International Journal of Thermophysics, Vol. 15, No. 6, 1994

Measurement of Thermophysical Properties by a Pulse-Heating Method: Thoriated Tungsten in the Range 1200 to 3600 K[1]

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Thoriated tungsten (tungsten, 98%; thorium oxide, 2%) is a widely used electrode material for inert-gas arc-welding. Data for the heat capacity, electrical resistivity, and hemispherical total emissivity of this material are reported for the temperature range 1200–3600 K. A subsecond pulse-heating technique was applied to rod specimens; radiance temperature was measured by high-speed pyrometry. Literature values of the temperature dependence of the normal spectral emissivity of tungsten were used to obtain true temperatures, using the melting point of thoriated tungsten as a calibration point. Reported uncertainties for the properties are 4% for heat capacity, 1.5% for electrical resistivity, and 7% for hemispherical total emissivity.

KEY WORDS: electrical resistivity; heat capacity; hemispherical total emissivity; high temperature; tungsten; welding electrodes.

1. INTRODUCTION

Pulse techniques are considered the most accurate method for the measurement of thermophysical properties at very high temperatures, because they overcome several limitations of steady-state experiments. Thoriated tungsten is a widely used electrode material for arc-welding, with several other industrial applications due to its electron emission properties and current carrying capacity.

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[1] Paper presented at the Twelfth Symposium on Thermophysical Properties, June 19–24, 1994, Boulder, Colorado, U.S.A.

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A research group at the CSIRO Division of Applied Physics (CSIRO DAP, Sydney, Australia) is presently attempting to perform computer modeling of arc–electrode interactions in gases at atmospheric pressures. Thoriated tungsten is one of the considered electrode materials and several thermophysical properties of commercial thoriated tungsten from approximately 1200 K to the melting point are useful as input data for the modeling. A scientific cooperation was set up between the respective research groups at CSIRO DAP and at the Istituto di Metrologia "G. Colonnetti" (IMGC, Torino, Italy), within the framework of the scientific cooperation agreement between the parent organizations CSIRO (Australia) and CNR (Italy).

The high-speed apparatus available at IMGC was used to determine several thermophysical properties of commercial thoriated tungsten samples, provided by CSIRO DAP. The experimental method is based on rapid resistive self-heating of the specimen from room temperature to high temperatures in times of the order of 1 s by the passage of an electrical current pulse. Experimental quantities such as the current through the specimen, the potential drop across the central portion of the specimen, and the temperature of the specimen are measured with submillisecond time resolution. Details regarding the construction and operation of the measurement system, the method of measuring experimental quantities, and other pertinent information, such as the formulation of relations for properties, uncertainty analysis, etc., are given in earlier publications [1-3].

2. MEASUREMENTS

The main technical features of the experimental apparatus are reported in Table I. The measurements were made on three thoriated tungsten rods identified as specimens S-1 to S-3; details on the specimens are given in Table II.

Temperature measurements were made with a high-speed pyrometer [4] operating near 0.9 μm. The interference filter had a bandwidth of 80 nm and the pyrometer target area was a circle 0.3 mm in diameter. A high-speed programmable amplifier was inserted between the pyrometer and the data acquisition system (DAS) and the software programmable gain (from 1000 to 1) was decreased as necessary during the experiment to avoid saturation of the DAS. In this way the temperature range 1100–3000 K was covered in one experiment. Measurements at higher temperatures were performed with a neutral density filter (nominal transmission, 0.25) mounted on the pyrometer target channel. In this way the

Table I. Measurement Technique and System Characteristics

General technique	Pulse-heating
Voltage measurement	Across tungsten knife-edge probes
Current measurement	Across standard resistor (1 mΩ) in series with the specimen
Temperature measurement	High-speed pyrometer operating near 0.9 μm [4]
Specimen environment	Either high vacuum ($\cong 10^{-3}$ Pa) or flowing argon at atmospheric pressure
Power source	Battery bank; eight series-connected 2-V elements with a capacity of $\cong$ 1600 Ah each
Data recording	High-speed data acquisition system (14 bits; 20 kHz; range, 0–10 V; least significant bit, 0.61 mV)
Temperature range	1200–3690 K (up to the melting point of thoriated tungsten) with pyrometer autorange [5]. Two subranges were used: 1200–3000 K (up to pyrometer saturation) and 1600–3690 K with a neutral density filter

temperature range 1600–3700 K (up to the melting point of the specimen) was covered in one experiment. Additional technical details of this pyrometer autorange feature may be found in an earlier publication [5].

