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Thermal Expansion of Molybdenum in the Range 1500–2800 K by a Transient Interferometric Technique[1]

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The linear thermal expansion of molybdenum has been measured in the temperature range 1500–2800 K by means of a transient (subsecond) interferometric technique. The molybdenum selected for these measurements was the Standard Reference Material SRM 781 (a high-temperature enthalpy and heat capacity standard). The results are expressed by the relation

$$(l-l_0)/l_0 = 6.0373\times10^{-3} - 1.1175\times10^{-5}T + 1.3557\times10^{-8}T^2$$
$$-4.6688\times10^{-12}T^3 + 6.9032\times10^{-16}T^4$$

where T is in K and l_0 is the specimen length at 20°C. The maximum error in the reported values of thermal expansion is estimated to be about 1% at 2000 K and not more than 2% at 2800 K.

KEY WORDS: high temperature; interferometry; molybdenum; pulse heating; thermal expansion.

1. INTRODUCTION

Measurements of thermal expansion at high temperatures ($T > 1100$ K) have generally relied on conventional steady-state or quasi-steady-state techniques such as push-rod dilatometry, X-ray diffractometry, and optical comparator methods. In these techniques, the long heating periods (minutes to hours) create problems associated with the increased heat transfer, loss of mechanical strength, specimen contamination and evaporation, etc., which become particularly severe at temperatures above 2000 K. To minimize such difficulties, we recently developed a high-speed

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interferometric technique [1] in which the entire experiment is performed in less than 1 s. This involved adapting a Michelson-type interferometer to the existing pulse-heating system [2, 3] at the National Bureau of Standards (NBS).

The method is based on rapid resistive self-heating of the specimen from room temperature to the maximum temperature of interest by the passage of a subsecond-duration electrical current pulse through it and on simultaneously measuring the specimen temperature by means of a high-speed photoelectric pyrometer [4] and the shift in the fringe pattern produced by the interferometer. The polarized beam from a He–Ne laser in the interferometer is split into two component beams, one which undergoes successive reflections from highly polished flats on opposite sides of the specimen and one which serves as the reference beam. The linear thermal expansion of the specimen is then determined from the cumulative fringe shift corresponding to a given specimen temperature. Details regarding the construction and operation of the measurement system and other pertinent information, such as formulation of relations to determine thermal expansion from the fringe shift, error analyses, etc., are given in the earlier publication [1].

In the present study, the technique was used to determine the linear thermal expansion of molybdenum at temperatures in the range 1500 to 2800 K. The molybdenum selected for the present work was the Standard Reference Material SRM 781, which has been certified by the Office of Standard Reference Materials (OSRM) at the NBS as a standard for enthalpy and heat capacity measurements at temperatures up to 2800 K [5].

2. MEASUREMENTS

The SRM 781 molybdenum used in the present work was supplied in the form of an annealed, polycrystalline rod by the OSRM. An earlier mass-spectrometric analysis [6] of a representative sample of this material yielded the following impurities (ppm by mass): W, 56; Fe and Si, 40 each; Mg, O, and Sn, <30 each; Ta, <25; K, 20; Ar and Ni, <20 each; Cu, 15; Cr, 12; Au and N, <10 each; Ca, 9; Cd, <8; Al, 7; Co, <7; Os and Pd, <6 each; Pt, <4; C, Ir, and Nb, <3 each; As and Mn, 2 each; and Ag, 1. The total amount of all other detected elements was less than 10 ppm, each element being below the 1-ppm detection limit.

The molybdenum rod was fabricated into three precision-machined tubes by an electroerosion technique. The nominal dimensions of each specimen tube were as follows; length, 76 mm: outside diameter, 6.35 mm; and inside diameter, 5.1 mm. A small rectangular sighting hole

(0.5 × 1 mm) was fabricated through the wall at the middle of each tube, thereby approximating a blackbody cavity for the pyrometric temperature measurements. The sighting hole was positioned 0.8 mm off center from the tube axis to improve the blackbody quality. In order to compensate for the cross-sectional nonuniformity created by the hole, a portion of the specimen was removed by grinding a flat along the length of the tube, excluding the 1-mm length of the hole. For the interferometric measurements, highly polished parallel flats were fabricated on opposite sides along the length of each tubular specimen. The distance between the optical flats (i.e., the specimen "length," l_0) was measured at 20°C by the Automated Production Technology Division at the NBS using a comparative gauge block technique. The results are given in Table I, along with estimates of the axial nonparallelism and departure from flatness of the two reflecting surfaces.

