Thermophysical Properties of Tantalum, Tungsten, Rhenium, and Their Alloys in the Temperature Range 1500–3000 °K*

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Dedicated to Professor August Guyer

Abstract

The thermal conductivities and total emittances of tantalum, tungsten, rhenium, and their alloys, Ta-10W, T_{111}, T_{222}, and W-25 Re, were measured in the temperature range 1500 to 3000 °K. From data in the literature, the high-temperature specific heats of these materials were computed.

Introduction

There is a great need for accurate thermophysical data on the high-temperature refractory metals and their alloys. This paper reports measurements on the thermal conductivities and total emittances of tantalum, tungsten, rhenium, and Ta-10W, T_{111}, T_{222}, and W-25 Re alloys in the temperature range 1500–3000 °K. Also, the

high-temperature specific heats of tungsten, tantalum, and rhenium are evaluated.

Experimental Method

Thermal Conductivity

The temperature distribution on the flat surface of a cylindrical disc heated in vacuum by induction has been used by HOCH et al.[1] to determine the thermal conductivity of solid materials. The following equation applies:

$$K = \frac{\varepsilon \sigma T_0^4}{4\left(\dfrac{dT}{dr^2}\right) a C^0}, \qquad (1)$$

where:
K = thermal conductivity
ε = total emittance
σ = Stefan-Boltzmann radiation constant
T = temperature on the flat surface, function of r
T_0 = temperature at center of flat circular surface
a = radius of the specimen
C^0 = constant, depending on the height-to-diameter ratio of the specimen
r = independent radial variable

This equation has been applied earlier to various materials (VARDI and HOCH,[2] HOCH and VARDI,[3] JUN and HOCH,[4] and HOCH and JUN[5]).

Total Emittance

When a specimen is heated to high temperature and then cooled in vacuum, the specimen loses heat from the surface to the surroundings only by radiation. From the power balance, NARASIMHAMURTY et al.[6] derived the relationship

$$\frac{Cp}{\varepsilon} = \frac{3\sigma A}{m} \cdot \frac{d\theta}{d(1/T^3)}, \qquad (2)$$

where:
Cp = specific heat
θ = time
A = area of the sample
m = mass of the sample

Equation (2) requires the knowledge of the specific heat to obtain the total emittance.

[1] M. HOCH, D. A. NITTI, C. F. GOTTSCHLICH, and P. E. BLACKBURN, New Method for the Determination of Thermal Conductivities Between 1000 and 3000 °C, *Progress in International Research on Thermodynamic and Transport Properties*, Academic Press, New York 1962, p. 512–8.
[2] J. VARDI and M. HOCH, Thermal Conductivity of Anisotropic Solids at High Temperatures—The Thermal Conductivity of Molded and Pyrolytic Graphites, *J. Amer. Cer. Soc. 46* (1963) 129–32.
[3] M. HOCH and J. VARDI, Thermal Conductivity of TiC, *J. Amer. Cer. Soc. 46* (1963) 245.
[4] C. K. JUN and M. HOCH, The Thermal Conductivity of Molybdenum at Elevated Temperatures: Influence of Grain Size, *Advances in Thermophysical Properties at Extreme Temperatures and Pressures*, The American Society of Mechanical Engineers, New York 1965, p. 296–300.
[5] M. HOCH and C. K. JUN, The Thermal Conductivity of TiB₂, ZrB₂, HfB₂, and NbB₂ at Elevated Temperatures, NPL *Thermal Conductivity Conference, Teddington (England) 1964.*
[6] H. V. L. NARASIMHAMURTY, A. S. IYER, and M. HOCH, Relation Between Specific Heat and Total Emittance in Tantalum, Niobium, Tungsten, and Molybdenum, *J. Physic. Chem. 69* (1965) 1420–3.

