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Measurements of linear thermal expansion coefficients of copper SRM 736 and some commercially available coppers in the temperature range 20–300 K by means of an absolute interferometric dilatometer*

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The thermal expansivities of several grades of copper were measured in the temperature range 20–300 K by means of an absolute laser interferometric dilatometer. The present results for copper standard reference material (NIST; SRM 736) and three commercially available kinds of copper, such as a high-purity copper (99.999% purity), an oxygen-free high-conductivity copper and a tough-pitch copper, show good agreement with the NIST certified values and the CODATA recommended values within 0.09 × 10^{-6} K^{-1} over the whole temperature range. © 1997 Elsevier Science Ltd.

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Copper is one of the most important materials in cryogenic engineering. It has many advantages as a structural material for cryogenic instruments, because of its very high thermal conductivity, high electric conductivity, machinability, weldability, etc. However, the linear thermal expansion coefficient (LTEC) is also a very important thermophysical property for cryogenic materials, as it is needed to estimate the thermal stress among materials with different thermal expansion coefficients. NIST supplies a high-purity copper code-named SRM 736 as a reference material for thermal expansion with certified data[1,2]. However, reliable expansivity data have been unavailable for commercially available industrial grade coppers. Therefore a method is required to measure and compare their LTECs with a kind of dilatometric instrument.

This paper presents measured results of LTECs between 20 and 300 K of a copper standard reference material (SRM 736, supplied by US NIST) and commercially available coppers, such as a high-purity copper (99.999% purity), an oxygen-free high-conductivity copper and a tough-pitch copper. One purpose of this work was to accumulate reliable data for standard materials such as copper, and to compare the measured results with NIST certified values and CODATA recommended values. Another was to obtain the LTECs of commercially available copper for common use, and to compare them with those of SRM.

Absolute expansivity measurements were carried out using an optical heterodyne laser interferometric dilatometer developed by Okaji *et al.*[3,4]. The measured uncertainties were about 1 nm in length measurement, about 10 mK in temperature measurement, and a few units × 10^{-8} K^{-1} in LTEC measurement.

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Interferometric dilatometer

An absolute interferometric dilatometer was used for the present measurements. This instrument is described in detail in our previous paper[3]. *Figure 1* shows its mechanical arrangement (partly revised from the original figure in reference 3). An improved interferometer, which enables more accurate length measurement than the previous one[3], was adopted for the present system. Its details are described elsewhere[4]. A nanometer accuracy length measurement is provided by the interferometer with optical heterodyne interferometry.

Details of the dilatometer's specimen cell are shown in *Figure 2*. A parallel spring was newly adopted as a specimen holding mechanism (*Figure 3a,b*). It was made from a beryllium–copper block by a wire-cutting machine. It was used to achieve parallelism between the two reflecting mirrors, which comprise part of the interferometer, so fine pol-