The high-speed pyrometer was focused on the surface of the rod specimen. Radiance temperature was measured and true temperature computed via normal spectral emissivity. Since the specimens are 98 % tungsten with some dispersed thoria (ThO_2) in the tungsten matrix, it was assumed that both the melting point and the temperature dependence of the normal spectral emissivity of thoriated tungsten are the same ones as tungsten.

Table II. The Thoriated Tungsten Specimens

Number of specimens	3
Source[a]	Teledyne Wah Chang Huntsville Inc., Huntsville, AL
Identification	Commercial grade 2% thoriated tungsten electrodes (98% tungsten, 2% thorium oxide)
Geometry	Rod form
Dimensions (nominal)	Total length, 89 mm; effective length,[b] 30 mm; diameter, 3.2 mm
Mass	Total mass, 13.8 g; effective mass,[b] 4.65 g
Characteristics	Density,[c] 18.872 $g \cdot cm^{-3}$
Special treatment	One of the specimens (S-3) was heated to high temperatures for several hours before the measurements

[a] The supplier is identified in order to characterize the specimen adequately; such identification does not imply recommendation or endorsement by IMGC or CSIRO DAP.

[b] "Effective" refers to the portion of the specimen between the voltage probes.

[c] The density measurements were performed at CSIRO DAP.

Literature data on the normal spectral emissivity of tungsten (at 900 nm) [6–8] were used to estimate the temperature dependence of the normal spectral emissivity, with a procedure previously used in similar measurements performed on tungsten [9, 10]. The emissivity curve was adjusted to the surface conditions of the specimens by using the melting plateau (assumed to be that of tungsten, 3693 K according to Ref. 11) as a calibration point. Several determinations of the melting plateau for each specimen were performed, making use of a new technique developed at IMGC, in which the specimen is brought to the melting point, but current is interrupted before the disintegration of the specimen [12]. A typical melting plateau for each specimen is presented in Fig. 1.

Previous measurements on tungsten [9, 10] were performed with two heating rates and showed no effects of the speed of the experiments on the measured properties. Consequently, experiments on thoriated tungsten were performed with one heating rate only, with the following typical conditions: experiment duration, 0.5–1.5 s; current pulse length, 0.4–0.7 s; imparted power, 3500–4900 W; current, 1200–1800 A; heating rate, 4400–6700 $K \cdot s^{-1}$, cooling rate, 120–800 $K \cdot s^{-1}$; and radiative heat loss 0.5–15% of input power.

