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Spectral Emissivity, Total Emissivity, and Thermal Conductivity of Molybdenum, Tantalum, and Tungsten above 2300°K

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The emissivities and thermal conductivities of high purity arc melted molybdenum and tantalum and powder metallurgy tantalum and tungsten were measured between 2300°K and their respective melting points. A method is presented for the determination of spectral and total emissivities from the determination of brightness temperature at the center of an electrically heated rod as a function of heat flow rate. The method does not depend on the experimental achievement of blackbody conditions. Spectral emissivities were also determined by measurement of brightness temperatures at the respective melting points. Thermal conductivities of electrically heated rods were determined by the Jain and Krishnan longitudinal heat flow method. The spectral emissivities of tantalum and tungsten decrease linearly with increase in temperature. The spectral emissivity of molybdenum is constant. The total emissivities of molybdenum, tantalum, and tungsten increase with increasing temperature. For any of the metals the spectral and total emissivities are closest to each other at the melting point. The melting point of tantalum is significantly lowered by small increase in impurity content. The thermal conductivities of molybdenum, tantalum, and tungsten decrease linearly with increasing temperature. The ranges of spectral emissivity, total emissivity, and thermal conductivity above 2300°K are as follows:

	ϵ_λ	ϵ_T	K (cal. cm^{-1} sec.$^{-1}$ °K^{-1})
Molybdenum	0.30	0.271–0.285	0.34–0.32
Tantalum	0.361–0.350	0.288–0.324	0.15–0.11
Tungsten	0.37 –0.36	0.268–0.352	0.38–0.30

I. INTRODUCTION

THE emissivities and thermal conductivities of refractory metals must be known to permit their intelligent use in high-temperature missile applications such as nozzles and jetevators. For instance, the temperature profile reached by a particular material is dependent upon its thermal conductivity and the rate at which heat flows through it; the heat flow rate, in turn, is dependent upon the emissivity (absorptivity) of the material.

Emissivity and thermal conductivity data in the literature show appreciable discord. Furthermore, calculation of thermal conductivity usually depends upon knowledge of spectral and total emissivity to permit necessary temperature corrections and heat flow rate determinations. Thus, it is desirable to measure emissivities and thermal conductivity on the same materials.

TABLE I. Material designations and details of manufacture of molybdenum, tantalum, and tungsten.

Material designation	Manufacturer	Methods of manufacture
Molybdenum	Fansteel Corp.	Cast, under inert gas, in watercooled copper mold using consumable electrode. Hot worked and hot rolled.
Tantalum No. 1	Fansteel Corp.	Pressed and sintered metal powder. Hot and cold rolled.
Tantalum No. 2	National Research Corp.	Cast, in vacuum, in water-cooled copper mold using consumable electrode. Cold rolled, swaged and cold drawn.
Tungsten No. 1	Fansteel Corp.	Pressed and sintered metal powder. Hot worked.
Tungsten No. 2	Wah Chang Corp.	Pressed and sintered metal powder. Hot worked.

A new method for the determination of spectral and total emissivities from a single experimental procedure, involving the determination of brightness temperature as a function of heat flow rate, has been applied for the first time. Spectral emissivities have also been separately determined by measurement of the brightness temperatures of the respective metals at melting.

Thermal conductivities have been determined by the Jain and Krishnan[1] method. This technique is based upon the experimental determination of the distribution of temperature along a thin rod which is electrically heated. The method assumes that heat is radiated from the rod's surface to an effectively black environment.

II. MATERIALS

All the materials were reasonably typical of commercial purity. The molybdenum was made by arc melting, under inert gas; the tantalum materials were produced by powder metallurgy procedures and vacuum arc melting; and the tungsten materials were produced by powder metallurgy procedures only. The material designations, details of manufacture, and chemical analyses are supplied in Tables I and II.