Each specimen tube, in turn, was mounted vertically between two water-cooled electrodes inside the test chamber which was then evacuated to a pressure of about 1 mPa ($\sim 10^{-5}$ Torr). The upper electrode is stationary, whereas the lower electrode is attached (through a linear guide) to a flexible connection which enables the specimen to expand downward along its length during pulse heating.

Prior to each pulse experiment, adjustments were made to the voltage from a battery bank and to a resistance in series with the specimen in order to achieve the desired heating rate. The specimen was then rapidly heated

Table I. Results of Measurements Performed on the Specimen Optical Flats by a Comparative Gauge Block Technique

	Specimen No.[a]		
	1	2	3
Distance between flats (mm)	5.8308	5.8237 (5.8237)[b]	5.9159 (5.9154)
Axial parallelism (min. of arc)	0.7	0.7 (1.1)	0.7 (1.4)
Flatness[c] (waves per 25 mm)	1	1 (2)	1 (2)

[a] Also, the chronological order in which the specimens, in turn, were pulse heated through the temperature range between 1500 and 2800 K.

[b] Results of measurements performed upon completion of the pulse-heating experiments are given in parentheses.

[c] Given in terms of the wavelength 632.8 nm.

from "room" temperature ($\sim$17°C) to the desired temperature by passing an electrical current pulse through it; the duration of the current pulse varied from about 650 to 760 ms. The temperature interval of the measurements (1500–2800 K) was divided into five overlapping temperature ranges in order to optimize the signal resolution of the high-speed pyrometer. For a given specimen, single pulse experiments were performed successively through each temperature range, beginning with the lowest range. Heating rates varied typically from about 3300 $K \cdot s^{-1}$ in the lowest temperature range to about 4500 $K \cdot s^{-1}$ in the highest range.

During each pulse experiment, the specimen temperature and the fringe shift were measured at rates of 1200 and 5000 times/s, respectively. These data were recorded by means of two digital storage oscilloscopes which have a full-scale signal resolution of about 1 part in 4000. After the experiment, the recorded data were transferred to a minicomputer for subsequent analyses.

Upon completion of the experiments, we calibrated the high-speed pyrometer using a tungsten-filament standard lamp which, in turn, had been calibrated against the NBS photoelectric pyrometer by the Radiometric Physics Division at the NBS. In addition, the reference dimension at 20°C, l_0, for two of the specimens (Nos. 2 and 3) was remeasured, and the results along with estimates of the nonparallelism and flatness of the reflecting surfaces after pulse heating are given (in parentheses) in Table I. All temperatures reported in this work, unless explicitly stated otherwise, are based on the International Practical Temperature Scale of 1968 [7].

Table II. Results of Fitting, by Means of the Least-Squares Method, the Expansion/Temperature Data Pairs for the Individual Specimens and the Combined Data for All Three Specimens by Polynomial Functions in Temperature (in K) of the Form $(l-l_0)/l_0 = a_0 + a_1 T + a_2 T^2 + a_3 T^3 + a_4 T^4$

Specimen No.	Number of data pairs[a]	SD (%)[b]	Polynomial coefficient[c] $a_0 \times 10^3$	$a_1 \times 10^6$	$a_2 \times 10^8$	$a_3 \times 10^{12}$	$a_4 \times 10^{16}$
1	81	0.06	4.9086	−8.2853	1.0963	−3.6883	5.5637
2	84	0.06	5.1218	−9.4133	1.2292	−4.2690	6.4418
3	83	0.09	3.5259	−6.6471	1.0561	−3.8028	5.9766
All specimens	248	0.15	6.0373	−11.175	1.3557	−4.6688	6.9032

[a] The number of data pairs fitted by the polynomial function represents only one of every six expansion/temperature data points recorded in the pulse experiments.

[b] Standard deviation of an individual value of $(l-l_0)/l_0$ from the smooth function.

[c] Based on the specimen reference length (l_0) at 20°C.