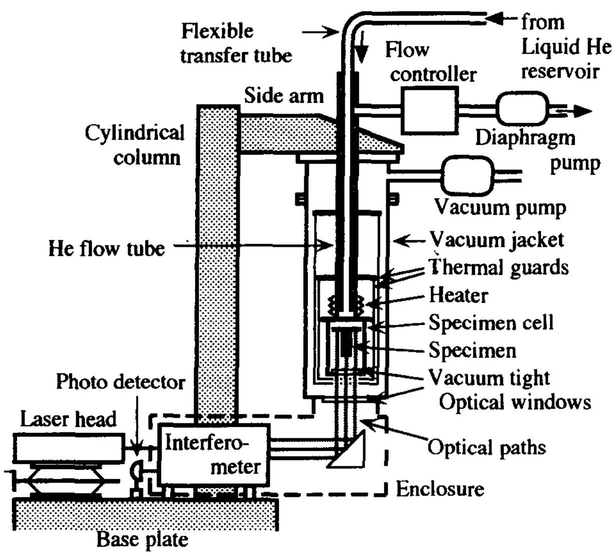

Figure 1 Mechanical arrangement of the dilatometer

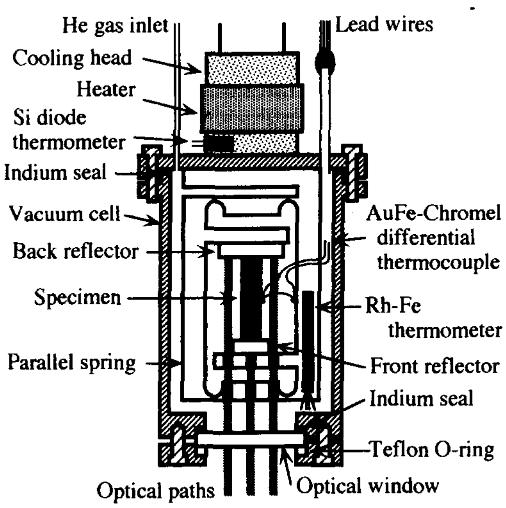

Figure 2 Details of the specimen cell

ishing of the measured specimen was unnecessary. The thickness of its hinges was adjusted properly so that only slight compressive force, about 1.5×10^3 Pa, was applied to the specimen. An Rh–Fe resistance thermometer (Rh–Fe RT), which was used to determine the absolute temperature of the specimen, was calibrated with an accuracy of better than 5 mK between 5 and 300 K. An AuFe–chromel differential thermocouple was added to measure possible temperature differences between the Rh–Fe RT and the specimen under a constant heating rate measurement condition, as mentioned later.

Measured specimens

Copper standard reference material (SRM 736) supplied by NIST, and three kinds of commercially available copper: high-purity (HP) copper (99.999% purity), oxygen-free high-conductivity (OFHC) copper and tough-pitch (TP) copper, were prepared as listed in *Table 1*. The purity of the SRM was mentioned as 99.99% in the NBS certificate, which was estimated from the residual resistivity ratio $R_{273\,K}/R_{4.2\,K}$[1]. OFHC copper is generally used under high vacuum circumstances with its very small out-gas characteristics, and TP copper is for electrical wires and parts with its high electric conductivity.

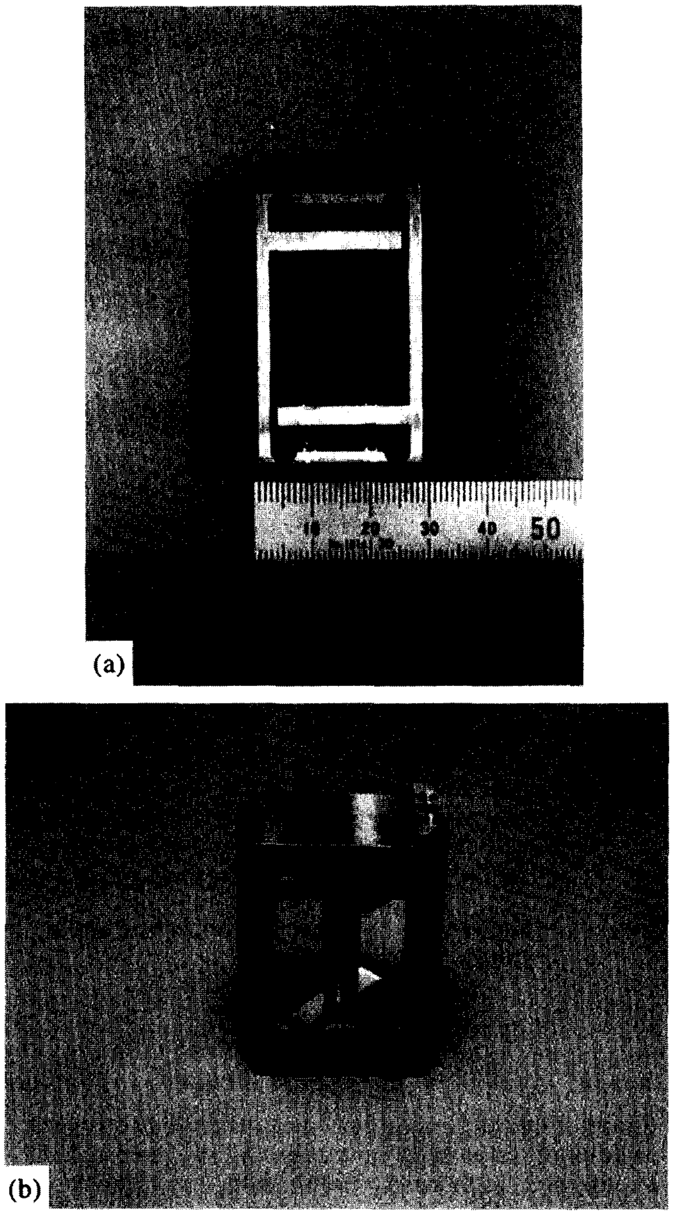

Figure 3 Photographs of the parallel spring used as a specimen holding mechanism: (a) the spring itself; (b) the spring with a specimen and two reflectors

The SRM material (originally 6.4 mm in diameter and 151 mm long) was cut into a 20 mm long specimen, both ends of which were polished flat (flatness within $\lambda/10$) and mutually parallel (parallelism within 5″). It was annealed at about 500°C for several hours in vacuum before the measurements. The other specimens were machined to cylindrical shape from bulk blocks. They were measured as machined without any heat treatment, because their characteristics may be similar to actual materials used at the cryogenic region.