III. DEFINITIONS AND THEORY

The total emissivity of a radiating body is the ratio of the heat flow rate from that body to the heat flow rate from a blackbody of the same dimensions at the same absolute temperature. Total emissivity, ϵ_T, may also be defined by the equation

$$\epsilon_T = Q/\sigma A(T_m^4 - T_e^4), \tag{1}$$

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[1] S. C. Jain and K. S. Krishnan, Proc. Roy. Soc. (London) A222, 167–180 (1954).

TABLE II. Chemical analyses of molybdenum, tantalum, and tungsten.

Material designation	Fe	Mn	Ti	Ni	Si	Cu	Nb	C	O_2	N_2	Mo	Ta	W	Others
Molybdenum	0.18	0.036	···	···	0.073	···	···	0.04	0.005	···	bal.	···	···	0.01
Tantalum No. 1	0.005	···	···	···	<0.02	···	···	0.0008	···	···	0.003	bal.	···	0.052
Tantalum No. 2	0.0028	···	···	···	···	···	0.0035	0.0016	0.0032 to 0.0050	<0.0010	···	bal.	···	0.0175
Tungsten No. 1	0.004	···	0.005	0.005	···	···	···	···	0.006	···	0.04	···	bal.	0.027
Tungsten No. 2	···	···	···	···	···	0.002	···	···	···	···	0.04	···	99.95	0.008

where Q is the heat flow rate, A is the area through which the heat flows, σ is the Stefan-Boltzmann constant, T_m is the temperature of the radiating element in degrees Kelvin, and T_e is the temperature of the environment in degrees Kelvin. In practice, T_e may be neglected if the environmental temperature is low.

Spectral emissivity, ϵ_λ, is defined by Wien's law:

$$\ln\epsilon_\lambda = C_2/\lambda\{1/T - 1/T'\}, \tag{2}$$

where C_2 is the second constant of Planck's law, λ is the wavelength of the pyrometer filter, T is the true temperature, and T' is the brightness temperature. The spectral emissivity must be determined in order to convert an experimental brightness temperature into a true temperature, since pyrometric temperature measurement is carried out at a single effective wavelength.[2]

Spectral emissivity may be expressed as a function of brightness temperature, heat flow rate, and the derivative of brightness temperature with respect to heat flow rate. The derivation is given below.

Neglecting the environmental temperature T_e, Eq. (1) becomes

$$Q = \epsilon_T \sigma A T^4. \tag{3}$$

By substituting Eq. (3) in Eq. (2)

$$\ln\epsilon_\lambda = \frac{C_2}{\lambda\{Q/\epsilon_T\sigma A\}^{\frac{1}{4}}} - \frac{C_2}{\lambda T'}. \tag{4}$$

By partial differentiation,

$$\frac{\partial T'}{\partial Q} = \frac{(\epsilon_T\sigma A)^{\frac{1}{4}}(T')^2}{4Q^{5/4}}, \tag{5}$$

$$\frac{\partial T'}{\partial \epsilon_T} = -\frac{(\sigma A)^{\frac{1}{4}}(T')^2}{4Q^{\frac{1}{4}}\epsilon_T{}^{\frac{3}{4}}}, \tag{6}$$

$$\frac{\partial T'}{\partial \epsilon_\lambda} = \frac{(T')^2\lambda}{\epsilon_\lambda C_2}. \tag{7}$$

On combining the preceding,

$$dT' = \frac{(\epsilon_T\sigma A)^{\frac{1}{4}}(T')^2}{4Q^{5/4}}dQ - \frac{(\sigma A)^{\frac{1}{4}}(T')^2}{4Q^{\frac{1}{4}}\epsilon_T{}^{\frac{3}{4}}}d\epsilon_T + \frac{(T')^2\lambda}{\epsilon_\lambda C_2}d\epsilon_\lambda. \tag{8}$$

Upon factoring (8) and upon substituting from (3),

$$\frac{dT'}{(T')^2} = \frac{dQ}{4TQ} - \frac{d\epsilon_T}{4T\epsilon_T} + \frac{\lambda d\epsilon_\lambda}{C_2\epsilon_\lambda}. \tag{9}$$

Since the sum of the second and third terms on the right is small relative to the first term, the equation may be written

$$1/T = [4Q/(T')^2][dT'/dQ]. \tag{10}$$

By substituting in (2),

$$\ln\epsilon_\lambda = \frac{C_2}{\lambda}\left[\frac{4Q}{(T')^2}\left(\frac{dT'}{dQ}\right) - \frac{1}{T'}\right]. \tag{11}$$

Equations (3) and (11) permit solutions for ϵ_T and ϵ_λ. Their approximate rates of change with heat flow rate or temperature can be readily calculated by solutions at a number of power levels.