Specific Heat

If the Debye theory holds for solids at elevated temperatures, the heat content of the solid can be expressed as:[7]

$$H_T = A + BT + CT^2 + H_f \exp\left(\frac{S_f}{R} - \frac{H_f}{RT}\right), \qquad (3)$$

where:
H_T = heat content
A, B, C = constants
H_f, S_f = heat and entropy of formation, respectively, of a vacancy in the metal
R = gas constant

The constants A, B, and C can be evaluated approximately from theoretical considerations. Differentiating Equation (3) with respect to temperature gives the specific heat. As pointed out earlier,[8] specific heat data obtained by calorimetric and by transient techniques do not agree; only calorimetric data which are more direct will be used to evaluate the constants in Equation (3). As also pointed out earlier,[7] the best method of evaluating the constants in Equation (3) is by computation of the mean specific heat.

$$\overline{Cp} = \frac{H_T - H_{298}}{T - 298} \quad \text{or} \quad \overline{Cp} = \frac{H_T - H_{273}}{H - 273}.$$

This computation of the mean specific heat rather than the true specific heat is advantageous, first, because it is a very sensitive function, and second, because for every experimental point using a drop calorimeter (which gives $H_T - H_{298}$ or $H_T - H_{273}$) one obtains one value for the mean specific heat. On the other hand, to obtain the true specific heat, $Cp = dH/dT$, the H_T vs T curve has to be evaluated mathematically and then differentiated with respect to T. In this way all points must be taken into consideration. The computation of the mean specific heat also permits the evaluation of the heat and entropy of formation of a vacancy.[9]

Equipment and Experimental Procedure

The equipment and experimental procedure have been described earlier.[2, 6]

Materials

The tantalum and tungsten were obtained from the Fansteel Metallurgical Corporation, North Chicago (Illinois). The Ta-10W, T_{111}, and T_{222} samples were supplied by the Thermophysics Branch, Air Force Materials Laboratory, Research and Technology Division, Wright-

[7] M. HOCH, High Temperature Specific Heat of Refractory Metals and Alloy, *Wright-Patterson Air Force Base, Air Force Materials Laboratory*, AFML-TR-66-360, November 1966.
[8] M. HOCH, Difference in Specific Heat Data Obtained by Transient Techniques and Calorimetric Methods, *Fourth Thermal Conductivity Conference, San Francisco (California) 1964.*
[9] M. HOCH, The Energy and Entropy of Vacancy Formation in Body-Centered Cubic Refractory Metals, *General Electric Company, Nuclear Materials and Propulsion Operation*, GE-TM-65-12-2, November 1965.

Table 1. Chemical composition of the samples (everything in p.p.m., exept where noted)

Impurity	Tantalum 1	2 3	4 5	Ta-10 W	T_{111}	T_{222}	W	Re	W-25 Re 1	2
C	19	137, 30*	0.5	25	22	95	7	47	23	25
H	17	2.7	0.9	ND**	5	12	< 0.5			2
N	17	16	18	ND	23	ND	< 5		< 5	ND
O	17	655,114,12*	36	20	34	6	< 5	11,16	14	22
W				9.0%	7.8%	8.5%				
Hf					2.0%	2.5%				
Re									25.04%	24.5%
Fe									10	

* Data indicates segregation.
** Not detected.

Patterson Air Force Base, Ohio. The rhenium and W-25 Re samples were supplied by General Electric Company, Nuclear Materials and Propulsion Operation, Cincinnati (Ohio).

The chemical composition of the various samples is given in Table 1. Grain size, density, and ratio of specific heat to total emittance are given in Table 2.

