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Improved Thermophysical Measurements on Solid and Liquid Tantalum[1]

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Wire-shaped tantalum samples are resistively pulse heated as part of a coaxially constructed capacitor discharge circuit. With heating rates of more than 10^9 K · s^{-1}, temperatures up to about 10,000 K are reached. The tantalum wire is contained, with water as the surrounding medium, in a high-pressure vessel with sapphire windows and a maximum pressure capability of 5 kbar. Time correlated measurements of the current through the wire and the voltage drop across it, as well as surface radiation and wire expansion, were performed to permit the determination of thermophysical properties of the solid and liquid tantalum.

KEY WORDS: electrical resistivity; enthalpy; high temperatures; liquid metal; pulse method; specific heat; tantalum.

1. INTRODUCTION

Thermophysical properties of tantalum have already been presented in an earlier publication [1]. In these investigations we were interested mainly in heating-rate dependences. The results were not presented in the form of least-squares fits polynomials. At that time, only Shaner et al. [2] and Lebedev and Mozharov [3, 4] had investigated liquid tantalum by a pulse heating method. The temperature scale reported by Shaner et al. was later corrected by Gathers [5]. Meanwhile, Berthault et al. [6] and Hixson et al. [7] also measured thermophysical properties of liquid tantalum. Thus, liquid tantalum is one of the most thoroughly investigated materials. Now it is of interest to extend the measurements to higher temperatures.

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To expand our experimental operation, we had to dismantle the entire measuring system and move it into another room of our laboratory. At the same time, we could improve our measuring system; instead of 200-MHz oscilloscopes we now use 100-Msample, 8-bit digital oscilloscopes. To test the new and better-arranged apparatus, we decided to remeasure the thermophysical properties of liquid tantalum. In this paper we present the results, in the form of least-squares fit polynomials, and compare the results obtained with the new data-aquisition system with those given in literature. Moreover, we reached temperatures of the superheated liquid which have not been investigated up to now.

2. EXPERIMENTAL

Wire-shaped tantalum samples were resistively pulse heated as part of a coaxially constructed capacitor discharge circuit. With heating rates greater than $10^9\,\mathrm{K\cdot s^{-1}}$, temperatures up to about 10 kK were achieved. The tantalum samples (Goodfellow, Cambridge, UK; 99.97%; diameter, 0.25 mm; length, 40 mm) were contained in a high-pressure vessel with two sapphire windows. The medium surrounding the wire sample was water to avoid peripheral gas discharges.

Time-correlated electrical measurements of the current through the wire and the voltage drop across it could be performed simultaneously with those of optical surface radiation and wire expansion. The behavior of the expanding liquid metal column was investigated additionally by analyzing Kerrcell photographs. The measurements were performed under a 2-kbar surrounding pressure. At this pressure, higher temperatures could be achieved in comparison to our earlier measurements and the optical expansion measurements with the help of the "shadowgraph" technique became more accurate. All measuring systems had rise times less than 10 ns. More experimental details are given in Ref. 8.

The measurements allow the determination of thermophysical properties of tantalum such as heat capacity and the mutual dependences among enthalpy, electrical resistivity, temperature, and volume up to highly superheated liquid states.

3. RESULTS AND DISCUSSION

Figure 1 presents temperature T as a function of the specific enthalpy H at a pressure of 2 kbar, as well as the values of Berthault et al. [6] and of Hixson [9]. Both also use the same pressure, but they use argon gas as the surrounding medium. Our results agree very well with their values. We could extend our measuring range up to enthalpy values of $2.0\,\mathrm{MJ\cdot kg^{-1}}$.

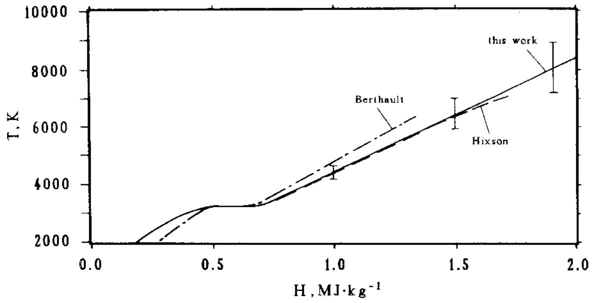

Fig. 1. Temperature T versus enthalpy H for tantalum.

