# HEAT CAPACITY OF GOLD FROM 80 TO 1000 K \*

#### YOICHI TAKAHASHI and HIDETOSHI AKIYAMA \*\*

Department of Nuclear Engineering, University of Tokyo, Hongo, Bunkyo-ku, Tokyo 113 (Japan)

(Received 30 June 1986)

### ABSTRACT

The heat capacity of gold has been measured by laser-flash calorimetry in the temperature range 80-1000 K. The results are compared with available low- and high-temperature heat capacities, and revised thermodynamic values of gold are given.

#### INTRODUCTION

Gold seems to be attractive as a possible standard material for high-temperature heat-capacity measurement, because of its relative inertness, high purity, and fabricability [1]. However, its heat-capacity values are not well established above room temperature. Although several authors [2-7] have reported heat capacities, the values differ by about 6% over the range 300-1200 K. It is therefore desirable to investigate the heat capacity of gold over a wide temperature range with sufficient accuracy.

In the present paper, we reported heat-capacity measurements on pure gold by laser-flash calorimetry [8] from 80 to 1000 K. The results are compared with other available data, and the revised thermodynamic values of gold are given.

#### EXPERIMENTAL

Measurements were made on 99.9% pure gold, a product of Ishifuku-kogyo Co. Two experimental specimens in the form of 10 mm diameter disks were used. The thickness and weight of the two specimens were 2.0 and 2.5 mm, and 2.98710 and 3.76068 g, respectively.

<sup>\*</sup> Dedicated to Professor Syûzô Seki in honour of his contribution to Calorimetry and Thermal Analysis.

<sup>\*\*</sup> Present address: Nuclear Energy Group, Toshiba Corp., Isogo, Yokohama, Japan.

Measurements were made with an improved laser-flash calorimeter, the details of which were described in a previous paper [8]. Experimental results on an alumina reference sample agreed with the National Bureau of Standards within  $\pm 0.5\%$  over the temperature range from 100 to 800 K, and within  $\pm 1\%$  from 800 to 1100 K.

A brief outline of the measurements reported here is as follows. First, the heat capacity of the sample at room temperature was determined by comparison with an  $\alpha$ -alumina reference sample. In these measurements the same absorbing disk made of glassy carbon (mass 0.0230 g) and a small mass (less than 0.002 g) of Apiezon-N grease as an adhesive were used. Correction was made for these materials using available data [9,10], and their total contributions to the gross heat capacity were determined as less than 3%. The temperature dependence of the heat capacity was then determined on the gold sample. The front surface of the sample was coated with a thin layer of Dry Graphite Film (Miracle Power Products Corp., Cleveland, OH) for these measurements.

## **RESULTS AND DISCUSSION**

The experimental results are listed in Table 1 in chronological order and are shown in Fig. 1 as a function of temperature. A molar mass of 196.9665 g mol<sup>-1</sup> was used for gold, and no correction for impurities was made. The heat capacities were reproducible within  $\pm 1.5\%$  on different series of experiments over the whole temperature range investigated.



Fig. 1. Molar heat capacity of gold. ( $\bigcirc$ ) Present work, ( $\longrightarrow$ ) Geballe and Giauque [2] and Franzosini and Clusius [3], (----) Jaeger et al. [4], (---) Vollmer and Kohlhaas [5], (----) Cordoba and Brooks [6], (-+-) Ferrier [7].