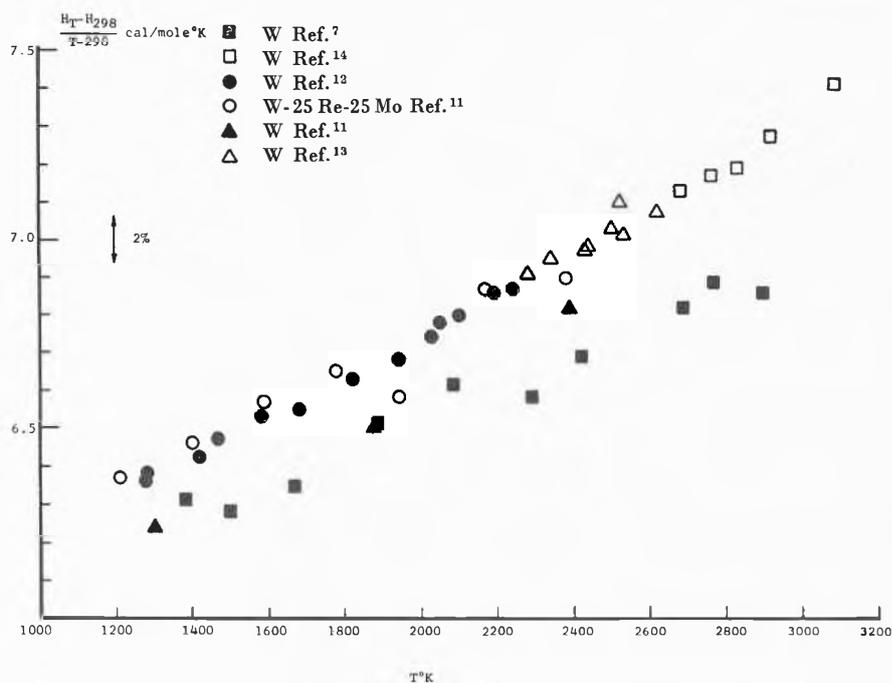## Experimental Results and Discussion

Specific Heat

The mean specific heats of tungsten and W-25 Re-25 Mo alloy are plotted in Figure 1 on a very extended scale. The agreement among the various authors[10, 11, 12, 13, 14] is very good. The figure shows that the mean specific heat is a straight line. Thus, in the temperature range considered, the temperature is too low for the last term in Equation (3) to contribute significantly. Using only

Table 2. Density, grain size, and ratio of specific heat to total emittance of the samples

Material	Sample	Density gm/cm³	Average Grain Diameter mm	Cp/ε cal/gm°K
Ta	1	16.65	0.26	0.210 ± 0.010
	2	16.63		0.195 ± 0.006
	3	16.62	1.04	0.195 ± 0.006
	4	16.63	1.23	0.210 ± 0.010
	5	16.60	1.86	0.219 ± 0.008
Ta-10 W	1	16.91		0.210 ± 0.008
	2	16.91		0.209 ± 0.006
T_{111}	1	16.73		0.199 ± 0.005
	2	16.73		0.209 ± 0.013
T_{222}	1	16.81		0.197 ± 0.004
	2	16.81		0.210 ± 0.004
Re	1	20.97	0.020	0.175 ± 0.012
W-Re	1	19.73	0.216	0.252 ± 0.004
	2	19.19	0.041	0.198 ± 0.003
W	1	18.89		0.142 ± 0.004
	2	19.03		0.153 ± 0.002
	3	19.23	0.035	0.182 ± 0.005

Fig. 1. Mean specific heat of tungsten

[10] M. HOCH and H. L. JOHNSTON, A High Temperature Drop Calorimeter. The Heat Capacities of Tantalum and Tungsten Between 1000 and 3000°K, *J. Physic. Chem.* 65 (1961) 855–60.

[11] J. B. CONWAY and R. A. HEIN, private communication.

[12] V. YA. CHEKHOVSKOY, B. YA. SHUMJATSKII, and K. A. YAKIMOVICH, The Enthalpy of Tungsten at 350–2000°C, *Inzh. Fiz. Zh. Akad. Nauk Belorussk* SSR 5 (1962) No. 10, p. 13.

[13] V. A. KIRILLIN, A. E. SHEINDLIN, and V. YA. CHEKHOVSKOY, Thermodynamic Properties of Tungsten in the Temperature Range 0–2400°C, *Teploenerg.* 9 (1962) No. 2, p. 63.

[14] V. A. KIRILLIN, A. E. SHEINDLIN, V. YA. CHEKHOVSKOY, and V. A. PETROV, Thermodynamic Properties of Tungsten in the Range 0–3500°K, *Zh. Fiz. Khim.* 37 (1963) 2249.

$\frac{H_T - H_{298}}{T - 298}$ cal/mole°K

■ W Ref.[7]
□ W Ref.[14]
● W Ref.[12]
○ W-25 Re-25 Mo Ref.[11]
▲ W Ref.[11]
△ W Ref.[13]

T°K

the data of CHEKHOVSKOY *et al.*[12] and KIRILLIN *et al.*[13, 14] one obtains the following expression for the true specific heat of tungsten:

$$Cp = 5.535 + 1.052 \times 10^{-3}\, T \text{ cal/mole } ^\circ\text{K}$$
$$1300 < T < 3000 \,^\circ\text{K}.$$