If necessary, a complete solution of Eq. (9) can now be obtained. On dividing, it becomes

$$\frac{1}{(T')^2}\frac{dT'}{dQ} = \frac{1}{4TQ} - \frac{1}{4T\epsilon_T}\frac{d\epsilon_T}{dQ} + \frac{\lambda}{C_2\epsilon_\lambda}\frac{d\epsilon_\lambda}{dQ}. \tag{12}$$

This may be rewritten

$$\frac{1}{(T')^2}\frac{dT'}{dQ} = \frac{1}{4TQ} + \alpha; \tag{13}$$

therefore,

$$\frac{1}{T} = 4Q\left[\frac{1}{(T')^2}\frac{dT'}{dQ} - \alpha\right], \tag{14}$$

and

$$\ln\epsilon_\lambda = \frac{C_2}{\lambda}\left[4Q\left(\frac{1}{(T')^2}\frac{dT'}{dQ} - \alpha\right) - \frac{1}{T'}\right]. \tag{15}$$

Successive solutions of Eqs. (15) and (3) will lead to successively more accurate determinations of α, ϵ_λ, and ϵ_T. The relaxation procedure should be carried out

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[2] The single effective wavelength is an average over a narrow band in the visible spectrum. The spectral emissivity shows negligible variation over the width of this band at a given temperature.

at several power levels since α is a function of several temperature dependent variables. Finally, ϵ_λ and ϵ_T can be plotted as functions of temperature.

The thermal conductivity of a substance, a numerical quantity which specifies its ability to conduct heat, is defined as the proportionality constant k in the equation

$$Q=-kA(dt/dx), \tag{16}$$

where Q is the heat flow rate, A is the area through which the heat passes, and dt/dx is the temperature gradient in a direction perpendicular to A. This equation may be used to determine the thermal conductivity of a rod-shaped specimen with a measurable temperature gradient parallel to its length, a constant heat flow rate perpendicular to all cross sections and a constant temperature parallel to any one cross section. In practice, such conditions are extremely difficult to obtain.

The Jain and Krishnan method makes possible the determination of the thermal conductivity of a long rod electrically heated between electrode-holders. Thermal conductivity is calculated from temperatures measured along the length of the rod. The following equation applies:

$$k=\frac{16\epsilon_T\sigma T_m{}^3x^2}{D\left[\ln\tau+\dfrac{\tau}{2T_m}+\dfrac{\tau^2}{16T_m{}^2}-\dfrac{\tau^3}{240T_m{}^3}-\ln t-\dfrac{t}{2T_m}-\dfrac{t^2}{16T_m{}^2}-\dfrac{t^3}{240T_m{}^3}\right]^2}, \tag{17}$$

where ϵ_T is the total emissivity, σ is the Stefan-Boltzmann constant, D is the diameter of the specimen, T_m is the temperature at the center, τ is the temperature difference between the center and end of the rod, and t is the temperature difference between the center and a point x distance from the end of the rod. Measured brightness temperatures must be corrected to true temperatures with the appropriate spectral emissivities. All temperatures are in degrees Kelvin. In practice, the cube terms in the denominator of Eq. (17) are usually small enough to be omitted.

The principal theoretical shortcoming of the method is that it does not take into account the radial component of the temperature gradient within the rod. This radial component, however, will be relatively less important if the test material has a positive temperature coefficient of resistivity. (Molybdenum, tantalum, and tungsten have positive coefficients.)

