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Thermophysical Properties by a Pulse-Heating Reflectometric Technique: Niobium, 1100 to 2700 K[1]

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Pulse-heating experiments were performed on niobium strips, taking the specimens from room temperature to the melting point is less than one second. The normal spectral emissivity of the strips was measured by integrating sphere reflectometry, and, simultaneously, experimental data (radiance temperature, current, voltage drop) for thermophysical properties were collected with sub-millisecond time resolution. The normal spectral emissivity results were used to compute the true temperature of the niobium strips; the heat capacity, electrical resistivity, and hemispherical total emissivity were evaluated in the temperature range 1100 to 2700 K. The results are compared with literature data obtained in pulse-heating experiments. It is concluded that combined measurements of normal spectral emissivity and of thermophysical properties on strip specimens provide results of the same quality as obtained using tubular specimens with a blackbody. The thermophysical property results on niobium also validate the normal spectral emissivity measurements by integrating sphere reflectometry.

KEY WORDS: electrical resistivity; heat capacity; hemispherical total emissivity; high temperatures; niobium; pulse-heating; refractory metals.

1. INTRODUCTION

Pulse-heating techniques are among the most versatile and accurate methods for the simultaneous determination of several thermophysical properties at high temperatures. Practical applications often require measurements to be performed on simple specimens. This paper describes accurate experimental

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measurements of heat capacity, electrical resistivity, and hemispherical total emissivity of niobium strip specimens over a wide temperature range performed recently at the Istituto di Metrologia "G. Colonnetti" (IMGC, Italy). The radiance temperature measured on the surface of the strip was converted into true temperature using normal spectral emissivity values obtained in the same experiments using a fast integrating sphere reflectometry technique developed at the IMGC [1]. This paper is complementary to the preceding one [2], which describes in detail the normal spectral emissivity measurements and the related experimental technique.

2. MEASUREMENTS

The measurements were performed with the high-speed multiproperty apparatus of the IMGC. 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 [3, 4].

The radiance temperature was measured using a high-speed pyrometer operating at 900 nm with a bandwidth of 82 nm. The temperature range from approximately 1100 K to the melting point of niobium was covered in a single experiment using an autorange technique [5]. The normal spectral emissivity data measured simultaneously with these thermophysical property measurements were used to obtain the true temperature from the radiance temperature measured in each experiment [2]. The pyrometer had a target area 0.8 mm in diameter and was focused at the center of the strip. The pyrometer calibration was based on long-term primary calibrations of its effective wavelength and its linearity. The melting point of niobium [6] (2749 K on the ITS-90) was used as a reference temperature. Most of the experiments reached the melting plateau (Fig. 1) and were stopped before destruction of the specimen using a measurement technique developed earlier at the IMGC [7]. The data at the melting plateau were used to calibrate the pyrometer (including the transmission of the window of the environmental chamber) and the integrating sphere reflectometer (details in Ref. 2). Pictures of the specimen taken after many measurements at the melting point (typical example in Fig. 2 of Ref. 2) indicate that a large central portion of the strip reached the melting point, including all of

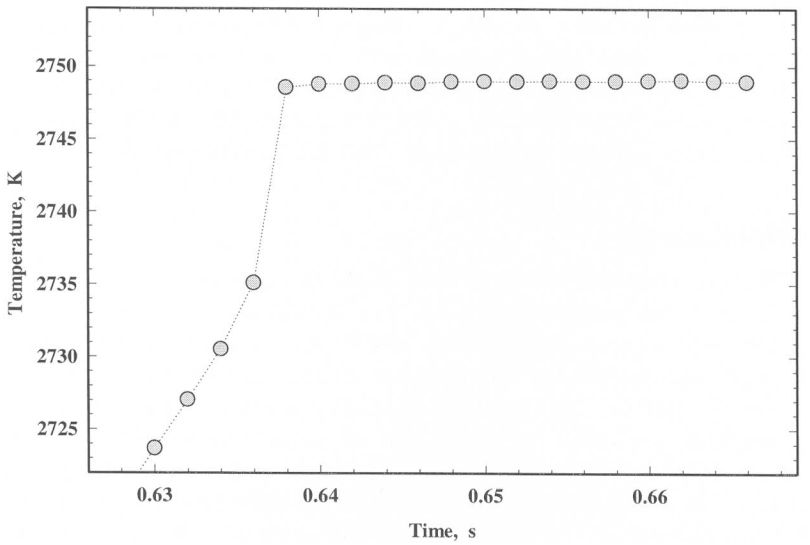

Fig. 1. Typical melting plateau of niobium used as temperature reference for the high-speed pyrometer.

the region between the voltage probes (spot-welded niobium wires of 0.05-mm diameter).