The same expression also applies to the W-25 Re alloy. From figures similar to Figure 1, the specific heat of tantalum and tantalum alloys and the specific heat of rhenium were computed as:[7]

$$\text{Ta} \quad Cp = 5.736 + 0.917 \times 10^{-3}\, T \text{ cal/mole}^\circ\text{K}$$
$$1300 < T < 2800\,^\circ\text{K},$$

$$\text{Re} \quad Cp = 5.630 + 1.080 \times 10^{-3}\, T \text{ cal/mole}^\circ\text{K}$$
$$1300 < T < 2600\,^\circ\text{K}.$$

Thermal Conductivity

The experimental results using the various samples are plotted in Figures 2 to 4. In Figure 2, all the data

Table 3. Comparison of tantalum thermal conductivity data

Author	K, watt/cm°K		
	1500°K	2000°K	3000°K
This research	0.652	0.631	0.587
KRAEV and STEL'MAKH[15]		0.631	0.606
WHEELER[16]	0.648	0.678	0.739

Table 3 gives the thermal conductivity of tantalum as calculated from Equation (4), as computed from the results of KRAEV and STEL'MAKH,[15] and of WHEELER.[16] Both used a thermal diffusivity method, and the specific heat given in this paper was used to compute the thermal conductivity data from their results. At 1500°K WHEELER's and our data agree quite well, whereas at 2000 and 3000°K KRAEV and STEL'MAKH's and our data agree. WHEELER's results increase with increasing temperature, whereas KRAEV and STEL'MAKH's and ours decrease. Earlier thermal conductivity data on tantalum has been compiled and compared with our results.[17]
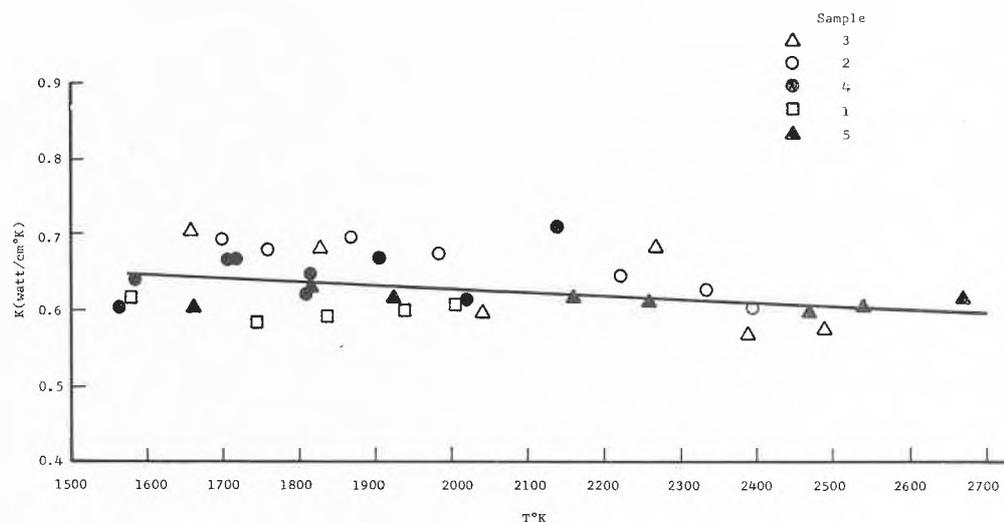

Fig. 2
Thermal conductivity
of tantalum

obtained on tantalum are represented. The best straight line obtained by least squares analysis is:

$$K = (0.718 \pm 0.006) - (0.435 \pm 0.208)$$
$$\times 10^{-4}\, T \text{ watt/cm}^\circ\text{K} \quad 1600 < T < 2700\,^\circ\text{K}. \quad (4)$$

It should be noted that all five samples of tantalum were machined from the same 1″ diameter tantalum rod. However, the samples were used differently, heated to high temperature, and several of them used in the study to show that Cp/ε^6 is constant, and finally in the thermal conductivity measurements shown here. It can be observed from Table 2 that an extremely large grain growth has occurred during these runs. The data presented in Figure 2 were obtained by two experimentalists; samples 1, 4, and 5 by one, and samples 2 and 3 by the other. Keeping all this in mind, one can see that the data are fairly consistent.

