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High-temperature thermal expansion of six metallic elements measured by dilatation method and X-ray diffraction

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The thermal expansion of silver, gold, copper, nickel, molybdenum and tungsten has been measured by the dilatation method. The thermal expansion values of silver, gold, copper and nickel have also been evaluated from lattice parameter measurements by X-ray diffraction. These six metals were found to exhibit a nearly uniform expansion over the temperature ranges covered. The thermal expansion values obtained by the dilatation method are in good agreement with those determined by X-ray diffraction. The present results appear also to agree well with those reported previously in the literature.

1. Introduction

There is an increasing need for thermal expansion data of various metals at high temperature, not only for application to composite refractory materials but also for discussion of thermodynamic properties such as the entropy of metals [1, 2]. However, the accuracy of the available information is not quantitatively sufficient in some cases to allow a definite interpretation of the thermal properties of metals with respect to the detected changes resulting from temperature variation.

The main purpose of this paper is to present thermal expansion data for six metallic elements, newly measured by the dilatation method and by X-ray diffraction.

2. Experimental procedures

The photograph in Fig. 1 shows an overall view of the dilatometer (Rigaku Denki Co., Tokyo) which consists of the data acquisition and computing unit with recorder, furnace and differential displacement detecting unit. The principle of this apparatus is to measure the difference in length change arising from the temperature variation between the sample and a standard material. Fig. 2 is a schematic diagram of the detecting unit. Both the sample and a standard material are sandwiched between the detect bar and the alumina support tube under a force of 5 g weight. The lower parts of the detect bars are connected to the magnetic core and coil, and their relative position can be detected by measuring the impedance of the coil. These detect bars are also balanced by the holding forces of the samples. The temperature of the sample is measured by a Pt/Pt–13% Rh thermocouple inserted into a hole in the standard material. The samples can be heated at a constant rate of 8 deg min^{-1} by the programmable temperature controller.

Alumina is frequently used as a standard material for the high-temperature measurement of thermophysical properties. However, its thermal expansion data are known to depend upon the quantity of polycrystalline material produced in the sintering process, and thus there often exist quantitative differences in detail between the measurements on one sample and another. For this reason, a cylindrical platinum rod (5 mm in diameter and 20 mm in length) with a hole for temperature measurement was employed in this work as a standard material, because reliable thermal expansion data for platinum are available [3], and with no phase transformation over a wide temperature range up to about 2000 K it is very well suited for the present purpose.

The measurements by the dilatation method were carried out under an atmosphere of Ar + 7% H_2 gas mixture, except for gold which was measured in air.

The total uncertainty of the thermal expansion data obtained by the present dilatation method was estimated to be within $\pm 2\%$ in the temperature range covered, based on the results of several measurements for copper and molybdenum or quartz, which is known to have a low and uniform thermal expansion.

X-ray diffraction studies under the atmosphere of Ar + 7% H_2 gas mixture were done with a diffractometer whose X-ray tube and detector rotate at the same angular rate in opposite directions around the horizontal axis. The techniques, including the high-temperature chamber and procedures for the analysis of measured intensity data, were almost identical to those employed in the previous high-temperature structural study of some refractory metals such as molybdenum and tungsten [4]. These procedures are now well established and reported in detail [4], and thus not duplicated here. The lattice parameters by X-ray diffraction were calculated by the Cohen least-squares method [5] and the uncertainty is about

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TABLE I Linear thermal expansions of silver, gold, copper, nickel, molybdenum and tungsten by the dilatation method

Temperature (K)	Linear thermal expansion (%)					
	Ag	Au	Cu	Ni	Mo	W
400	0.20	0.15	0.17	0.14	0.05	0.05
500	0.39	0.29	0.35	0.28	0.11	0.09
600	0.59	0.45	0.53	0.43	0.17	0.14
700	0.81	0.60	0.72	0.60	0.23	0.19
800	1.03	0.76	0.91	0.76	0.29	0.24
900	1.27	0.93	1.12	0.93	0.36	0.29
1000	1.51	1.10	1.36	1.11	0.43	0.34
1100		1.28	1.68	1.29	0.49	0.39
1200				1.49	0.58	0.45
1300				1.70		0.51
1400				1.91		0.56
1500				2.13		0.60
1600				2.36		0.65
1700						0.68

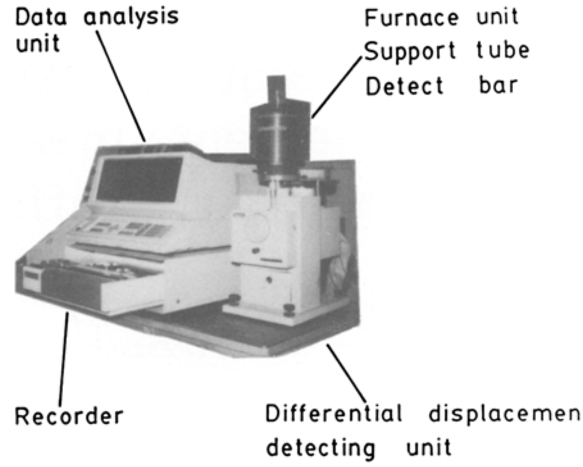

Figure 1 An overall view of the dilatometer, which consists of the data acquisition and computing unit with recorder, furnace and differential displacement detecting unit.