3. RESULTS

The linear thermal expansion of the specimen was determined at each recorded temperature from the cumulative fringe shift Δn by means of the relation

$$(l-l_0)/l_0=(\lambda/2l_0)\,\Delta n \qquad (1)$$

where λ is equal to 632.8 nm and l_0 is the specimen "length" at 20°C. In order to account for any difference between the initial temperature of the specimen and 20°C, a "zero" correction was applied to Δn on the basis of

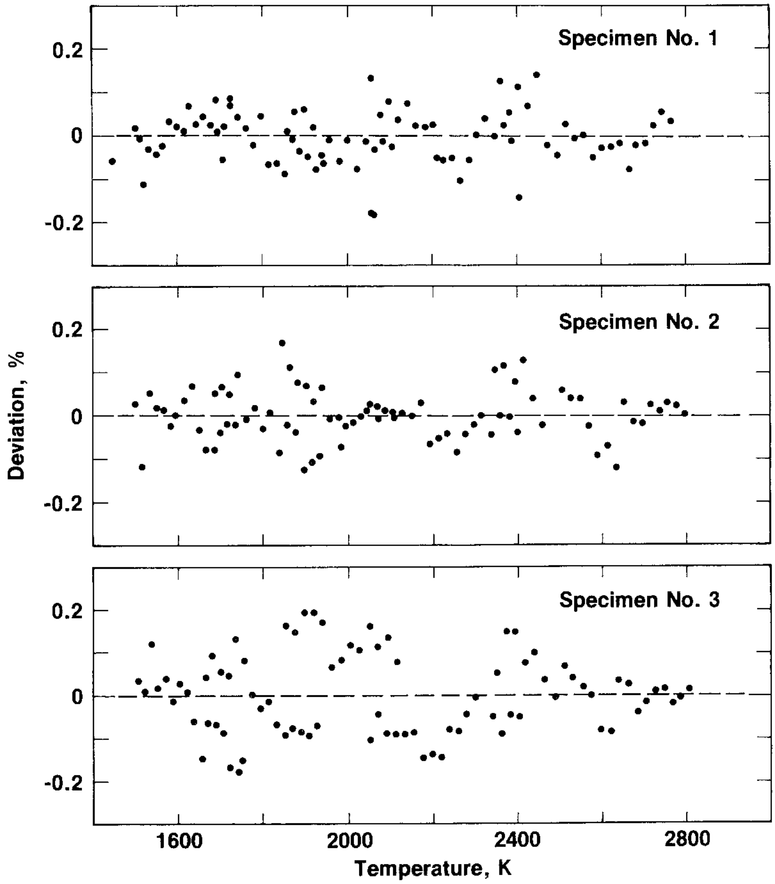

Fig. 1. Deviation of expansion/temperature data pairs from the smooth functions (see Table II) representing the least-squares fits to data for the individual specimens.

expansion data near room temperature reported in the literature [8]. The expansion/temperature data pairs for each specimen were then fitted by a quartic polynomial function of temperature by the least-squares method. The polynomial functions representing the results for individual specimens are given in Table II. The deviation of individual data points from the smooth function for each specimen is illustrated in Fig. 1. The random fluctuation among data points within a given temperature range is due primarily to the uncertainty in determining the fringe count from a limited number of data points per fringe (about 12 at 1500 K, decreasing to about 5 at 2800 K). In certain cases, differences between data from overlapping temperature ranges also contribute to the scatter of data points.

The thermal expansion of specimen 1 was also measured in the next higher temperature range, up to its melting point (~2895 K). These results (open circles) are compared with those for the next lower temperature range (filled circles) in Fig. 2 by plotting the percentage deviation from the smooth polynomial function based on data for the spcimen in the five lowest ranges. The good agreement suggests that the polynomial functions for individual specimens may be extrapolated beyond 2800 K to yield thermal expansion values at temperatures within a few degrees of melting without serious error.