The strip specimens were machined at the IMGC from commercial niobium foil of nominal purity 99.9%. The typical analysis provided by the manufacturer indicated the following impurities (ppm): Ta, 500; Si and O, 100; W, <100; Fe, 30; C, 25; N, 20; Mo, 10; B, H, Ti, and Zr, <10; and Cu and Ni, <5. Measurements were performed on three strip specimens with the following nominal dimensions: total length, 83 mm; effective length (between the voltage probes), 24 mm; width, 10 mm; and thicknesses, 0.5 mm (specimens S1 and S2) and 1 mm (specimen S3). The specimens were preheated to their melting point before the final experiments to obtain a stable surface condition for normal spectral emissivity measurements [2].

The experimental data were obtained in fast experiments with typical current pulse durations of 0.5 to 0.7 s and heating rates in the range of 2200 to 5700 $K \cdot s^{-1}$. The experiments were conducted in high vacuum (better than 10^{-3} Pa). All the temperatures reported in this paper are based on the International Temperature Scale of 1990 (ITS-90) [8].

3. RESULTS

The thermophysical properties of niobium were determined from measurements taken on the central portion of the specimen between the

voltage probes. Several experiments were performed for each specimen, and the results are based on 9 experiments performed on specimen S1, 25 experiments performed on S2, and 10 experiments performed on S3. For computations a value of 8.572 $g \cdot cm^{-3}$ was assumed for the density of niobium, and a value of 92.91 $g \cdot mol^{-1}$ for its atomic mass [9].

3.1. Heat Capacity

The experimental results of heat capacity are adequately represented by a fourth-order polynomial (Table I). The function was computed with a specific procedure developed at the IMGC that requires only temperature differences and avoids the computation of temperature derivatives [10]. The deviation of the heat capacity of each specimen from the final fitting polynomial is presented in Fig. 2, and all values are within $\pm 1\%$. The radiative heat loss needed in the computation of heat capacity was obtained from the total hemispherical emissivity function reported in Table I. At temperatures below 2000 K, extrapolated values were used. At the highest temperatures the correction term for radiative heat losses was always less than 20%.

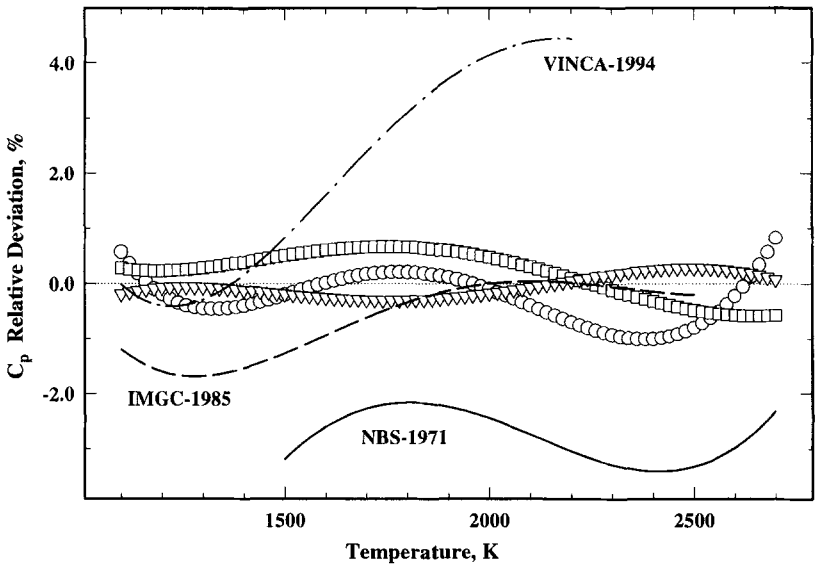

Fig. 2. Relative deviation of the experimental results on the heat capacity and of literature values from the value established by the least-squares fit as a function of temperature. (○) Specimen S1; (□) specimen S2; (▽) specimen S3. Literature curves are labeled as in the text.

Table I. Functional Representation of the Results for Niobium by Fitted Polynomials

	Heat capacity ($J \cdot mol^{-1} \cdot K^{-1}$)	Electrical resistivity ($n\Omega \cdot m$)	Total hemispherical emissivity
Polynomial	$a+bT+cT^2+dT^3+eT^4$	$a+bT+cT^2+dT^3$	$a+bT$
a	-0.633771	73.731	6.9345×10^{-2}
b	6.012135×10^{-2}	3.7793×10^{-1}	7.37435×10^{-5}
c	-4.549868×10^{-5}	-3.5349×10^{-5}	
d	1.530586×10^{-8}	2.6896×10^{-9}	
e	$-1.654222 \times 10^{-12}$		
Temp. range (K)	1100 to 2700	1100 to 2700	2000 to 2700
SD (%)[a]	0.67	0.34	1.5

[a] Relative standard deviation computed from the difference between the experimental values for each specimen and those from the smooth functions reported above.