$\pm 0.01\%$, while that of thermal expansion is about 2 or 3%.

Samples of 5 mm diameter and 20 mm length for dilatation were purchased from Wakoh Chemicals and Tanaka Noble Metal Co. Ltd., and were of nominal purity 99.9% for silver, gold, nickel, molybdenum and tungsten, and 99.99% for copper. Powder metal samples were used for X-ray diffraction.

3. Experimental results and discussion

The thermal expansion data obtained by the dilatation method are summarized in Table I. The lattice parameters and their variation with respect to temperature change measured by X-ray diffraction are given in Table II. As easily seen from the results in Figs 3 to 5, the six metals appear to exhibit a nearly uniform expansion over the temperature range covered. They are best represented by the equation

$$\Delta L/L = A_1(T - 293) + A_2(T - 293)^2 \quad (1)$$

where L is the length of a sample at temperature 293 K and ΔL is the change in length between 293 K and temperature T on the absolute temperature scale. The values of A_1 and A_2 determined in this work for the six metallic elements are tabulated in Table III. It may be noted that these coefficients are applicable over the quoted temperature range with a mean deviation of $\pm 0.3 \times 10^{-2}$ for dilatation results and of $\pm 0.3 \times 10^{-3}$ for X-ray diffraction results.

Some specific comments are given below for each element.

3.1. Silver, gold, copper and nickel

The structures of silver, gold, copper and nickel were found to remain face-centred cubic over the quoted temperature range, just as they are at room temperature.

Mauer and Bolz [6] made measurements on silver over the temperature range from 273 to 1150 K by means of X-ray diffraction. On the other hand, Esser and Eusterbrock [7] reported the thermal expansion of

TABLE II Lattice parameters, a, and linear thermal expansions of silver, gold, copper and nickel from 293 K to stated temperatures by X-ray diffraction

Run No.	Silver			Gold			Copper			Nickel		
	T (K)	a (nm)	$\Delta L/L$ (%)	T (K)	a (nm)	$\Delta L/L$ (%)	T (K)	a (nm)	$\Delta L/L$ (%)	T (K)	a (nm)	$\Delta L/L$ (%)
1	293	0.4086	0	293	0.4072	0	293	0.3613	0	293	0.3524	0
2	576	0.4109	0.59	574	0.4091	0.41	577	0.3630	0.50	578	0.3538	0.40
3	671	0.4117	0.78	676	0.4097	0.56	680	0.3636	0.68	676	0.3544	0.57
4	774	0.4126	1.00	777	0.4103	0.72	777	0.3644	0.88	825	0.3552	0.80
5	875	0.4135	1.22	875	0.4110	0.88	874	0.3650	1.06	1007	0.3563	1.11
6	975	0.4144	1.45	971	0.4117	1.05	979	0.3658	1.26	1123	0.3571	1.33
7	1076	0.4155	1.71	1075	0.4125	1.23	1076	0.3667	1.51	1256	0.3581	1.62
8	1174	0.4166	1.99	1179	0.4134	1.47	1179	0.3672	1.65	1345	0.3587	1.79
9	1228	0.4172	2.13	1280	0.4141	1.69	1275	0.3684	2.01	1433	0.3595	2.02
10	293	0.4083	0	1324	0.4145	1.75	1343	0.3692	2.21	1553	0.3603	2.24
11	574	0.4109	0.59	293	0.4075	0	293	0.3612	0	1676	0.3615	2.58
12	676	0.4116	0.78	576	0.4091	0.41	574	0.3634	0.51	293	0.3524	0
13	775	0.4127	1.01	680	0.4096	0.56	672	0.3636	0.67	629	0.3542	0.51
14	878	0.4137	1.22	772	0.4103	0.72	775	0.3644	0.88	754	0.3548	0.68
15	977	0.4145	1.45	874	0.4109	0.88	874	0.3651	1.06	976	0.3562	1.08
16	1078	0.4154	1.70	978	0.4118	1.05	978	0.3657	1.26	1107	0.3570	1.31
17	1171	0.4167	2.01	1074	0.4124	1.24	1080	0.3666	1.51	1225	0.3578	1.53
18	1226	0.4175	2.14	1172	0.4133	1.46	1176	0.3671	1.64	1383	0.3590	1.87
19				1276	0.4144	1.69	1271	0.3685	2.03	1543	0.3604	2.27
20				1325	0.4146	1.75	1337	0.3691	2.20	1671	0.3614	2.55