Results and discussion

In this work, all measurements were carried out at a constant heating rate which was fixed at 12 K h^{-1} for the whole temperature range. A data set of length and temperature recorded every 10 s (360 sets h^{-1}) and a LTEC was calcu-

Table 1 Measured specimens (unit mm)

Material	Shape	Purity	Heat treatment
NIST SRM 736 copper	6.4 $\phi \times$ 20.002 L	99.99%	Yes (annealed at 500°C)
HP copper	6.0 $\phi \times$ 20.001 L	99.999%	No (as-machined)
OFHC copper	6.0 $\phi \times$ 20.005 L	>99.97%[a]	No (as-machined)
TP copper	6.0 $\phi \times$ 19.992 L	>99.95%[a]	No (as-machined)

[a] Nominal purity

lated from the 150 continuous data sets of length and temperature, which correspond to the average LTEC over a 5 K temperature interval. Here, LTEC ($\bar{\alpha}$) was defined by

$$\bar{\alpha} = \Delta L/(L_0 \Delta T) \quad (1)$$

where ΔL and ΔT are the average length and temperature changes of a specimen, and L_0 is a specimen length at 293 K. The LTEC with the finite temperature interval measurement generally differs from the instantaneous LTEC, defined by $\alpha = dL/(L_0 dT)$. However, the difference between them is estimated to be only 6×10^{-9} K^{-1} at the maximum[5], and such a value is much smaller than the scatter of the data. Consequently such a difference was negligibly small in this case.

The thermal expansivity data of SRM 736 copper, obtained by the process described above, are plotted as filled points in *Figure 4*. The NIST certified data and the CODATA recommended data are shown as solid triangles and open squares, respectively. Only 1% of the total recorded data are plotted in the figure, because the total is over 8000.

These values were fitted to the following function:

$$\alpha\,(10^{-6}\,\mathrm{K}^{-1}) = a + bT + cT^2 + dT^3 + eT^4 + fT^6 + gT^6 + hT^7 \quad (2)$$

where $a = 1.986 \pm 0.003$, $b = -(2.5179 \pm 0.0001) \times 10^{-1}$, $c = (1.062553 \pm 0.000003) \times 10^{-2}$, $d = (-1.326651 \pm 0.000001) \times 10^{-4}$, $e = (8.523575 \times 0.000003) \times 10^{-7}$, $f = (-3.045470 \pm 0.000001) \times 10^{-9}$, $g = (5.748850 \pm 0.000003) \times 10^{-12}$, $h = (-4.47461 \pm 0.0003) \times 10^{-15}$. These parameters were determined using a least-squares method. The deviation of each data point from the fitting function (Equation (2)) is shown in *Figure 5*. The standard deviation of the data from the fit was calculated to be 0.028×10^{-6} K^{-1}. The present results calculated from Equation (2) are listed in *Table 2*.

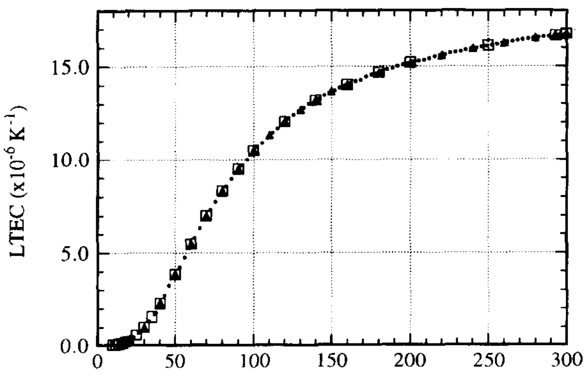

Figure 4 Measured data of SRM 736 copper: ●, present data; ▲, NIST certified data; □, CODATA recommended data

Figure 6 shows the difference between the present measured expansivity of the SRM specimen and the NIST-certified data and the CODATA-recommended data[6], which are within 0.03×10^{-6} K^{-1} of one another. The data scatter of the present result was calculated to be 0.028×10^{-6} K^{-1}. On the other hand, the scatter level of the data is 0.06×10^{-6} K^{-1} according to NIST calibration, and the maximum probable error of the CODATA recommended data is 0.02×10^{-6} K^{-1}. Consequently, these data show no significant statistical differences.

