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Microsecond-Resolution Measurements of the Thermophysical Properties of Liquid Gold

E. Kaschnitz,[1] G. Nussbaumer,[1] G. Pottlacher,[1] and H. Jäger[1]

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New experimental results obtained using an accurate technique for electrical and optical measurements on pulse-heated gold samples are given. An almost-constant current pulse is used for resistive self-heating of the sample over a time interval of 50 μs. Because of the high heating rate, the sample maintains its cylindrical shape in the liquid phase. High pressures are used to extend the investigated range of the liquid phase by suppressing boiling. The stability of the liquid sample is demonstrated by short-time photographs, obtained with a kerr-cell camera. Measurements of current through the sample, voltage drop across the sample, surface radiation, and volume expansion allow the determination of the selected thermophysical properties. Specific enthalpy, electrical resistivity, temperature, density, and their mutual dependencies are obtained. In addition, the enthalpy of melting, as well as the specific heat capacity at constant pressure, is determined.

KEY WORDS: density; electrical resistivity; enthalpy; gold; liquid metals; heat capacity; high temperatures.

1. INTRODUCTION

Experimentally obtained data for liquid gold reported in the literature are quite sparse. The investigated temperature range is limited to about 100 K above melting, with estimated values extending to 1700 K [1]. The use of fast pulse-heating techniques is necessary to reach higher temperatures. These techniques successfully overcome the limitations of conventional static measurements. A recently developed very sensitive high-speed pyrometer [2] allows us to investigate the entire liquid phase of gold starting in the solid. High pressures are used to extend the range of the liquid

[1] Institut für Experimentalphysik, Technische Universität Graz, Petersgasse 16, A-8010 Graz, Austria.

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phase by suppressing boiling. Thermophysical properties, such as specific enthalpy, electrical resistivity, density, and their mutual dependencies, are determined. In addition, the enthalpy of melting, as well as the specific heat capacity at constant pressure, is obtained.

2. EXPERIMENTS AND DATA REDUCTION

Wire-shaped (diameter, 0.5 and 0.25 mm; length, 40 mm) gold samples (purity, 99.999%) are resistively heated far into the liquid phase by passing a large current pulse through them. The energy used for pulse heating is stored in a capacitor bank. Time-resolved (microsecond-time scale) quantities measured are current through the sample, voltage drop across it, surface radiation of the sample, and increase in sample radius, all at pressures from 1 to 2000 bar.

The experimental details of our microsecond pulse-heating apparatus are reported in another paper [3]. The high conductivity of gold has led to changes in our voltage measurement system. Two knife edge probes are used to avoid errors in the measured voltage caused by voltage drops at the clamping contacts. The two voltage signals are measured with ohmic voltage dividers connected to a single ground point.

The enthalpy of a sample heated along an isobaric path may be calculated from

$$H(t) = \frac{1}{m} \int_0^T U(t)\, I(t)\, dt \tag{1}$$

where m is the mass of the sample, $I(t)$ is the time-dependent current through the sample, and $U(t)$ is the time-dependent voltage drop across the sample. Enthalpy is related to that at room temperature. We also obtain values of enthalpy at the beginning of melt H_s, and at the end of melt H_L, and the difference is the enthalpy of melting ΔH. Temperatures are calculated from the surface radiance $J(t)$ using the Planck law

$$T(t) = c_2/\lambda \ln \left\{ 1 + \frac{\varepsilon(t)\, J_m(T_m)}{\varepsilon(T_m)\, J(t)} \left[\exp(c_2/\lambda T_m) - 1 \right] \right\} \tag{2}$$

where c_2 is the second radiation constant, and λ is wavelength. It is assumed that normal spectral emissivity, ε, in the liquid phase is constant. The surface radiance at melting J_m with its known temperature T_m is used as the calibration point. The specific heat capacity at constant pressure, c_P, for liquid gold may be derived from the relation between enthalpy and temperature. The density is determined from photographs, made with a kerr-cell camera. The diameter of the sample is related to the enthalpy

at the instant the photograph is taken. The electrical resistivity ρ_0 without correction for thermal expansion is given by

$$\rho_0(t) = \frac{U(t)\,\pi r^2}{I(t)\,l} \tag{3}$$

where r is the radius of the cold sample and l the length of the sample. As the expansion takes place only radially, it is possible to calculate the electrical resistivity ρ with correction for thermal expansion from

$$\rho(H) = \rho_0(H)\,\frac{d_C}{d(H)} \tag{4}$$

where d is the density of the hot sample, and d_C is the density of the cold specimen. A more detailed description of data reduction is given in a previous paper [4].

3. RESULTS

The large amount of data points obtained for enthalpy, electrical resistivity, volume, and temperature are presented in the form of least-squares fits. Our results for gold are summarized in Table I.