TABLE 1

Experimental heat capacity of gold  $(M(Au) = 196.9665 \text{ g mol}^{-1})$ 

<b>T</b>	C	 T	C	<u>т</u>	<u> </u>	 Т	C
1 ( <b>K</b> )	$C_p$	I (K)	$C_p$	$(\mathbf{K})$	$C_p$	1 ( <b>K</b> )	$C_p$
( <b>K</b> )	$(\mathbf{J} \mathbf{K})$ $(\mathbf{mol}^{-1})$	( <b>K</b> )	$(\mathbf{J} \mathbf{K})$ mol <sup>-1</sup> )	( <b>K</b> )	$mol^{-1}$	( <b>K</b> )	$mol^{-1}$
Sarias I		Sariac IV		Sarias VI		Sarias VII	,
200.01	25 11	806.16	28.26	86 1 <i>A</i>	20.40	82 74	20.12
299.01	25.44	791.00	20.20	00.14	20.49	04.14	20.12
277.20	25.27	780.11	27.60	80.70 80.40	20.03	0 <del>4</del> .JZ 96.21	20.18
290.90	25.41	787.41	27.00	07.40	20.77	00.51	20.34
299.01	25.55	210 441	27.90	92.37	21.12	00.07	20.44
299.11	25.20	819.00	20.09	95.00	21.40	07.04	20.36
290.94	25.22	843.99 871 31	20.32	90.74	21.50	91.07	20.93
270.77	25.22	0/1.21 807 20	28.50	101.70	21.05	93.13	21.00
299.12	25.26	897.39	28.00	104./1	21.98	98.42	21.38
Curing II		928.41	28.79	107.00	22.11	101.62	21.65
Series II	25 (2	953.41	28.85	111.91	22.23	104.77	21.85
324.52	25.63	981.79	29.09	110.10	22.55	107.85	22.07
346.30	25.72	1002.19	29.37	120.21	22.55	110.86	22.19
370.53	25.71	a · •		124.24	22.80	115.33	22.43
394.32	25.95	Series V		128.19	23.04	119.72	22.61
423.39	25.94	327.62	25.34	132.04	23.11	124.02	22.86
443.49	26.14	362.79	25.69	135.89	23.23	128.24	23.05
463.91	26.20	382.39	25.93	139.74	23.40	132.01	22.92
483.92	26.53	401.80	25.81	143.65	23.41	136.07	23.24
500.27	26.25	442.71	26.12	147.64	23.53	140.04	23.34
513.00	26.51	477.94	26.36	153.25	23.63	143.93	23.31
532.20	26.46	492.51	26.37	159.27	23.71	147.74	23.50
546.77	26.54	516.97	26.53	160.88	23.81	151.91	23.54
559.02	26.85	539.25	26.49	165.27	23.89	156.58	23.74
572.19	26.66	559.44	26.67	170.12	24.13	161.31	23.75
590.65	26.80	581.66	26.89	174.65	24.08	165.93	23.74
604.06	26.62	612.92	27.02	179.71	24.34	170.51	23.98
594.82	27.15	634.60	27.25	185.13	24.22	175.48	24.16
620.01	27.10	658.56	27.23	190.65	24.36	181.06	24.02
645.55	27.07	682.99	27.42	195.91	24.35	187.00	24.17
672.60	27.46	709.28	27.65	201.09	24.47	192.86	24.23
		734.34	27.84	207.69	24.48	198.28	24.27
Series III		756.55	27.87	214.63	24.59	204.96	24.45
393.29	25.77	781.69	28.05	221.06	24.88	212.19	24.54
498.80	26.13	806.17	28.18	227.34	24.92	218.93	24.76
500.64	26.35	827.42	28.44	233.01	24.76	225.93	24.84
510.24	26.30	854.44	28.57	239.85	24.96	233.35	24.95
536.59	26.78	878.12	28.66	246.31	24.89	239.92	25.04
562.23	26.72	900.84	28.83	251.51	24.97	246.40	25.12
589.96	26.65	926.75	29.06	256.90	25.02	253.25	25.11
618.35	27.04	955.44	29.28	263.41	25.13	259.56	25.31
651.76	27.30	977.45	29.41	269.22	25.31	271.80	25.33
682.48	27.41			274.98	25.29	277.74	25.39
711.38	27.42	Series VI		288.28	25.16	284.23	25.60
742.10	27.64	80.91	20.05	293.14	25.29	290.41	25.41
772.61	27.84	82.71	20.13	298.31	25.50		
		84.44	20.47				