Figure 7 shows the expansivity deviations for the coppers (HP, OFHC and TP) from the CODATA recommended data. These deviations from the CODATA data are within 0.09×10^{-6} K^{-1}. Data scatters for these specimens are of the same order as that of the SRM. Hence, it is confirmed that the LTEC of all specimens agree very well.

Consequently, certified expansivity data by NIST and CODATA or the data presented here are applicable generally to normally used coppers when cryogenic instruments are designed and manufactured. Also, commercially available coppers (HP, OFHC and TP) can be used as reference materials for differential dilatometers such as push-rod dilatometers, thermo-mechanical analyzers and capacitance dilatometers.

Conclusions

The conclusions obtained in the present work are as follows.

1. Thermal expansivity measurements of four kinds of copper were measured by means of a laser interferometric absolute dilatometer in the temperature range 20–300 K.
2. Measured specimens were a copper standard reference

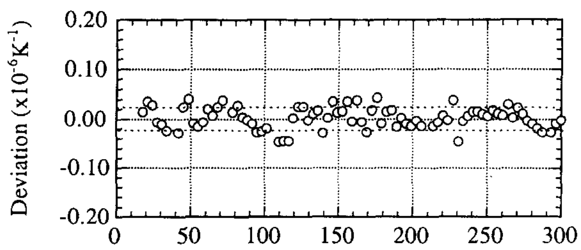

Figure 5 Deviation of each data point from fitting function (Equation (2)) (dotted line represents the standard deviation of each data point)

Table 2 Generated smooth values of LTEC from the fit (Equation (2)) (unit × 10^{-6} K^{-1})

T (K)	LTEC	*T* (K)	LTEC	*T* (K)	LTEC
		110	11.34	210	15.42
20	0.27	120	12.06	220	15.62
30	1.03	130	12.67	230	15.79
40	2.32	140	13.18	240	15.94
50	3.84	150	13.63	250	16.09
60	5.41	160	14.02	260	16.23
70	6.91	170	14.36	270	16.38
80	8.27	180	14.67	280	16.52
90	9.46	190	14.95	290	16.64
100	10.48	200	15.20	300	16.70

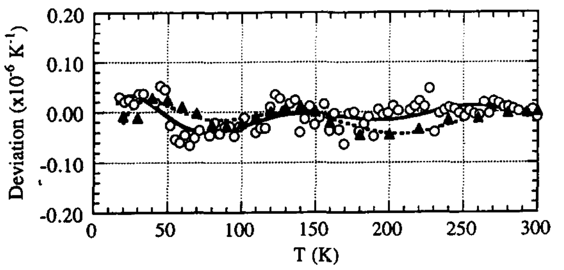

Figure 6 Expansivity deviations for the present measured expansivity of SRM specimen and the NIST certified data from the CODATA recommended data: – o –, present data; · ·▲· ·, NIST certified data

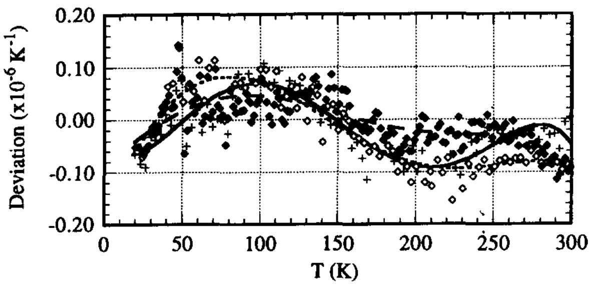

Figure 7 Expansivity deviations for HP, OFHC and TP coppers from the CODATA recommended data: – + –, HP copper; · ·◇· ·, OFHC copper; · ·◆· ·, TP copper

material (SRM 736, supplied by NIST) and commercially available coppers, such as a high-purity (HP) copper (99.999% purity), an oxygen-free high-conductivity (OFHC) copper and a tough-pitch (TP) copper.

3. The present results of the copper SRM 736 agree very well with the NIST certified value and the CODATA recommended value within 0.04 × 10^{-6} K^{-1} over the whole temperature range.
4. The LTEC of the three commercially available coppers agree well with each other and with the NIST certified data and the CODATA recommended data within 0.09 × 10^{-6} K^{-1} over the whole temperature range.
5. It is also focused on that no distinguishable differences in LTEC were observed between the annealed SRM specimen and machined commercially available specimens.

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