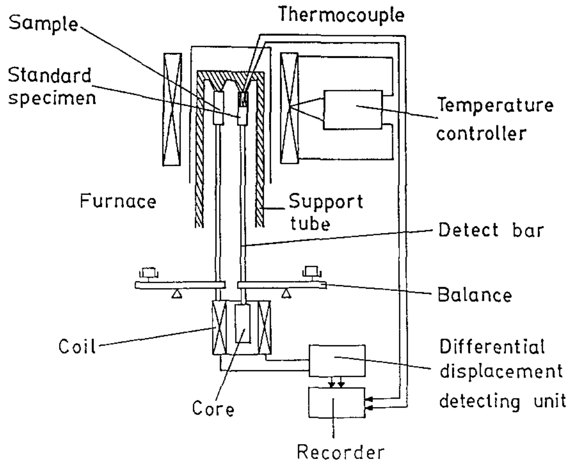

Figure 2 Schematic diagram of the dilatometer presently used.

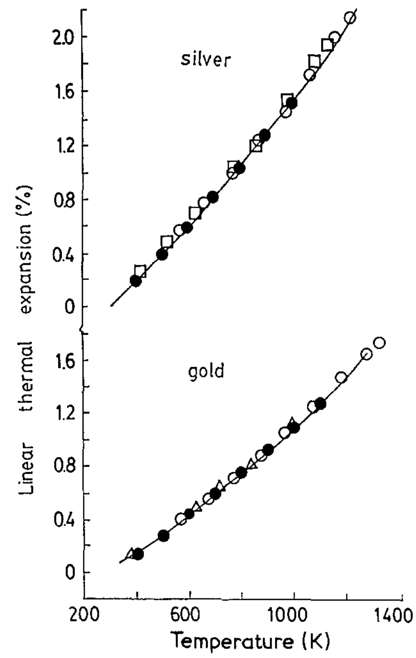

Figure 3 Linear thermal expansion of silver and gold. (○) This work (X-ray); (●) this work (dilatometer); (——) Esser and Eusterbrock [7]; (□) Mauer and Bolz [6]; (△) Nix and MacNair [8].

silver measured by the comparator method. As shown in Fig. 3, the present thermal expansion data for silver agree well with those of Mauer and Bolz [6] and Esser and Eusterbrock [7].

Nix and MacNair [8] determined the thermal expansion of gold between 86 and 1003 K by an interferometric dilatometer involving photographic recording. Esser and Eusterbrock [7] also provide thermal expansion data for gold between 273 and 1223 K. As shown in Fig. 3, the thermal expansion data reported here agree well with those of the earlier workers.

The present results for copper obtained by both dilatation and X-ray diffraction are plotted in Fig. 4 with those obtained by Esser and Eusterbrock [7] and Fieldhouse *et al.* [9]. Their results agree well with ours.

Hidnert [10] measured the thermal expansion of nickel, which was cold-swaged and annealed at 1223 K, by a micrometric thermal expansion apparatus between 293 and 1173 K and Allen [11] made measurements over the temperature range from 293 to 1033 K. Their results are given in Fig. 4 with the values obtained in this work, and ours appear to be in good agreement with the data previously reported.

3.2. Tungsten and molybdenum

Tungsten and molybdenum are known to remain body-centred cubic up to about 2613 K by X-ray diffraction, just as they are at room temperature [4].

As shown in Fig. 5, the present thermal expansion data for molybdenum by the dilatation method reported here are in good agreement with those of Worthing [12] and Demarquay [13], who used the comparator method, and the X-ray diffraction results of Waseda *et al.* [4].

Demarquay [13] reported thermal expansion data for tungsten at temperatures between 293 and 2423 K by X-ray diffraction and Levingstein [14] reported the thermal expansion values of tungsten between 297 and 1422 K by the comparator method. It is easily seen from Fig. 5 that the present results obtained by the dilatation method agree well with those reported previously as well as the X-ray diffraction results of Waseda *et al.* [4].