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TABLE 2

T	$C_p^0$	S <sup>0</sup>	$H^0(T) - H^0(298)$	$-[G^{0}(T) - H^{0}(298)]/T$
(K)	$(\mathbf{J}^{F}\mathbf{K}^{-1} \text{ mol}^{-1})$	$(J K^{-1} mol^{-1})$	$(J \text{ mol}^{-1})$	$(J K^{-1} mol^{-1})$
80	19.68	16.74	- 5188	81.59
100	21.49	21.35	- 4775	69.09
150	23.47	30.50	- 3643	54.79
200	24.39	37.39	-2444	49.61
250	24.94	42.90	-1210	47.74
298.15	25.31	47.32	0.0	47.32
300	25.32	47.48	46.84	47.32
400	25.90	54.85	2609	48.32
500	26.40	60.68	5224	50.23
600	26.91	65.54	7890	52.39
700	27.47	69.73	10609	54.57
800	28.07	73.43	13385	56.70
900	28.74	76.78	16225	58.75
1000	29.47	79.84	19135	60.71

Molar thermodynamic properties of gold ( $M(Au) = 196.9665 \text{ g mol}^{-1}$ )

Figure 1 shows a comparison of the present heat capacities with those from other investigations. At 298.15 K the present value for the heat capacity is  $(25.31 \pm 0.15)$  J K<sup>-1</sup> mol<sup>-1</sup>, which can be compared with the value of Geballe and Giauque [2], 25.36 J K<sup>-1</sup> mol<sup>-1</sup>. At temperatures below 300 K, the present values agree well with the literature values [2,3]. Above 300–1000 K, our data lie between the two sets of literature values; namely 2–3% higher than the values of Jaeger et al. [4] and Vollmer and Kohlhaas [5], and 2–3% lower than those of Cordoba and Brooks [6] and Ferrier [7].

The close agreement between our data and literature values below 300 K indicates the reliability of the present investigation. Using the present values, the smoothed heat capacity and the derived thermodynamic functions were calculated by a least-squares method. The resulting values at selected temperatures are given in Table 2, where the entropy value at 80 K is taken from the value of Geballe and Giauque [2]. It is to be noted that the resulting values agree very well with the selected values of Hultgren et al. [11]. Their heat-capacity values at 400, 700 and 1000 K are 25.81, 27.24 and 28.83 J K<sup>-1</sup> mol<sup>-1</sup> and lower by 0.3, 0.8 and 2.2%, respectively, than the present values.

## REFERENCES

- 1 C.R. Brooks and E.E. Stansbury, Can. Metall. Q., 13 (1974) 345.
- 2 T.H. Geballe and W.F. Giauque, J. Am. Chem. Soc., 74 (1952) 2368.
- 3 P. Franzosini and K. Clusius, Z. Naturforsch., Teil A, 18 (1963) 1243.

- 4 F.M. Jaeger, E. Rosenbohm and J.A. Bottems, Proc. R. Acad. (Amsterdam), 35 (1932) 772.
- 5 O. Vollmer and R. Kohlhaas, Z. Metallkd., 59 (1968) 273.
- 6 G. Cordoba and C.R. Brooks, Phys. Statos Solidi A, 6 (1971) 581.
- 7 A. Ferreir, cited in AGARD-R-606, by E. Fitzer, 1973, p. 57.
- 8 Y. Takahashi, H. Yokokawa, H. Kadokura, Y. Sekine and T. Mukaibo, J. Chem. Thermodyn., 11 (1979) 379.
- 9 Y. Takahashi and E.F. Westrum, Jr., J. Chem. Thermodyn., 2 (1979) 847.
- 10 H. Kadokura, H. Yokokawa and Y. Takahashi, Calorim. Therm. Anal. (Jpn.), 4 (1977) 52.
- 11 R. Hultgren, P.D. Desai, D.T. Hawkins, M. Gleiser, K.K. Kelley and D.D. Wagman, Selected Values of the Thermodynamic Properties of the Elements, Am. Soc. for Metals, Metal Park, 1973, p. 38.