In Figure 3 are plotted the thermal conductivities of the tantalum base alloys. All the thermal conductivities are a little smaller than that of tantalum and their slopes are parallel to each other. The only larger-than-expected fluctuation is observed in the Ta-10 W in which sample No. 1 was first run, then heated to 2700°K, and the other points, marked sample No. 2, taken. This seems to indicate that sample No. 1 was not in equilibrium. The data can be represented as:

[15] O. A. KRAEV and A. A. STEL'MAKH, Thermal Diffusivity of Tantalum, Molybdenum, and Niobium at Temperatures Above 1800°K, *Teplofizika Vysokikh Temperatur 2* (1964) No. 2, p. 302.
[16] M. J. WHEELER, Thermal Diffusivity at Incandescent Temperatures by a Modulated Electron Beam Technique, *Brit. J. Appl. Physics 16* (1965) 365.
[17] C. K. JUN and M. HOCH, Thermal Conductivity of Tantalum, Tungsten, and Tantalum-Tungsten Alloys, *Wright-Patterson Air Force Base, Air Force Materials Laboratory*, AFML-TR-65-191, May 1965.
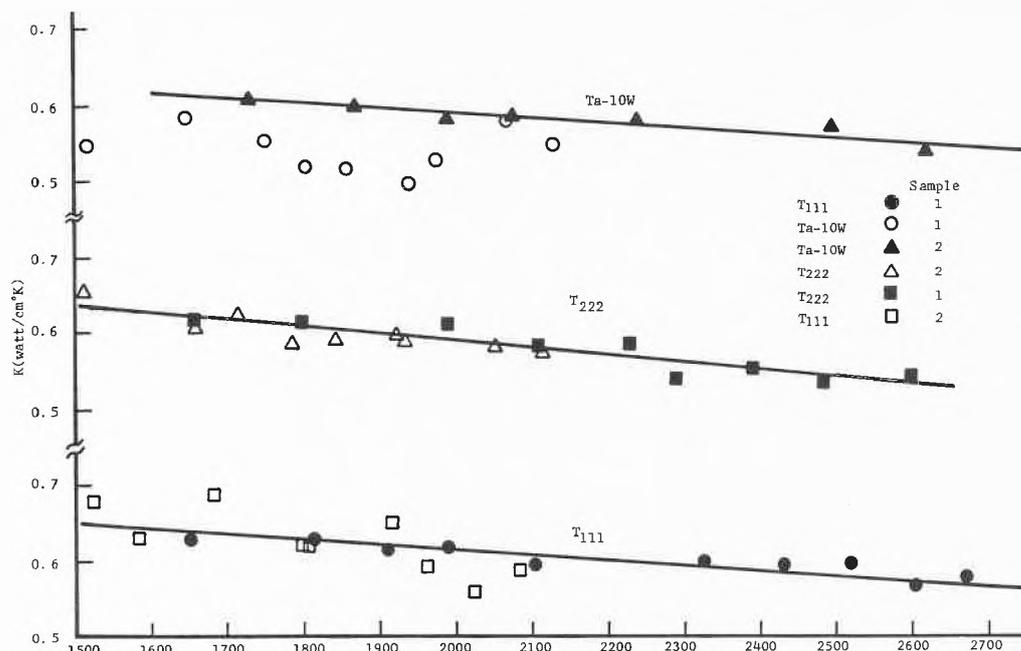
Fig. 3
Thermal conductivity
of tantalum alloys

Ta-10 W $K = (0.720 \pm 0.004) - (0.655 \pm 0.082)$
$\times 10^{-4} T \, \text{watt/cm} \, ^\circ \text{K} \quad 1600 < T < 2700 \, ^\circ \text{K},$

T_{111} $K = (0.749 \pm 0.005) - (0.675 \pm 0.141)$
$\times 10^{-4} T \, \text{watt/cm} \, ^\circ \text{K} \quad 1600 < T < 2700 \, ^\circ \text{K},$

T_{222} $K = (0.770 \pm 0,003) - (0.910 \pm 0.103)$
$\times 10^{-4} T \, \text{watt/cm} \, ^\circ \text{K} \quad 1600 < T < 2700 \, ^\circ \text{K}.$