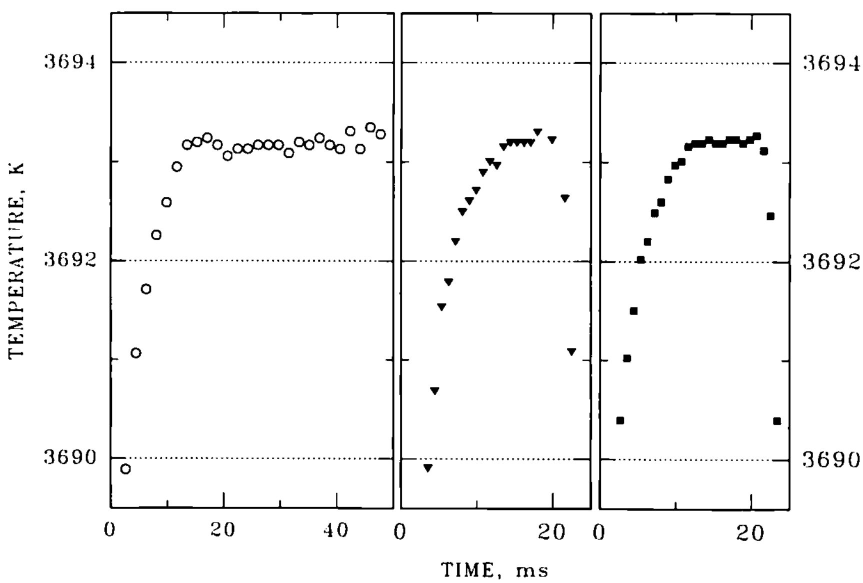

Fig. 1. Typical melting plateaux of thoriated tungsten. (○) Specimen S-1, argon; (▼) S-2; (■) S-3.

Specimens S-1 and S-2 were used as received from the manufacturer, while specimen S-3 was heat treated at temperatures higher than 3000 K for a period of more than 3 h. The heat treatment was performed at CSIRO DAP and consisted in direct current heating of the specimen held between water-cooled electrode holders under a protective atmosphere of flowing argon. The length of the heated part of the specimen was approximately 50 mm and the DC current was provided by a conventional welding power supply. Currents ranging from 300 to 420 A were used, keeping the specimen at temperatures between 2240 and 3400 K for 10 min for each current value (11 current steps). For the final heat treatment the specimen was held at 3415 K for 120 min, using a current of 420 A. The temperature in the middle of the specimen was measured with a two-wavelength pyrometry technique.

Most of the experiments were performed in high vacuum (better than 10^{-3} Pa), with no apparent major problems due to evaporation of the specimen or coatings on the chamber window. Some experiments on specimen S-1 were also performed in flowing argon at atmospheric pressure. Experimental results for specimen S-1 are presented for both environments, even if the difference in heat capacity and electrical resistivity does not appear to be significant (same order of magnitude of the reproducibility of experiments).

All the temperatures reported in this paper are based on the International Temperature Scale of 1990 [13] except where explicitly noted otherwise.

3. RESULTS

Individual experiments by the pulse method may provide up to 1000 data points per experiment, measured at random temperatures in the experimental range. Therefore, some data reduction is necessary. Thermophysical properties were computed for each experiment every 10 K, interpolating the experimental quantities (temperature, current, voltage drop). Heating rates and cooling rates were obtained by a low-order polynomial fit of temperature versus time and by differentiation of the fitting function.

The functions representing the thermophysical properties of thoriated tungsten are obtained from the data for specimens S-1 and S-2. Data from specimen S-3 are also shown in the various figures, to show that the thermophysical properties of this material after prolonged heat treatment do not differ much from those of the as-received specimens.

For heat capacity and electrical resistivity, the results of different experiments were averaged to obtain a single value for each specimen at each temperature, with the exception of specimen S-1 for which two data sets are presented, corresponding to the results obtained for measurements in high vacuum or in argon at atmospheric pressure. For hemispherical total emissivity, measurements were performed primarily on specimen S-2, and checks on specimen S-1 indicated that the same values applied to that specimen. The heat-treated specimen S-3 had different surface conditions and its hemispherical total emissivity was markedly different.

Experimental data were collected from approximately 1100 K to the melting point of tungsten, but the smoothed analytical functions representing the various properties are valid in different temperature ranges. Details are provided in the section concerning each property.

3.1. Heat Capacity (C_p)

The heat capacity of thoriated tungsten over a wide temperature range could not be represented by a low-order polynomial, on account of the heat capacity increase at high temperatures. An arbitrary separation temperature was chosen (3300 K) and the experimental results are adequately represented by two splines (third-degree polynomials reported in Table III) joining smoothly at 3300 K. The values obtained from the splines are presented in Table IV. The deviation of the heat capacity of each set of experimental values from the heat capacity of tungsten is shown in Fig. 2, along with the smooth values obtained from the two splines.