IV. EQUIPMENT AND PROCEDURES

Experimental determinations of total and spectral emissivity and thermal conductivity were carried out in a Pyrex bell jar in a protective atmosphere of argon or in partial vacuum. The base plate was machined from a glass-bonded synthetic mica. Test temperatures were achieved by direct current electrical self resistance heating of test specimens. Power leads, voltage measurement leads, and atmosphere control ports through the base were sealed with epoxy cement.

Test specimens were 0.125 in. in diam and 10 in. long. They were successively polished with No. 1-, 0-, 00-, 000-, and 0000-abrasive papers. Each specimen was connected in a vertical position between a rigid copper bus bar at the top and a flexible copper lead at the bottom (to allow for thermal expansion). The vertical distance between electrical connections was 8 in. Two voltage leads of 0.020-in. diam tungsten wire were spot welded 1 in. apart, symmetrically on either side of the center of each emissivity specimen.

Direct current was supplied by a 60-v 2000-amp motor generator set. Temperatures were measured with a disappearing filament pyrometer. Observed temperatures were corrected to brightness temperatures with an experimental "window factor." Electrical measurements were made with a recording dc ammeter and a vacuum tube voltmeter. Measurements of temperature and electrical quantities were not recorded until a steady state had been reached. After any change in electrical power setting, continuous readings of the center temperature were made until it stopped changing.

Oxidation and/or sublimation of molybdenum, tantalum, and tungsten were difficult to overcome. The most damaging consequence of these reactions was accumulation of a translucent deposit on the bell jar wall. Such a deposit seriously lowered the observed temperatures and decreased the quantity of radiant energy leaving the jar. To reduce the sublimation and subsequent deposition, emissivity runs on molybdenum were carried out in a partial pressure of argon; the inert gas flowed through the jar at the rate of 10 cu ft/hr. Emissivity runs on tungsten and tantalum and all thermal conductivity runs were carried out in argon at a pressure slightly greater than one atmosphere. (The argon flowed through the jar at the rate of 10 cu ft/hr and bubbled through 0.1 in. of water at the system's exit.)

The test procedures for emissivity were of two types: (1) measurement of brightness temperature at the melting point by slowly increasing power and closely following the sample temperature until melting broke the circuit. This procedure was carried out a minimum of three times for each metal. (2) Simultaneous measurements of brightness temperature, current, and potential drop across the test section at a series of power levels. These readings were recorded at 10-amp intervals within the brightness temperature range between 1500°C and 2800°C.

Spectral and total emissivities were calculated with the following steps:

1. The spectral emissivity for 0.65 μ was obtained at each melting point[3] by substitution of brightness and true temperatures into Eq. (2).
2. Heat flow rates at the various power levels were calculated from Eq. (18),

$$Q=JEI \qquad (18)$$

in which Q is the heat flow rate, J is a conversion factor, E is potential drop, and I is current.

3. The spectral emissivities (0.65 μ) and total emissivities at temperatures below melting were obtained from Eqs. (3), (11), and (15) according to the procedures described under "Definitions and Theory."

The test procedures for thermal conductivity are described below. At each power level, temperature readings were obtained at the center of the specimen, and distance readings were obtained at a point 100°C cooler than the center. The approximate temperature of the upper copper clamp was determined with a chromel-alumel thermocouple, immediately after each power shutoff.

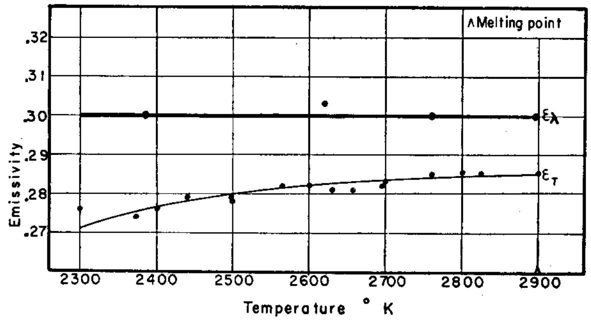

FIG. 1. Total and spectral emissivities (at 0.65 μ) of molybdenum above 2300°K.

