deviation from Lambert's cosine law.

The more nearly constant factors

$$\frac{Td\rho}{\rho dT},\ \frac{TdB_n}{B_n dT},\ \text{and}\ \frac{Td\eta}{\eta dT}$$

are the slopes on corresponding log plots.

There are very little published data for molybdenum on the properties at incandescence which are considered here. Greenslade[5] has published in an abstract spectral energy curves for various temperatures, but the plots are in such a form that comparisons cannot well be made. Somerville[6] has studied the resistance change with temperature between room temperature and 1000°C. The results do not check with what is given here. The cause is probably due to impurities in Somerville's molybdenum which was prepared when the metallurgy

[5] Greenslade, Phys. Rev., **15,** 150, 1920.
[6] Somerville, Phys. Rev., **31,** 261, 1910.

of molybdenum was much less well developed. In contrast with the temperature coefficient of resistance between 0°C and 100°C, α_{0-100}, of 0.00463 1/deg. as obtained by Giess and van Liempt and by the present author, as reported above, Somerville obtained 0.0033 1/deg. More recently Blom[7] has published resistivity results for the range 0°C to 2000°C. Unfortunately the check here is not good. Perhaps here also lack of purity of the specimens used is the cause for discrepancy. Expressed in micro-ohm cm, he found 4.4 as the resistivity at 0°C, as against 5.78 for the very pure and 6.41 for the commercial specimens used by the present author. For α_{0-100}, Blom obtained 0.00414 1/deg. as against 0.00463 1/deg. Reduced to the temperature scale employed here, Blom's results give for $\frac{T}{\rho}\frac{d\rho}{dT}$ values which increase gradually from 1.11 at 0°C to 1.23 at 1600° to 2000°K; whereas the present work gives results which starting with 1.265 at 0°C decrease gradually to the constant value of 1.145 which applies above 1000°K. From the standpoint of variations in α_{0-100} and in $T/\rho\ d\rho/dT$, the differences might be explained by assuming Blom's wire to have been less pure than the present author's. The actual resistivities found at room temperature do not fit in with this explanation, however.

Tantalum

From some of the same wire that was used in the spectral emissivity work (see preceding paper), a lamp with voltage pick-offs of fine tungsten wire was made. This lamp had a good vacuum. The filament was well aged at a temperature of about 2700°K, and its surface, during the measurements made on it, was well polished and quite free from pits and any apparent damage resulting from any residual gases. The dimensions of the part between the potential leads were: diameter, 0.496 mm; length, 30.5 mm. At 23°C its resistance was 0.0228 oms.

Determinations of temperature distribution along the length of the filament were made for four widely separated temperatures to take account of variations due to lack of constancy in diameter. Then for several currents, resistance, wattage and thermal expansion measurements were made. Methods and precautions were the same as in the study of molybdenum.

Resistivity. In curve *A*, Fig. 3, resistivity as a function of temperature ($1500°K < T < 2800°K$) is shown on a logarithm scale. The locus is nearly a straight line with the slope $\frac{T}{\rho}\frac{d\rho}{dT}$ equal to 0.785. Pirani's[8]

[7] Blom, Phys. Rev., **13,** 309, 1919.

[8] Pirani, Verh. Deut. Phys. Gesell., **12,** 301, 1910.

data plotted similarly gives a noticeably greater slope, namely, 0.86. This difference cannot be ascribed to a difference in temperature scales. It may be found, perhaps, in Pirani's method of eliminating cooling effects due to leads. He used the method of differences as obtained from two filaments of unequal length. This method is good in case the filaments have the same cross-section, but such a coincidence occurs too seldom according to the present writer's experience, and instead he has used the potential lead method.