4. Conclusion

To summarize the results of this work, the thermal expansion data for silver, gold, copper, nickel, molybdenum and tungsten obtained by the dilatation method and those of silver, gold, copper and nickel evaluated from lattice parameter measurements by X-ray

TABLE III The values of constants A_1 and A_2 in Equation 1 for various metals

Element	A_1	A_2	Temperature range (K)	Method
Ag	1.77×10^{-5}	5.16×10^{-9}	293 to 1000	Dilatometer
	1.81×10^{-5}	4.74×10^{-9}	293 to 1228	X-ray diffraction
Au	1.36×10^{-5}	2.74×10^{-9}	293 to 1100	Dilatometer
	1.27×10^{-5}	4.25×10^{-9}	293 to 1325	X-ray diffraction
Cu	1.38×10^{-5}	7.96×10^{-9}	293 to 1100	Dilatometer
	1.46×10^{-5}	5.51×10^{-9}	293 to 1343	X-ray diffraction
Ni	1.30×10^{-5}	3.91×10^{-9}	293 to 1600	Dilatometer
	1.23×10^{-5}	4.40×10^{-9}	293 to 1676	X-ray diffraction
Mo	5.08×10^{-6}	1.41×10^{-9}	293 to 1200	Dilatometer
	3.91×10^{-6}	1.81×10^{-9}	298 to 2614	X-ray diffraction*
W	4.97×10^{-6}	3.16×10^{-11}	293 to 1700	Dilatometer
	5.37×10^{-6}	1.81×10^{-10}	298 to 2621	X-ray diffraction*

*Taken from Waseda *et al.* [4].

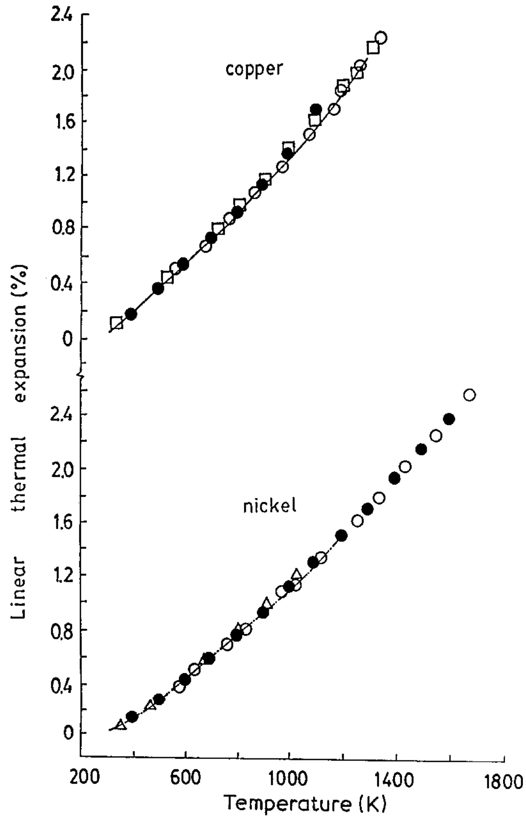

Figure 4 Linear thermal expansion of copper and nickel. (○) This work (X-ray); (●) this work (dilatometer); (——) Esser and Eusterbrock [7]; (□) Fieldhouse *et al.* [9]; (–·–) Hidnert [10]; (△) Allen [11].

diffraction appear to be quite reliable, as can be judged by comparing them with the data reported previously in the literature. It would be interesting, therefore, to extend the present thermal expansion values for investigating various high-temperature properties of metals.

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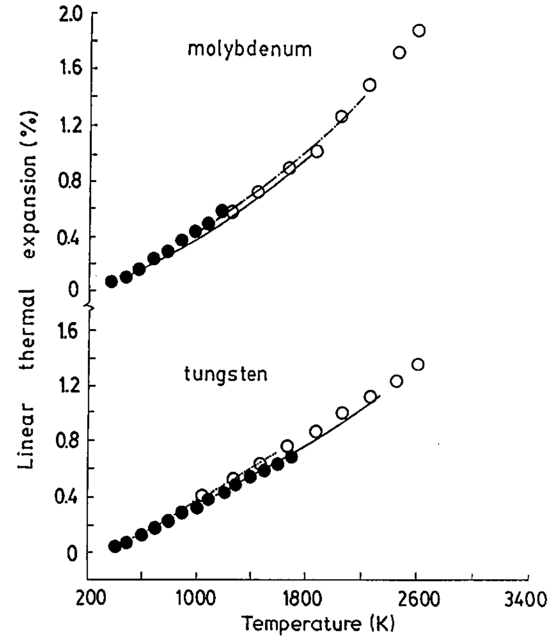

Figure 5 Linear thermal expansion of molybdenum and tungsten. (●) This work (dilatometer); (○) Waseda *et al.* [4] (X-ray); (——) Demarquay [13]; (–·–) Worthing [12]; (····) Levingstein [14].

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