In detail, a temperature was first read at the center. Then the pyrometer reading was decreased by 100°C and the pyrometer rotated through a vertical angle until a match was obtained with the specimen at some point between center and end. The linear distance between the latter point and the end was measured by observation, through the pyrometer, of a stainless steel scale affixed to the outside of the bell jar. This scale reading was corrected with a calibration curve which had been obtained by substituting a second steel scale for the sample and correlating the corresponding readings from the two scales. The measuring procedure was repeated several times to obtain an average temperature at the center and an average location of a temperature 100°C less. The power was then turned off and the temperature of the sample grip immediately determined with a thermocouple. The true temperature of the clamp was obtained by extrapolation from a temperature-time curve established from successive readings during cooling.

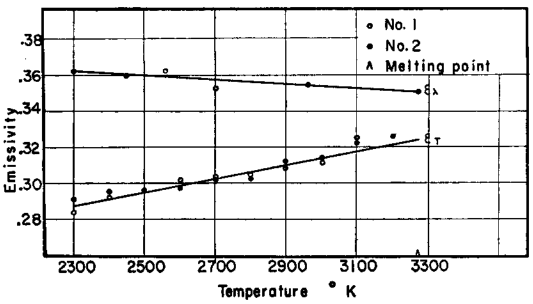

FIG. 2. Total and spectral emissivities (at 0.65 μ) of tantalum above 2300°K.

Thermal conductivities were calculated from Eq. (17).

V. RESULTS AND DISCUSSION

Total and spectral emissivities are plotted vs true temperature in Figs. 1–3. The total emissivities of molybdenum, tantalum, and tungsten increase with increasing temperature. Total emissivities of tantalum No. 1, tantalum No. 2, and tungsten No. 1 are linear functions of temperature over the respective temperature intervals from 2300°K to melting. The total emissivity of tungsten No. 2 increases with temperature at an increasing rate. The total emissivity of molybdenum increases with temperature at a decreasing rate from 2300°K to melting.

The spectral emissivities of tantalum and tungsten decrease with increase in temperature between 2300°K and the respective melting points; the relationships are linear. The rates of variation with temperature are appreciably less than those determined for total emissivity. The spectral emissivity of molybdenum is constant, within limits of experimental error, between 2300°K and the melting point.

For any one of the three metals, the spectral and total emissivities are closest to each other at the melting point.

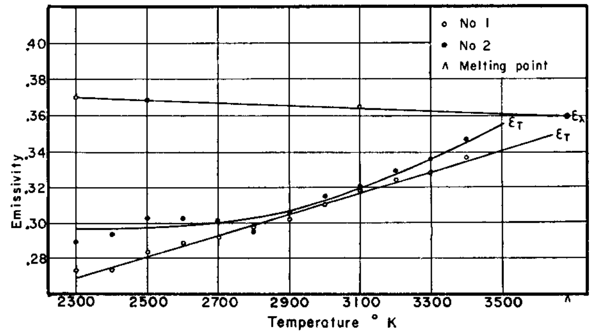

FIG. 3. Total and spectral emissivities (at 0.65 μ) of tungsten above 2300°K.

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[3] Melting points were taken from *Metals Handbook*, edited by Taylor Lyman (The American Society for Metals, 1948), pp. 20–21.

Tantalum No. 1 and tantalum No. 2 yielded emissivities which were identical within experimental error, in the temperature region below melting; this was true of both spectral and total emissivity. The brightness temperature of tantalum No. 1 at the melting point was 43°C less than the brightness temperature of No. 2. This discrepancy is probably indicative of an actual difference in melting point. The lower purity of tantalum No. 1 supports this interpretation of the data.