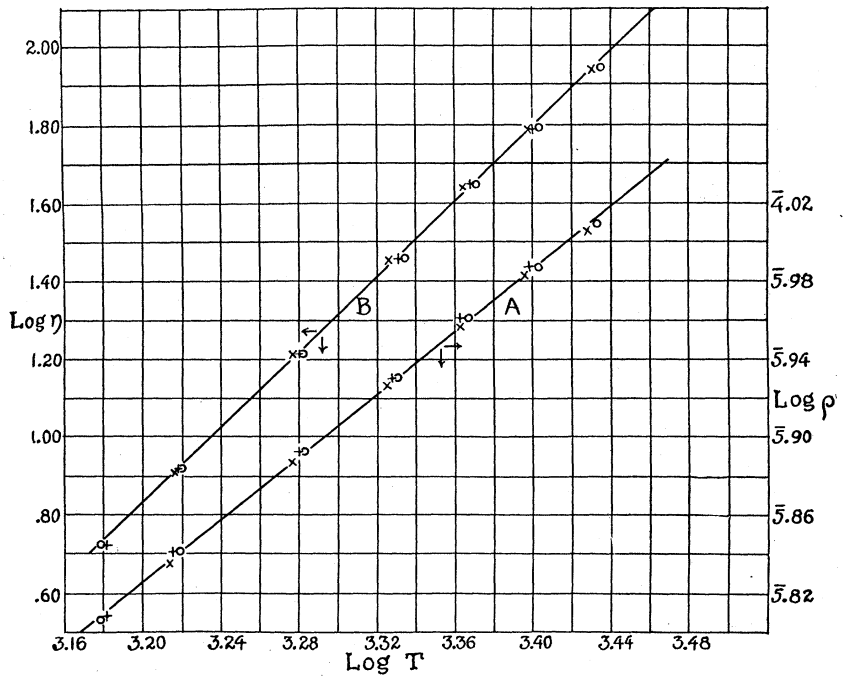

Fig. 3. Variation of resistivity ρ in ohm cms (Curve A) and radiation intensity η in watts/cm^2 (Curve B) with temperature T in °K for well seasoned tantalum.

At the lowest temperature shown in Fig. 3, there is indication of an increase in the slope $\frac{T}{\rho}\frac{d\rho}{dT}$. This is consistent with the average slope of .915 between room temperature and 1500°K which is obtainable from the data given. This low temperature value agrees satisfactorily with Holborn's[9] data and also with Pirani's for this range, but not with Bridgman's[10] as is indictated by Holborn's comparison.

[9] Holborn, Ann. Physik, **59**, 145, 1919.
[10] Bridgman, Proc. Nat. Acad. Sci., **3**, 10, 1917.

The resistivity of 14.6 microohm-cm. at 300°K does not check with Pecheux's[11] 17.5 microohm-cms.

In contrast with tungsten and molybdenum, for which $\frac{T}{\rho}\frac{d\rho}{dT}$ at incandescent temperatures are 1.200 and 1.145, this quantity for tantalum is lower than unity, being about 0.78.

Radiation intensity. In curve *B*, Fig. 3, the variation in the radiation intensity of tantalum with temperature is shown. Three sets of measurements taken in the order ×, +, o are indicated. Just why

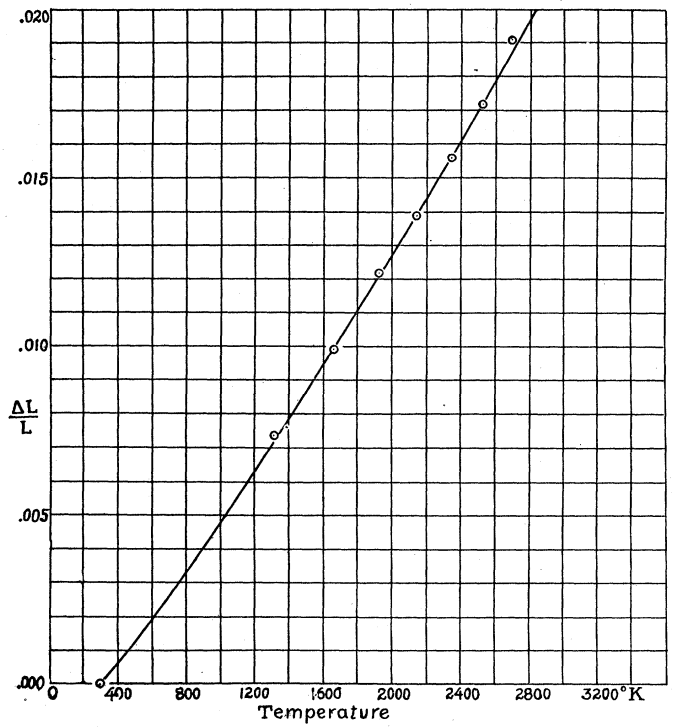

Fig. 4. Thermal expansion of tantalum.