Table III. Functional Representation of the Results for Thoriated Tungsten by Fitted Polynomials

	Heat capacity ($J \cdot kg^{-1} \cdot K^{-1}$)	Heat capacity ($J \cdot kg^{-1} \cdot K^{-1}$)	Electrical resistivity ($n\Omega \cdot m$)	Hemispherical total emissivity (vacuum experiments)
Polynomial	$a+bT+cT^2+dT^3$	$a+bT+cT^2+dT^3$	$a+bT+cT^2+dT^3+eT^4$	$a+bT+cT^2$
a	133.18	-1.1560416×10^4	-205.29	6.5478×10^{-2}
b	2.5201×10^{-2}	10.655742	6.3125×10^{-1}	1.1269×10^{-4}
c	-1.1637×10^{-5}	-3.233013×10^{-3}	-2.5119×10^{-4}	-7.7165×10^{-9}
d	4.6872×10^{-9}	3.3007874×10^{-7}	8.8217×10^{-8}	
e			-1.0455×10^{-11}	
Temp. range (K)	1400–3300	3300–3600	1200–3690	2090–3600
s^a (%)	0.14		0.12	0.68

[a] Relative standard deviation (1 σ level) as computed from the difference between the average experimental values for each specimen and that from the smooth functions reported above.

The radiative heat loss term needed in the computation of heat capacity was obtained from the hemispherical total emissivity values reported in Tables III and IV. At low temperatures extrapolated values were used.

The smooth C_p functions of Table III are valid in the temperature ranges 1400–3300 and 3300–3600 K, respectively. At temperatures below 1400 K noise in the temperature measurements via pyrometry prevented the accurate computation of the heating rate. The experimental data extend up to the melting point of thoriated tungsten, but some complex phenomena occur at temperatures above 3600 K. Thermophysical properties of thoriated tungsten in this temperature range are discussed in a complementary publication.

Table IV. Smoothed Properties of Thoriated Tungsten Calculated from the Polynomials Listed in Table III

Temperature (K)	Heat capacity ($J \cdot kg^{-1} \cdot K^{-1}$)	Electrical resistivity ($n\Omega \cdot m$)[a]	Hemispherical total emissivity
1200		321.3	
1300		354.8	
1400	158.51	388.0	0.2081[b]
1500	160.62	421.2	0.2172[b]
1600	162.91	454.5	0.2260[b]
1700	165.42	488.0	0.2348[b]
1800	168.17	521.8	0.2433[b]
1900	171.20	556.1	0.2517[b]
2000	174.53	590.9	0.2600[b]
2100	178.19	626.2	0.2681
2200	182.21	662.1	0.2760
2300	186.61	698.6	0.2838
2400	191.43	735.5	0.2915
2500	196.69	772.9	0.2990
2600	202.42	810.6	0.3063
2700	208.65	848.7	0.3135
2800	215.40	886.8	0.3205
2900	222.71	924.9	0.3274
3000	230.60	962.8	0.3341
3100	239.11	1000.2	0.3407
3200	248.25	1036.9	0.3471
3300	258.06	1072.7	0.3533
3400	268.89	1107.3	0.3594
3500	282.40	1140.4	0.3654
3600	300.56	1171.6	0.3712

[a] Based on the room-temperature (293 K) dimensions of the specimen.
[b] Extrapolated value.