Two different polynomials are used to describe temperature (in kK) as a function of enthalpy (in $\mathrm{MJ \cdot kg^{-1}}$). For the range $0.35 \leqslant H \leqslant 0.51\ \mathrm{MJ \cdot kg^{-1}}$,

$$T = -2.9555 + 30.783H - 51.698H^2 + 29.413H^3 \tag{1}$$

and for the range $0.70 \leqslant H \leqslant 2\ \mathrm{MJ \cdot kg^{-1}}$,

$$T = -0.90816 + 7.7595H - 3.2297H^2 + 0.8679H^3 \tag{2}$$

The dependence of enthalpy versus temperature $H(T)$ is not plotted here, but a least-squares fit was made for the liquid range $3.5 \leqslant T \leqslant 8.5$ kK, which gives H in $\mathrm{MJ \cdot kg^{-1}}$:

$$H = +0.30624 - 9.8927 \times 10^{-3}T + 5.0648 \times 10^{-2}T^2 - 3.1085 \times 10^{-3}T^3 \tag{3}$$

From the constant radiation plateau at the melting transition in the temperature measurement, we obtain enthalpy values of $0.51\ \mathrm{MJ \cdot kg^{-1}}$ at the beginning of melting and $0.66\ \mathrm{MJ \cdot kg^{-1}}$ at the end of melting. These new computed values lie somewhat lower than those reported earlier [1] ($0.54\ \mathrm{MJ \cdot kg^{-1}}$ at the beginning of melting and $0.77\ \mathrm{MJ \cdot kg^{-1}}$ at the end of melting), due to better data aquisition and more precise temperature measurements, as the use of annealed wire samples gives a better resolution of the melting plateau.

In Table I, a summary of enthalpy values at the beginning and at the end of melting reported by different authors is given. The enthalpy values of the other authors for the beginning of melting vary within ±6% and for the end of melting within ±9% from the values of this work.

Our enthalpy values give a heat of fusion of $0.15\ \mathrm{MJ \cdot kg^{-1}}$. A summary of values for the heat of fusion reported by different authors is given in Table II.

Table I. Summary of Values for Enthalpy, Electrical Resistivity, and Relative Volume at the Beginning and the End of Melting (Subscript s, Solid; Subscript l, Liquid; V_0, Volume at 293 K) Reported by Different Authors

Authors	Ref. No.	H_s $(\mathrm{MJ \cdot kg^{-1}})$	H_l $(\mathrm{MJ \cdot kg^{-1}})$	ρ_s $(\mu\Omega \cdot \mathrm{m})$	ρ_l $(\mu\Omega \cdot \mathrm{m})$	V_s/V_0	V_l/V_0
This work		0.51	0.66	1.12	1.29	1.09	1.12
Hixson	9	0.54	0.68		1.29		1.15
Berthault et al.	6	0.50	0.67	1.16	1.34	1.08	1.14
Shaner et al.	2	0.52	0.72	1.14	1.29	1.09	1.15
Gathers	5	0.52	0.72	1.14	1.29	1.09	1.15
Arpaci and Frohberg	14	0.52	0.70				
Frohberg and Betz	15	0.51	0.70				
Lebedev and Savvatimskii	16			1.14	1.26		
Desai et al.	13			1.22	1.31	1.05	

For the specific heat of liquid tantalum we obtained $c_p = 0.250\ \mathrm{J \cdot g^{-1} \cdot K^{-1}}$ for temperatures up to 8.5 kK. The value reported earlier [1], $c_p = 0.245\ \mathrm{J \cdot g^{-1} \cdot K^{-1}}$, is within our uncertainty bars. In Table II, there is also a summary of c_p values, obtained by pulse experiments and by means of levitation calorimetry, given by other authors.