the O values of curve *B* should lie so much further from the × and + values than in case of curve *A* is uncertain. The supposition of an error of determination of the temperature combined with an appreciable filament vaporization between sets does not fit. The supposition of deleterious effects due to residual gases is also eliminated, for the filament surface maintained a high polish throughout. Taken al-

[11] Pecheux, Comptes Rendus, **153,** 1140, 1911.

together, there is an indication of some change in resistivity or in radiation intensity or both.

All three sets of measurements show separately the same percentage change in wattage for a given percentage change in temperature, $\frac{T}{\eta}\frac{d\eta}{dT}$ amounting to 4.80. Theoretically with increase in T, $\frac{T}{\eta}\frac{d\eta}{dT}$ must finally decrease to 4, the black body value. That line *B*, Fig. 3, is straight may indicate an error in the locating of the spectral emissivity curve as presented in the preceding paper. This does not tend, however, to indicate essential correctness in other reported temperature scales for tantalum such as those resulting from Mendenhall's and Forsythe's or McCauley's emissivity data, for on their temperature scales an appreciable increase in $\frac{T}{\eta}\frac{d\eta}{dT}$ with temperature would follow.

The same comments with regard to published data that are made above relative to resistivity at incandescent temperatures apply equally here.

TABLE IV

Physical properties of well seasoned tantalum.

T Temperature °K	Spectral emissivity at 0.665μ	Spectral emissivity at 0.463μ	Brightness Temp. °K $s_{0.665\mu}$	Color Temp.* °K T_c	Resistivity ρ μ-ohm-cm	$\frac{T}{\rho}\frac{d\rho}{dT}$	Radiation Intensity η watts/cm^2	$\frac{Td\eta}{\eta dT}$	Total Emissivity e_t	Radiation Temperature T_r °K
300	.493	.56	....	....	.....	.....	.....	....	....	....
1000	.459	.52	966	....	.....	.....	.....	....	....	....
1200	.450	.51	1149	....	.....	.....	.....	....	....	....
1400	.442	.50	1329	....	.....	.....	.....	....	....	....
1600	.434	.49	1506	1642	67.6	0.785	1.65	4.80	.194	1062
1800	.426	.48	1680	1859	74.1	0.785	2.90	4.80	.213	1222
2000	.418	.47	1851	2075	80.5	0.785	4.81	4.80	.232	1390
2200	.411	.46	2018	2288	86.9	0.785	7.60	4.80	.251	1556
2400	.404	.45	2180	2497	92.9	0.785	11.5	4.80	.269	1730
2600	.397	.44	2339	2705	99.1	0.785	16.9	4.80	.287	1901
2800	.390	...	2495	2911	105.0	0.785	24.2	4.80	.304	2080
3000	.384	...	2647	....	.....	.....	.....	....	....	....
3300mp	.375	...	2870	....	.....	.....	.....	....	....	....

*As determined by Forsythe.

Thermal expansion. The data for a particular run are shown in Fig. 4. The curve is represented with considerable accuracy by the equation

$$\frac{\Delta l}{l} = 6.60\times10^{-6}(T-300°)+5.2\times10^{-10}(T-300°)^2,$$

For 0°C this yields a coefficient of 6.57×10^{-6} deg.$^{-1}$. This is in good agreement with Disch's[12] equation for the range $0°C<t<400°C$, obtained by the Fizeau method,

$$\frac{\Delta l}{l}=6.46\times10^{-6}t+9.0\times10^{-10}t^2.$$

Collected data. In Table IV data on properties of tantalum between room temperature and the melting point where known with reasonable certainty are shown at 200° intervals.

The writer is greatly indebted to Lester Boss, Ruth Sublett Haas, C. H. Prescott Jr. and Pauline Perkins for aid in carrying out the experimental work.

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NELA PARK, CLEVELAND,
September, 1925.

[12] Disch, Z. Physik, **5**, 173, 1921.