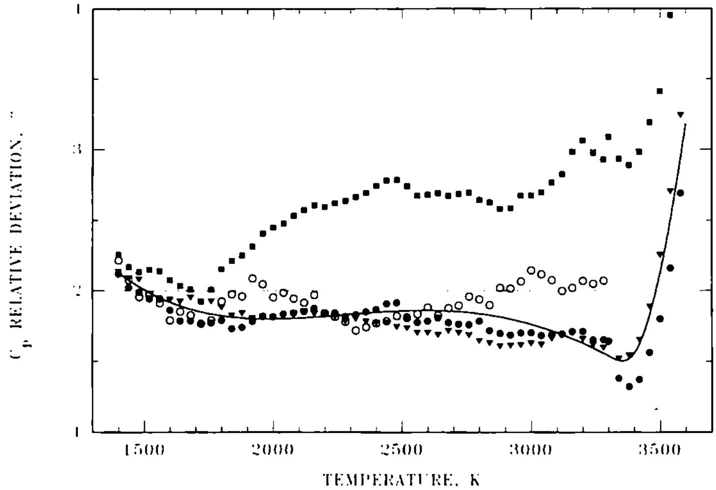

Fig. 2. Relative deviation of the experimental results on the heat capacity of thoriated tungsten from the IMGC results on the heat capacity of tungsten. (●) Specimen S-1, vacuum; ( ) S-1 argon; (▼) S-2; (■) S-3. The solid line represents the least-squares fitting in the temperature range 1400–3600 K with two splines. One datum of four is plotted.

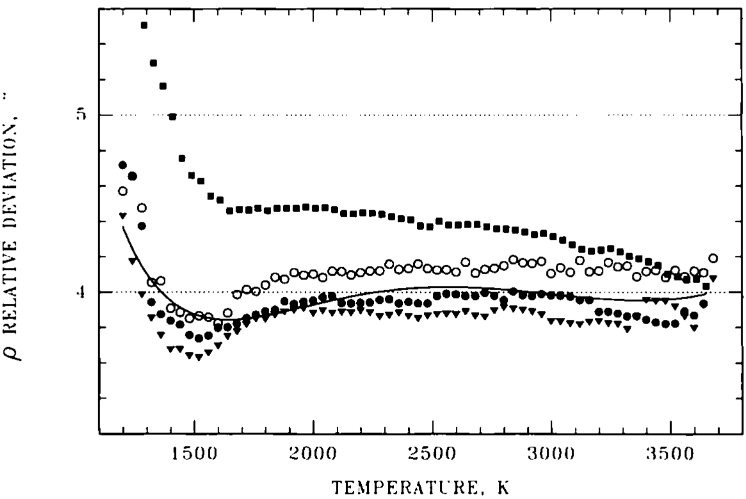

Fig. 3. Relative deviation of the experimental results on the electrical resistivity of thoriated tungsten from the IMGC results on the electrical resistivity of tungsten. (●) Specimen S-1, vacuum; ( ) S-1, argon; (▼) S-2; (■) S-3. The solid line represents the least-squares fitting in the temperature range 1200–3690 K. One datum of four is plotted.

3.2. Electrical Resistivity (ρ)

The experimental values of the electrical resistivity are based on geometric dimensions at room temperature (293 K): no thermal expansion correction was applied. The coefficients of the least-squares fitting polynomial are presented in Table III and the values obtained from the smooth function are in Table IV. The deviation of each set of experimental values from the electrical resistivity of tungsten is shown in Fig. 3, along with the smooth values from the fitting polynomial.

The smooth ρ function of Table III is valid in the entire temperature range 1200–3690 K. The noise in the data of Fig. 3 is typical of these experiments, because the electrical resistivity results are based on the

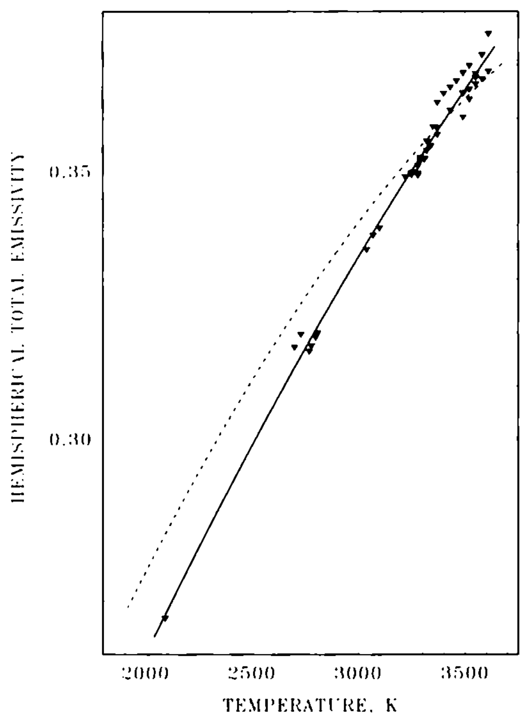

Fig. 4. Hemispherical total emissivity of thoriated tungsten. (▼) Specimen S-2. The solid and dashed lines are the least-squares fittings for thoriated tungsten and tungsten, respectively (vacuum experiments). One datum of three is plotted.

measured experimental quantities (temperature, current, voltage drop), with no smoothing except for the averaging of different experiments on the same specimen.