Table I. Thermophysical Properties of Gold: Enthalpy, H, Temperature, T, Density, d, and Electrical Resistivity, ρ_0 and ρ (Without and with Correction for Thermal Expansion)[a]

H (kJ · kg^{-1})	T (K)	d (kg · m^{-3})	ρ_0 ($\mu\Omega$ · m)	ρ ($\mu\Omega$ · m)
148	1337(s)[b]		0.149	
210	1337(1)	17,100	0.296	0.33
225	1435	17,000	0.309	0.35
250	1590	16,700	0.329	0.38
275	1750	16,500	0.350	0.41
300	1910	16,300	0.371	0.44
325	2070	16,000	0.392	0.47
350	2230	15,800	0.412	0.50
375	2390	15,600	0.433	0.54
400	2550	15,400	0.454	0.57
425	2710	15,100	0.475	0.61
450	2870	14,900	0.495	0.64
475	3025	14,700	0.516	0.68
500	3185	14,400	0.537	0.72
525	3345	14,200	0.558	0.76
550	3500	14,000	0.579	0.80
575	3660	13,700	0.599	0.84
600	3820	13,500	0.620	0.89

[a] Reference point: $T = 293$ K, $p = 2000$ bar, and $\rho = 19300$ kg · m^{-3}.
[b] s, solid; l, liquid.

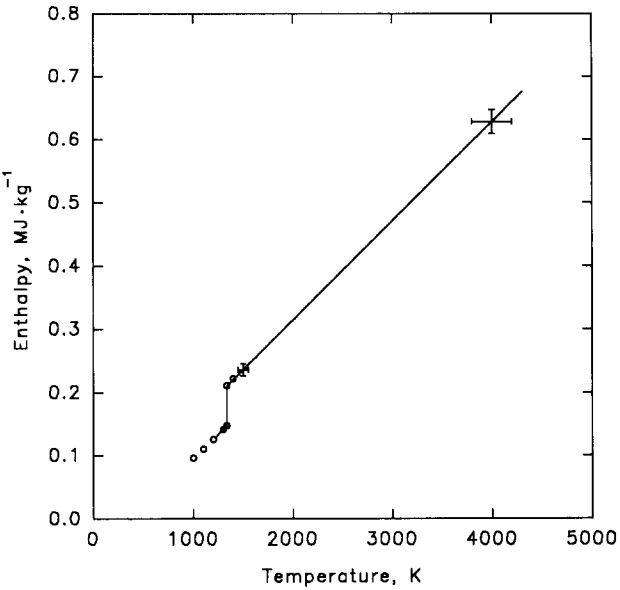

Fig. 1. Enthalpy versus temperature for gold. The solid line represents values of this work; open circles show values of Hultgren et al. [1].

Figure 1 shows variation of enthalpy versus temperature. The least-squares fit to these data for the range $1337 < T < 4300$ K is given by

$$H = 1.57 \times 10^{-4}\, T \tag{5}$$

where H is in $\mathrm{MJ \cdot kg^{-1}}$. From this fit, a value of $157\ \mathrm{J \cdot kg^{-1} \cdot K^{-1}}$ ($7.39\ \mathrm{cal \cdot mol^{-1} \cdot K^{-1}}$) is obtained for c_P, the specific heat capacity at constant pressure. This is in very good agreement with the estimated value of Hultgren et al. [1] of $7.4\ \mathrm{cal \cdot mol^{-1} \cdot K^{-1}}$ and that of Vollmer and Kohlhaas [5] of $156\ \mathrm{J \cdot kg^{-1} \cdot K^{-1}}$; the value of Tester [6] et al. of $7.953\ \mathrm{cal \cdot mol^{-1} \cdot K^{-1}}$ is slightly higher.

Our values for volume, converted to density and plotted against enthalpy, are shown in Fig. 2. The best fit to our data for the range $0.21 < H < 0.6\ \mathrm{MJ \cdot kg^{-1}}$ is given by

$$d = 19033 - 9193.7\ \mathrm{H} \tag{6}$$

where d is in $\mathrm{kg \cdot m^{-3}}$. Densities may also be fit against temperature, and the result for the range $1337 < T < 3820$ K is

$$d = 19033 - 1.4434\ T \tag{7}$$

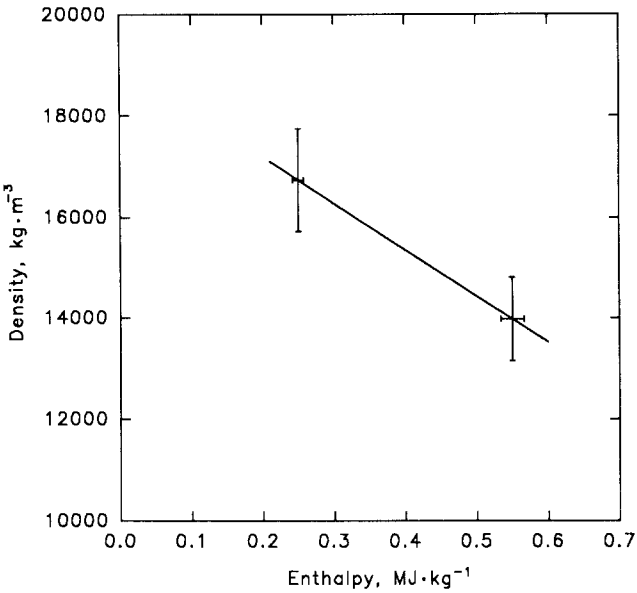

Fig. 2. Density versus enthalphy for liquid gold.