The final results were obtained by combining the expansion/temperature data pairs for the three specimens and then fitting them by a quartic polynomial function of temperature. The function that represents

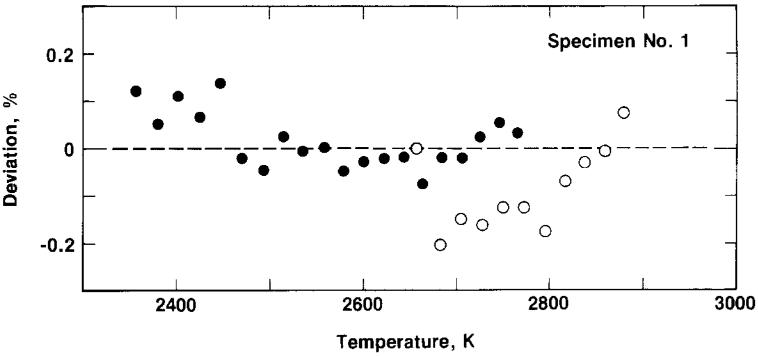

Fig. 2. Deviation of expansion/temperature data pairs for specimen 1 from the smooth polynomial function (see Table II) based on data in the five lowest temperature ranges. The filled circles represent data from the highest of these ranges and the open circles represent data from an additional higher range which includes the melting point (~2895 K).

Table III. Smoothed Results[a] for the Linear Thermal Expansion of Molybdenum

Temperature (K)	$10^2 \times (l-l_0)/l_0$ (%)	Temperature (K)	$10^2 \times (l-l_0)/l_0$ (%)
1500	0.752	2200	1.353
1600	0.826	2300	1.456
1700	0.905	2400	1.567
1800	0.987	2500	1.685
1900	1.072	2600	1.811
2000	1.161	2700	1.949
2100	1.254	2800	2.098

[a] Based on the specimen reference length (l_0) at 20°C.

(SD = 0.15%) the results for linear thermal expansion of molybdenum (SRM 781) in the temperature range 1500–2800 K is

$$(l-l_0)/l_0 = 6.0373 \times 10^{-3} - 1.1175 \times 10^{-5}T + 1.3557 \times 10^{-8}T^2 - 4.6688 \times 10^{-12}T^3 + 6.9032 \times 10^{-16}T^4 \quad (2)$$

where T is in K. The smoothed results, as defined by Eq. (2), are illustrated in Fig. 3 and are given at intervals of 100 K in Table III.

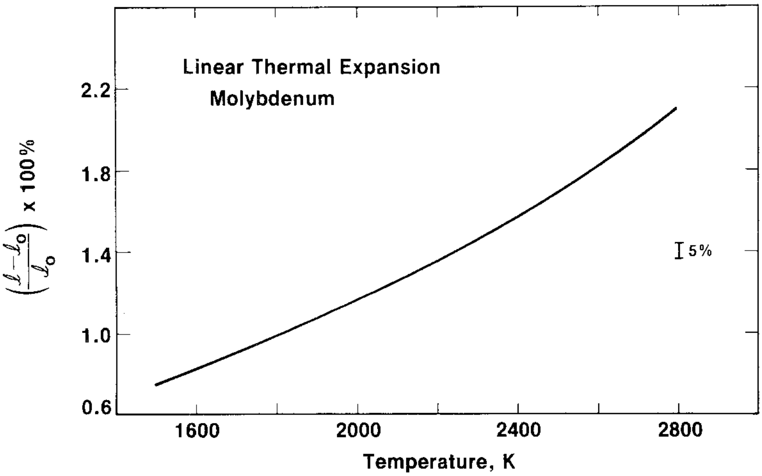

Fig. 3. Smoothed results for the thermal expansion of molybdenum as expressed by Eq. (2).

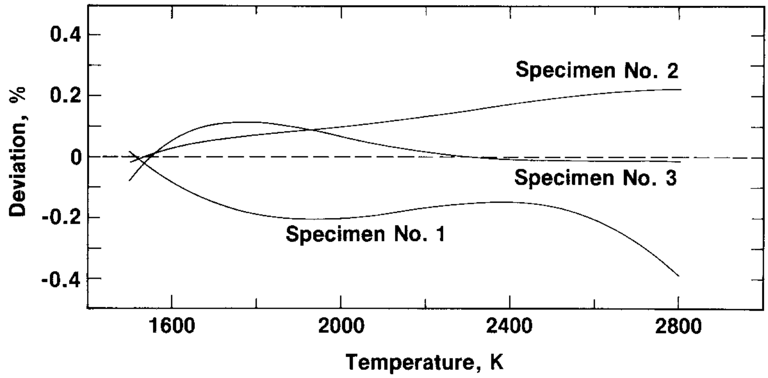

Fig. 4. Deviation of the smoothed thermal expansion results for the individual specimens from Eq. (2), which represents the least-squares fit to the combined data for three specimens.