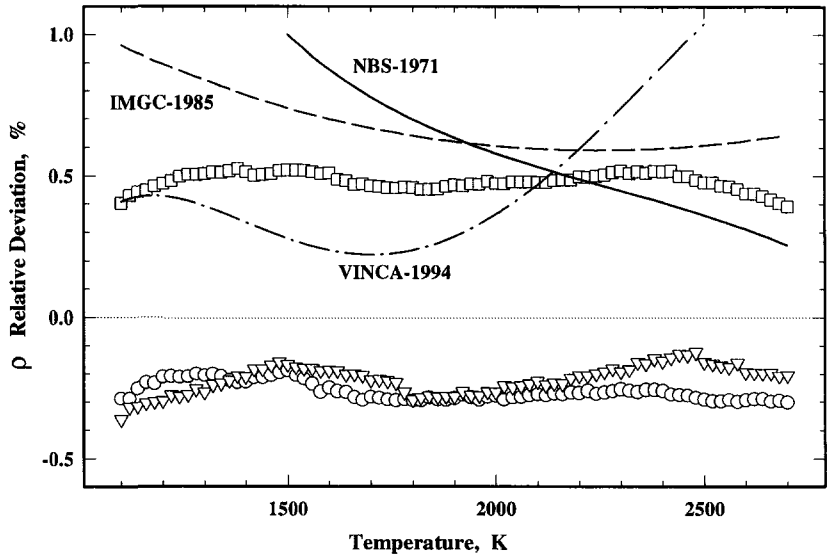

Fig. 3. Relative deviation of the experimental results on the electrical resistivity and of literature values from the value established by the least squares fit as a function of temperature. (○) Specimen S1; (□) specimen S2; (▽) specimen S3. Literature curves are labeled as in the text.

3.2. Electrical Resistivity

The experimental values of electrical resistivity are based on geometric dimensions at room temperature; no thermal expansion correction was applied. The coefficients of the least-squares fitting polynomial are presented in Table I, and the deviations of individual specimens from the fitting polynomial in Fig. 3. The results for individual specimens are average

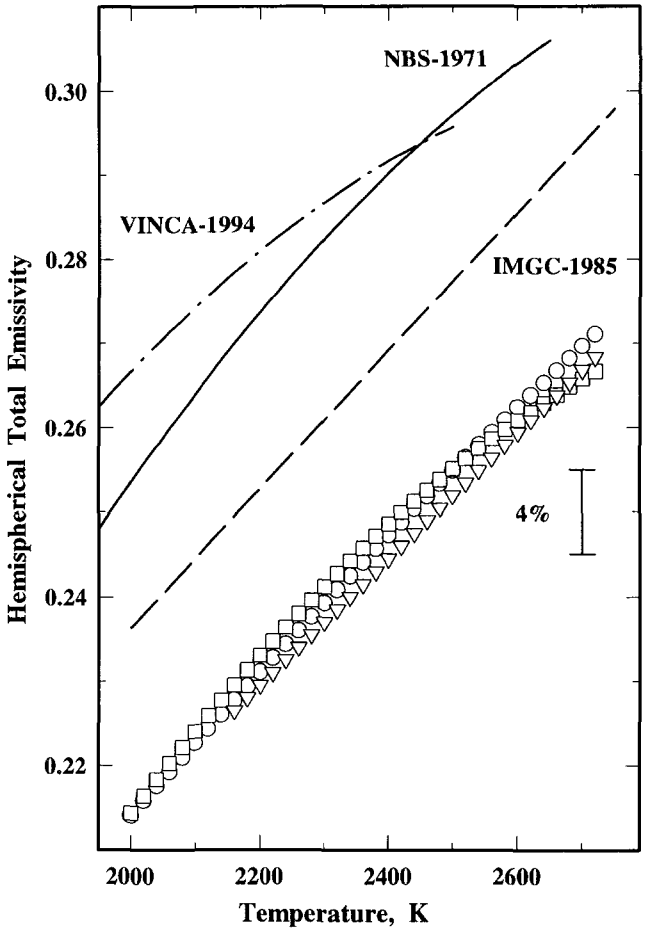

Fig. 4. Experimental results on the total hemispherical emissivity of niobium as a function of temperature. Results from the present work: (○) specimen S1; (□) specimen S2; (▽) specimen S3. Literature curves are labeled as in the text.

values computed at fixed temperatures by interpolation. It may be noted that the electrical resistivity of S2 is approximately 0.8% higher than the resistivity of the other specimens. Investigations of this discrepancy did not reveal any specific reason for the systematic difference, and, therefore, all specimens were retained with identical weight in the final computation.