There is a good agreement between our values and data given by Crawley [7].

Electrical resistivities without thermal expansion may be obtained from our data; the results are shown in Fig. 3. The density data allow correction for thermal expansion; the corrected values for electrical resistivity are also shown in Fig. 3. The least-squares fit to the data of the uncorrected electrical resistivity for the range of $0.148 < H < 0.21$ MJ · kg^{-1} is

$$\rho_0 = -0.2019 + 2.371\ H \tag{8}$$

where ρ_0 is in $\mu\Omega \cdot$ m and is for the range of $0.21 < H < 0.6$ MJ · kg^{-1},

$$\rho_0 = 0.1216 + 0.8308\ H \tag{9}$$

The volume-corrected electrical resistivity can be obtained using Eq. (4) as well as the fits given by Eqs. (6) and (9), where the density at room temperature is 19,300 kg · m^{-3}. Our values for resistivity are slightly higher than the data of Matula [8], but they are within the estimated error bars.

Values obtained for the specific enthalpy at the onset and end of melting, melting enthalpy, electrical resistivity at end of melting, and

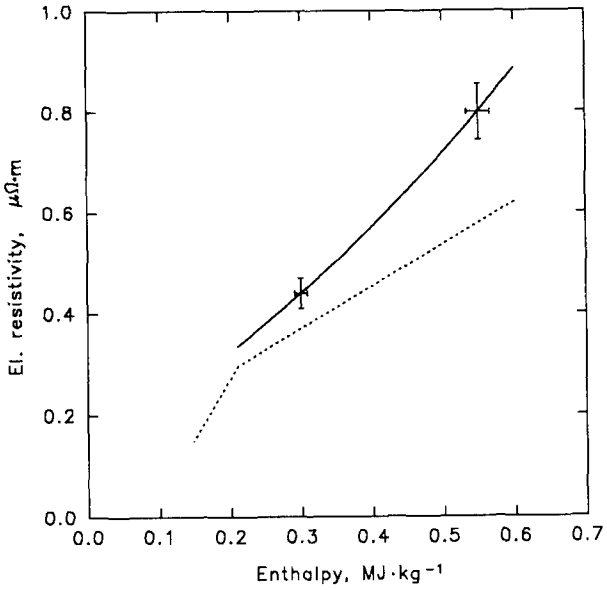

Fig. 3. Electrical resistivity versus enthalpy for gold. The solid line represents electrical resistivity with correction for thermal expansion; the dashed line shows electrical resistivity without correction.

specific heat capacity of liquid gold in the present work and those reported in the literature are given in Table II.

4. ESTIMATION OF ERRORS

Since enthalpy is the most accurate of all the properties measured, with an estimated uncertainty of $\pm 3\%$, all other properties are related to

Table II. Thermophysical Properties of Gold: Specific Enthalpy at Beginning, H_S, and End, H_L, of the Melting Phase Transition, Melting Enthalpy, ΔH, Electrical Resistivity, ρ_L (Thermal Expansion Corrected) at End of Melting, and Specific Heat Capacity, c_P, for the Liquid Phase, Compared with the Values of Other Investigators

H_S (kJ·kg^{-1})	H_L (kJ·kg^{-1})	ΔH (kJ·kg^{-1})	ρ_L ($\mu\Omega$·m)	c_P (J·kg^{-1}·K^{-1})	Ref. No.
148	210	62	0.33	157	This work
147.6	211.3	63.7		157	[1]
145.0	209.5	64.5		156	[5]
148.0	211.0	63.0		169	[6]
		64.8		149	[9]
			0.3108		[8]

enthalpy. The uncertainties in temperature measurements are $\pm 2\%$ in the melting region, increasing to $\pm 5\%$ at the highest temperatures. An error of $\pm 6\%$ is estimated for density. The error bars on the calculated values of uncorrected electrical resistivity are estimated to be $\pm 3\%$; the errors in the corrected electrical resistivity increase to $\pm 7\%$. The estimated uncertainty in the value of melting enthalpy is $\pm 5\%$, and that in the value of specific heat capacity is $\pm 6\%$.

5. CONCLUSIONS

A rapid resistive self-heating technique in the microsecond range has been used to investigate the thermophysical properties of liquid gold. The mutual dependencies among enthalpy, temperature, density, and electrical resistivity have been obtained, as well as the melting enthalpy and the specific heat capacity. The results are compared with the values given for the onset of the liquid phase in the literature, and they are in good agreement. Our measured data extend the investigated range up to boiling of gold.

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