3.3. Hemispherical Total Emissivity (ε_{ht})

Hemispherical total emissivity depends on a combination of data obtained during heating and cooling. Measurements were performed on specimens S-2 and checks were performed on specimen S-1. The hemispherical total emissivity function is reported in Table III and computed in Table IV. The experimental results and the fitting polynomial are presented in Fig. 4, along with the data for tungsten [9].

4. DISCUSSION

The results of the uncertainty analysis expressed according to BIPM suggestions [14, 15] are presented in Table V. The second column takes into account only statistical considerations. The third column also includes estimated contributions of various sources: uncertainties after calibrations, combined effects of measured quantities on computed properties, uncertainties of literature data, etc. Details of the method used in preparing the uncertainty analysis are given in an earlier publication [2]; data were recomputed whenever the present experimental conditions differed from those of earlier experiments. The large uncertainties in temperature are due to the value of the melting point of tungsten, taken from the literature [11] with its corresponding uncertainty of 15 K, and to the assumed normal spectral emissivity-versus-temperature function (also taken from literature data).

Table V. Uncertainty of Measured and Computed Quantities

Quantity	Uncertainty[a]	
	Type A	Type B
Temperature		
At 1500 K	0.4 K	4 K
At 2500 K	0.2 K	8 K
At 3500 K	0.2 K	14 K
Heat capacity	0.14%	4%
Electrical resistivity	0.12%	1.5%
Hemispherical total emissivity	0.68%	7%

[a] See text for the designation of Type A and Type B uncertainties.

The literature contains extensive data on the electron emission properties of thoriated tungsten, but very few data on its thermal properties. A recent comprehensive book on tungsten and related materials [16] discusses in detail the production and usage of thoriated tungsten but reports no specific values on its thermal properties. An updated summary of recent reviews on the thermal properties of tungsten was reported in the earlier publications [9, 10].

The results of the present work on thoriated tungsten are compared (see Figs. 2 and 3) with the results obtained on pure tungsten [9, 10] at IMGC, using the same experimental technique. The heat capacity of thoriated tungsten (Fig. 2) is approximately 1.5–2% higher than that of tungsten up to 3300 K, then increases rapidly to 3% above the value of tungsten. The electrical resistivity of thoriated tungsten (Fig. 3) is approximately 4% higher than that of tungsten and exhibits the same temperature dependence up to the melting point.

From the experimental results reported in Figs. 2 and 3, it is evident that several hours of heat treatment at temperatures above 3000 K under the usual argon protecting environment do not change significantly the thermal properties of thoriated tungsten. The heat-treated specimen S-3 exhibits maximum differences in heat capacity and electrical resistivity of the order of 1% with respect to the as received specimens. Part of this difference might also be attributable to geometrical dimension measurements, which were difficult on S-3 on account of the bent shape assumed during the heat treatment.

5. CONCLUSIONS

This work presents experimental data on the thermophysical properties of thoriated tungsten up to very high temperatures. They were obtained by using simple rod specimens and by assuming both the melting point and the temperature dependence of the normal spectral emissivity from the literature data on tungsten. It was assumed that the melting point of thoriated tungsten is the same one as that of tungsten, and this melting point was used as a calibration point, by forcing the normal spectral emissivity function to reproduce the true melting temperature at the plateau. In the case of tungsten and thoriated tungsten, this simple technique has provided adequate experimental results, even if with a larger uncertainty than using tubular specimens with a blackbody hole.

ACKNOWLEDGMENTS

This work was performed in the framework of the CNR-CSIRO scientific cooperation agreement. The work was supported in part by the Progetto Finalizzato Materiali of CNR.

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