3.3. Total Hemispherical Emissivity

The total hemispherical emissivity depends on a combination of experimental data obtained during heating and cooling. A classical evaluation procedure was followed, computing heating and cooling rates in appropriate temperature intervals near the maximum temperature reached in each experiment. The fitting polynomial is presented in Table I, and the results for individual specimens are shown in Fig. 4. The low total hemispherical emissivity values obtained in these experiments are a direct result of the measurement technique. The repeated cycling of the specimens through the melting point creates a shiny surface with very large grains (details in Fig. 2 of Ref. 2). Such a surface exhibits a lower emissivity with respect to specimens that did not reach the melting point.

Total hemispherical emissivity measurements using tubular specimens with a blackbody hole require much attention because, in short times after current interruption, the physical model fails. This happens because round-shaped temperature profiles (due to thermal conduction toward the clamps) cause errors in the cooling rate measured in the blackbody. The physical situation is more favorable in measurements on strips. The radiance temperature measured in the center of the strip is affected by thermal conduction processes only after some time, and consequently, using strips, it is possible to follow the cooling of the specimen for a temperature range of several hundred degrees.

4. DISCUSSION

The results of the uncertainty analysis (two-standard deviation level) expressed according to BIPM recommendations [11, 12] are presented in Table II. The second column (Type A uncertainty) takes into account only statistical considerations. The third column (Type B uncertainty) 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 [3]; data were recomputed whenever the present experimental conditions differed from those of earlier experiments. The somewhat large uncertainties

Table II. Uncertainty of Computed Thermophysical Properties

Quantity	Uncertainty (%)[a]	
	Type A	Type B
Heat capacity	±1.3	±4
Electrical resistivity	±0.7	±1.5
Hemispherical total emissivity	±3.0	±6

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

in the properties with respect to measurements on tubular specimens are due to the indirect method of temperature measurement (via radiance temperatures and normal spectral emissivities).

Recent papers on thermophysical properties of niobium at high temperatures [13, 14] have presented literature surveys of available data. In this paper literature comparisons are limited to measurements performed on the same material using pulse-heating techniques, to investigate possible differences of results of strip specimens (after the determination of the normal spectral emissivity) with respect to the use of other types of specimens. The following literature data were taken into consideration:

- measurements performed at the National Bureau of Standards, USA (NBS; presently NIST), in 1971 using tubular specimens with a blackbody hole [15] (referred to as NBS-1971 in the text and figures);
- measurements performed by our research group at the IMGC in 1985 using tubular specimens with a blackbody hole [13] (referred to as IMGC-1985); and
- measurements performed at the Institute of Nuclear Sciences VINCA (Yugoslavia) in 1994 using cylindrical specimens and measuring temperature by a combination of spot-welded thermocouples and pyrometry [14] (referred to as VINCA-1994).

If necessary, the data on literature references were adjusted to the ITS-90, so that comparisons were made using the same temperature scale.

Literature comparisons for heat capacity are presented in Fig. 2. The data from the present work are in good agreement with the IMGC-1985 results over the entire temperature range, with a maximum deviation of less than 2%. There is good agreement with the VINCA-1994 data up to 1500 K; then the results indicate a different temperature trend. There is a systematic difference of 2 to 3% from the NBS-1971 data, even if the two

sets indicate the same temperature trend. This difference had already been found with the IMGC-1985 measurements, and it is confirmed. It should be noted that in all cases the differences are well within the combined uncertainties indicated by the laboratories, and consequently, several sets of consistent heat capacity data from different laboratories are now available.

Literature comparisons of electrical resistivity are presented in Fig. 3. The data in this case seem to indicate slightly different temperature trends, but the four data sets (NBS-1971, IMGC-1985, VINCA-1994, and the results of the present work) are in agreement within 1%. Also, in this case, the differences are all within the combined uncertainties estimated by each laboratory.

Differences in total hemispherical emissivity are presented in Fig. 4. Absolute values of total hemispherical emissivity are strongly dependent on surface conditions, and, therefore, large differences are possible and expected. The reason for the low values found in the present work from repeated cycles through the melting point has already been explained. It is noteworthy that the temperature trend (not dependent on surface conditions) is highly similar in all the data sets.

It may be concluded that the thermophysical property measurements performed on strip specimens with simultaneous determination of the normal spectral emissivity provide accurate data of the same quality as measurements performed on tubular specimens with a blackbody hole. The results of the present work also validate the normal spectral emissivity data obtained by integrating sphere reflectometry in the same experiments.

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