Figure 4 presents the data on pure rhenium, tungsten, and W-25 Re alloy. The tungsten samples 1, 2, and 3

were all machined from the same tungsten piece. Sample No. 1 was repolished and machined to obtain sample No. 2, after which it was again repolished to obtain sample No. 3. Inspection of Figure 4 shows that there is a marked decrease in the thermal conductivity of tungsten as one goes from sample No. 1 to No. 2, and an especially large decrease when one goes to sample No. 3. From Table 2 it can be seen that the density increases as the sample number increases. It seems, therefore, that the thermal conductivity of tungsten is
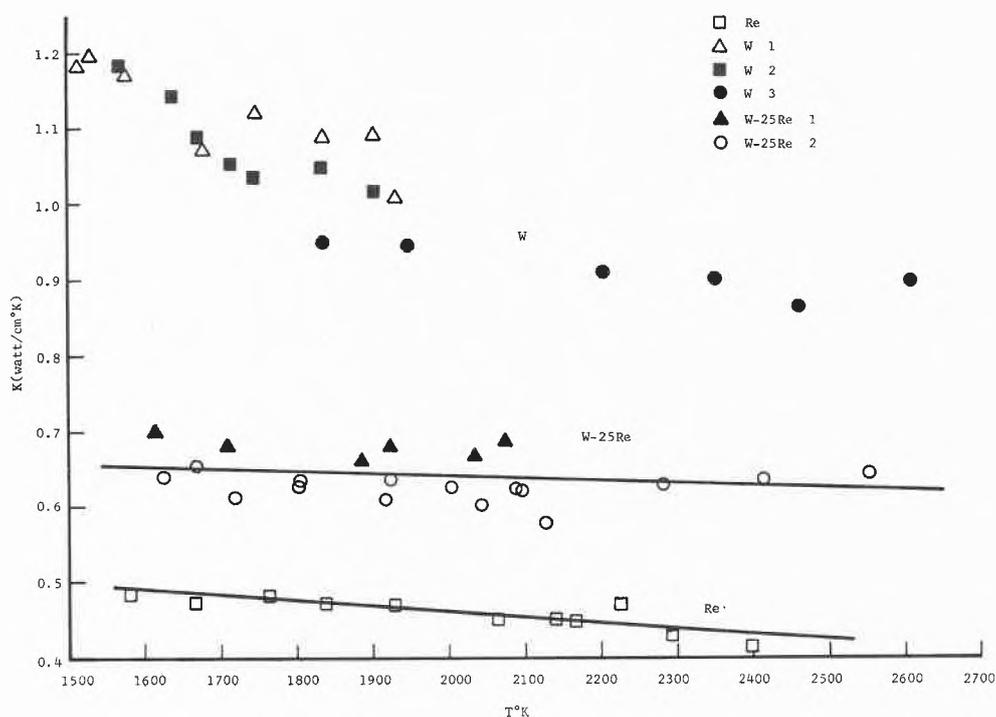

Fig. 4
Thermal conductivity of
tungsten, rhenium an W-25 Re

dependent on the density, and as the density increases, the thermal conductivity decreases. In the W-25 Re material, sample No. 2 is a powdered metallurgy sample and sample No. 1 is an arc-cast sample; the difference between the two shows up in the density and grain size. The thermal conductivities can be expressed as:

Re $\quad$ $K = (0.600 \pm 0.005) - (0.712 \pm 0.154)$
$\quad\quad\quad \times 10^{-4}T \text{ watt/cm}°K \quad 1600 < T < 2500°K,$

W $\quad$ $K = (1.109 \pm 0.008) - (0.876 \pm 0.280)$
$\quad\quad\quad \times 10^{-4}T \text{ watt/cm}°K \quad 1800 < T < 2700°K,$

W-25 Re $\quad K = (0.701 \pm 0.007) - (0.304 \pm 0.270)$
$\quad\quad\quad \times 10^{-4}T \text{ watt/cm}°K \quad 1600 < T < 2700°K.$

Table 4 gives a comparison of the authors' results on tungsten with the "most probable" values compiled by

Table 4. Comparison of tungsten thermal conductivity data

Author	K, watt/cm°K	
	2000°K	3000°K
This research	0.934	0.846
National Bureau of Standards[18]	1.00	0.91

the National Bureau of Standards.[18] The agreement is extremely good.

Acknowledgment

[18] R. W. POWELL, C. Y. HO, and P. E. LILEY, Thermal Conductivity of Selected Materials, *National Standard Reference Data Series – National Bureau of Standards – 8, Washington (D.C.) 1966.*