The mathematical analyses employed to determine spectral emissivity at temperatures below the melting point and total emissivities at all temperatures assume that all energy supplied to a sample within the central portion is dissipated by radiation. Two departures from this condition are considered: (1) convective heat transfer from the specimen to the inert gas flowing through the bell jar and (2) conductive heat transfer from the specimen through the voltage leads. The percentage of heat transferred to the gas was very small. A calorimetric calculation showed that less than 0.5% of the total heat generated in the test section at 2500°C was removed by convection. Calculations based on temperature profiles along the voltage leads indicated that only about 0.5% of the heat generated within the test section was removed by conduction through the leads. The amount of heat lost by conduction through the voltage leads was sufficient, however, to produce a slight temperature drop at the points where these were spot-welded to the specimen. The temperature drop from midway between the voltage leads to either lead averaged less than one percent. Temperature profiles along the test section indicated that the temperature at the midpoint did not differ from the mean temperature by more than 0.3%.

TABLE III. Total emissivities of molybdenum.

Temperature (°K)	This investigation	Worthing[a]
2400	0.277	0.248
2600	0.282	0.265
2800	0.285	0.281
2895	0.286	0.290

[a] See footnote reference 4.

TABLE IV. Total emissivities of tantalum.

Temperature (°K)	This investigation	Worthing[a]
2400	0.292	0.269
2600	0.300	0.287
2800	0.308	0.304

[a] See footnote reference 4.

TABLE V. Total emissivities of tungsten.

Temperature (°K)	This investigation No. 1	This investigation No. 2	*American Institute of Physics Handbook*, 1957
2400	0.277	0.294	0.296
2600	0.289	0.298	0.311
2800	0.300	0.304	0.323
3000	0.312	0.314	0.334
3200	0.323	0.329	0.341
3400	0.335	0.347	0.348
3600	0.347	···	0.354

The total emissivity data from this investigation are in reasonable accord with published values. Worthing[4] determined the total emissivities of molybdenum and tantalum from experimental measurements of radiation intensity at various temperatures. His results are compared with this investigation in Tables III and IV. Comparative tungsten data are given in Table V.

The spectral emissivities (at $0.65\,\mu$) determined during the present investigation are somewhat lower than values published by Worthing.[4,5] The differences are least at the melting points of the metals and greatest at the lowest temperature considered (2300°K).

In order to theorize about these discrepancies, it is necessary to briefly describe Worthing's method. Tubular specimens with openings in the walls were electrically heated and outside and inside temperatures were determined with a disappearing filament pyrometer. To determine the actual temperature difference between inside and outside walls, thermal conductivity, electrical resistivity, and current density were substituted into the well-known Angell equation

$$\Delta T=\frac{i^2\rho}{2K}\left(r_i\ln\frac{r_0}{r_i}-\frac{r_0^2-r_i^2}{2}\right), \tag{19}$$

where ΔT is the temperature difference between inner and outer surfaces, i is the current density, ρ is the resistivity, k is the thermal conductivity, r_i is the internal radius, and r_0 is the external radius. The temperature difference was subtracted from the inside temperature to obtain the true outside temperature.

Spectral emissivity was obtained from Eq. (2) by substitution of the outside brightness and true temperatures. The method assumes that blackbody conditions obtain at the inside temperature stations.

Worthing[6] has mentioned three factors which produce high emissivities: (1) use of poorly polished specimens, (2) insufficient approach to blackbody conditions, and (3) use of impure specimens. Without question, the tube method of determining spectral emissivity is subject to error due to departure from perfect blackbody conditions. Furthermore, departure from blackbody conditions within a tube is probably greater at lower temperatures because the length of the constant temperature zone is a function of temperature; therefore, an inside temperature station "sees" a larger cold area at low temperature than at high. For this

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[4] A. G. Worthing, Phys. Rev. **28**, 190–210 (1926).

[5] A. G. Worthing, Phys. Rev. **10**, 377–394 (1917).

[6] A. G. Worthing, Phys. Rev. **25**, 846–857 (1925).

reason, higher apparent spectral emissivities would be obtained at lower temperatures.

The method employed in this investigation does not depend on the experimental achievement of blackbody conditions.

The comparative spectral emissivity data are presented in Tables VI–VIII.

Thermal conductivities of molybdenum, tantalum, and tungsten decrease with increasing temperature from 2300°K to their respective melting points. Thermal conductivity temperature curves for all three metals are linear with relatively slight slopes. The data are shown in Figs. 4–6. Single curves obtain for both tantalum materials and both tungsten materials. Thus, differences in purity were insufficient to produce measureable differences in thermal conductivity.