The deviation of the smoothed results for individual specimens from those of Eq. (2) is given in Fig. 4. In all cases, the maximum deviation of these results from the overall least-squares fit is about 0.2% at temperatures below 2400 K and less than 0.4% at higher temperatures.

4. ESTIMATE OF ERRORS

A detailed analysis of errors in such experimental quantities as temperature, fringe count, and specimen "length" at 20°C was given in an earlier publication [1]. Specific items in the error analysis were recomputed whenever the present conditions differed from those in the earlier publication. The resultant estimated maximum error in the reported values of thermal expansion is about 1% at 2000 K and not more than 2% at 2800 K.

5. DISCUSSION

A comparison of expansion data for molybdenum reported in the literature with the present results is presented in Fig. 5. The curves, which are numbered chronologically, give the percentage deviation of literature data obtained by push-rod dilatometry, X-ray diffractometry, and optical comparator methods from the present results as expressed by Eq. (2). It can be seen that the literature data do not show any significant bias toward a given steady-state measurement technique. A "zero" correction was applied to the data whenever the reported reference temperature differed from 20°C. No attempt was made to convert the reported temperatures to

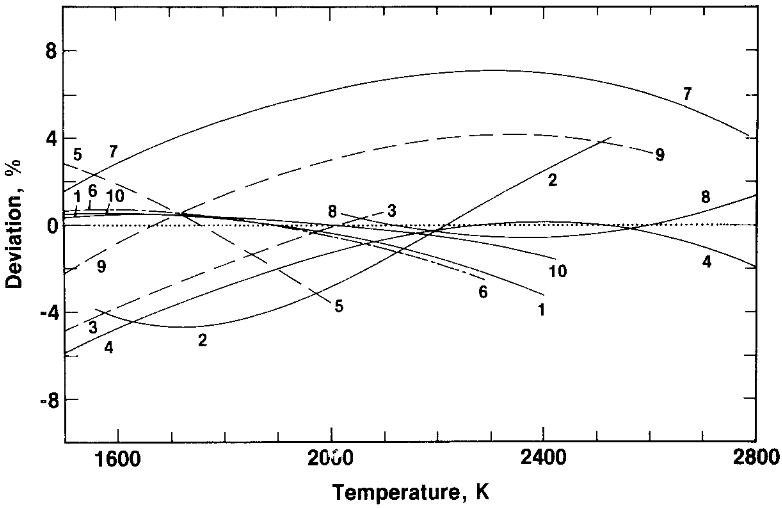

Fig. 5. Deviation of thermal expansion data for molybdenum reported in the literature from the present results as expressed by Eq. (2) (the dotted baseline). Measurement techniques: push-rod dilatometry (— · —); X-ray diffractometry (——); optical comparator methods (——). Curve numbers refer to (1) Worthing [9], (2) Demarquay [10], (3) Edwards et al. [11], (4) Rasor and McClelland [12], (5) Ross and Hume-Rothery [13], (6) Amonenko [14], (7) Conway and Losekamp [15], (8) Petukhov and Chekhovskoi [16], (9) Waseda et al. [17], and (10) Petukhov et al. [18].

IPTS-68 since the particular temperature scale used in the different studies could not be determined unambiguously in all cases and since such "corrections" would be relatively small (less than 0.5%).

The molybdenum expansion data reported by Worthing (curve 1 [9]), Amonenko et al. (curve 6 [14]), Petukhov and Chekhovskoi (curve 8 [16]), and Petukhov et al. (curve 10 [18]) are in rather good agreement with the present results, differing by less than 1% at temperatures up to 2000 K and by about 2% or less at temperatures above 2200 K. The other reported values of thermal expansion exhibit different trends with changing temperatures and deviate from the present results by as much as 7%. A common trend may be seen in the values obtained by Edwards et al. (curve 3 [11]), Rasor and McClelland (curve 4 [12]), Conway and Losekamp (curve 7 [15]), and Waseda et al. (curve 9 [17]); however, the disagreement among these data is as large as 8%.

As mentioned earlier, molybdenum SRM 781 is certified as a standard for enthalpy and heat capacity measurements at temperatures up to 2800 K. As yet, existing SRMs for thermal expansion cover the temperature range only up to 2000 K [5]. The present study suggests that SRM 781 would be a suitable candidate as a standard material for thermal expansion measurements up to 2800 K.

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