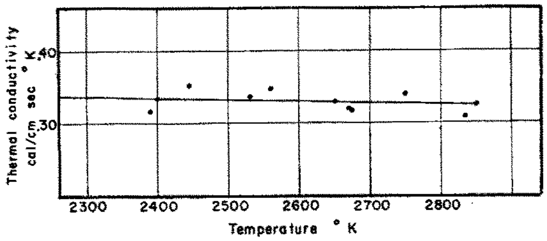

FIG. 4. Thermal conductivity of molybdenum above 2300°K.

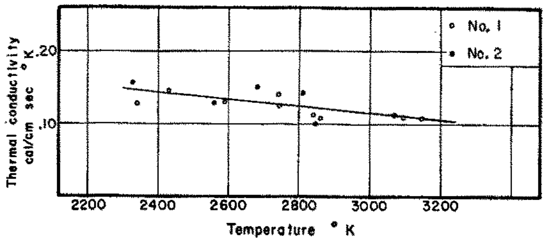

FIG. 5. Thermal conductivity of tantalum above 2300°K.

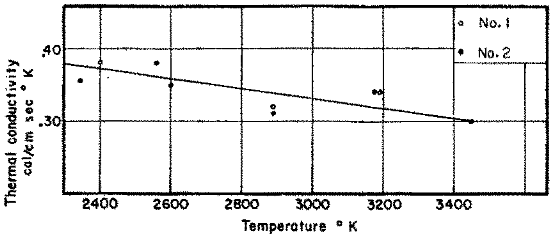

FIG. 6. Thermal conductivity of tungsten above 2300°K.

Very limited comparison of these thermal conductivities with published data has been made because of the paucity of data on very pure metals at these very high temperatures. Available data are characterized by inconsistencies among the reported magnitudes and temperature coefficients of thermal conductivity. For instance, thermal conductivities of molybdenum reported by Zuikker[7] are not in agreement with values reported by Rasor and McClelland[8] either with respect to magnitude or temperature dependence. The thermal conductivities determined in this investigation are in better agreement with Zuikker's results.

TABLE VI. Spectral emissivities (0.65 μ) of molybdenum

Temperature (°K)	This investigation	Worthing[a]
2400	0.30	0.341
2600	0.30	0.336
2800	0.30	0.331
2898	0.30	0.328

[a] See footnote reference 4.

TABLE VII. Spectral emissivities (0.65 μ) of tantalum.

Temperature (°K)	This investigation	Worthing[a]
2400	0.361	0.404
2600	0.358	0.397
2800	0.356	0.390
3000	0.353	0.384
3269	0.350	0.375

[a] See footnote reference 4.

TABLE VIII. Spectral emissivities (0.65 μ) of tungsten.

Temperature (°K)	This investigation	Worthing[a]
2400	0.370	0.429
2600	0.368	0.425
2800	0.367	0.420
3000	0.365	0.415
3200	0.364	0.410
3400	0.363	0.405
3683	0.360	0.397

[a] See footnote reference 4.

Similar discrepancies exist among the published thermal conductivities for tantalum and tungsten.

VI. CONCLUSIONS

1. The total emissivities of molybdenum, tantalum, and tungsten increase with temperature, usually at a decreasing rate, between 2300°K and the respective melting points.
2. The spectral emissivities of tantalum and tungsten decrease linearly with temperature between 2300°K and the respective melting points. The spectral emissivity of molybdenum is constant within this temperature span.
3. The thermal conductivities of molybdenum, tantalum, and tungsten decrease linearly with temperature from 2300°K to the respective melting points.

VII. ACKNOWLEDGMENTS

This work was carried out under U. S. Navy contract. Technical assistance was rendered by D. R. Blair and A. H. Hussung.

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[7] C. Zuikker, Physica 7, 71–74 (1927).

[8] N. S. Rasor and J. D. McClelland, WADC Tech. Rept. 56–400, Pt. I (1957).