Table II. Summary of the Change of Enthalpy and of Electrical Resistivity During the Melting Transition, the Volume Ratio, and the Ratio of the Resistivities for this Interval (Subscript s, Solid; Subscript l, Liquid) and of Specific Heat for the Liquid Phase of Tantalum Reported by Different Authors

Authors	Ref No.	ΔH $(\mathrm{MJ \cdot kg^{-1}})$	$\Delta\rho$ $(\mu\Omega \cdot \mathrm{m})$	V_l/V_s	ρ_l/ρ_s	c_p $(\mathrm{J \cdot g^{-1} \cdot K^{-1}})$
This work		0.15	0.17	1.03	1.15	0.250
Hixson	7, 9	0.14				0.256
Berthault et al.	6	0.17	0.18	1.05	1.15	0.210
Shaner et al.	2	0.20	0.15	1.05	1.13	0.380
Gathers	5	0.20	0.15	1.06	1.13	0.327
Arpaci and Frohberg	14	0.18				0.233
Frohberg and Betz	15	0.19				0.221
Lebedev and Savvatimskii	16	0.20	0.12			
Desai et al.	13		0.08		1.07	

Our value is in good agreement with that of Hixson [9]; the value of Berthault et al. [6] lies about 16% lower. The value of Shaner et al. [2], corrected by Gathers [5], still seems to be very high. The wide range in values is due to the fact that determination of c_p depends on temperature, which is the least accurate measured quantity in this kind of pulse experiments.

In Fig. 2, the relative volume V/V_0 versus enthalpy H for this work at a pressure of 2 kbar can be seen (V_0 volume at room temperature). The measured values show a nearly linear behavior, with a volume ratio increase of 0.03 for each 0.1 MJ · kg^{-1}. Above 0.7 MJ · kg^{-1}, our mean values are somewhat lower than those of Berthault et al. [6], Ivanov et al. [10], and Hixson [9] but are still within our experimental uncertainty of 8%.

The corresponding polynomial fit to our values is for the range $0.35 \leqslant H \leqslant 1.75$ MJ · kg^{-1}:

$$V/V_0 = +1.0039 + 0.1619H + 7.6431 \times 10^{-3}H^2 + 1.3453 \times 10^{-2}H^3 \quad (4)$$

Table I also gives values for the volume ratio at the melting transition reported by different authors. The ratio of the values at the beginning and the end of melting is given in Table II.

The volume expansion versus enthalpy given in our previous paper [1] showed quite clearly a dependence on the heating rate if a pressure of only 1 bar in the surrounding water is used. For enthalpy values above 1 MJ · kg^{-1}, a very strong increase in the volume ratio was observed. For this work, a surrounding pressure of 2 kbar was used, which showed a quite different behavior. A strong volume increase could not be detected at this pressure.

Ivanov et al. [10] considered the problem of expansion measurements using an optical method with water as the surrounding medium of the

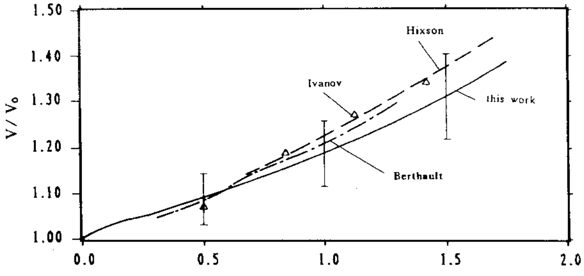

Fig. 2. Relative volume V/V_0 versus enthalpy H for tantalum. V_0, volume at room temperature.

investigated wire. Their conclusion is that if the wire is heated in water, a layer of vapor is created in the water around the wire. In this case, its measured thickness is essentially influenced by the temperature and the rate of increase of temperature in the metal. The layer of vapor, whose refractive index is smaller than that of water, creates an expansion of the investigated metal, which is much stronger than it really would be, as the thickness of the layer is added to the thickness of the sample. This is the case especially for high-melting metals, where very high temperatures are reached.

In this work, we studied the dependence of the wire expansion on the static pressure in the surrounding water with the help of the shadowgraph technique. Using a Kerrcell camera, the pictures (exposure time, 30 ns) show, in the case of a pressure of 1 bar (Figs. 3a and b), a very strong

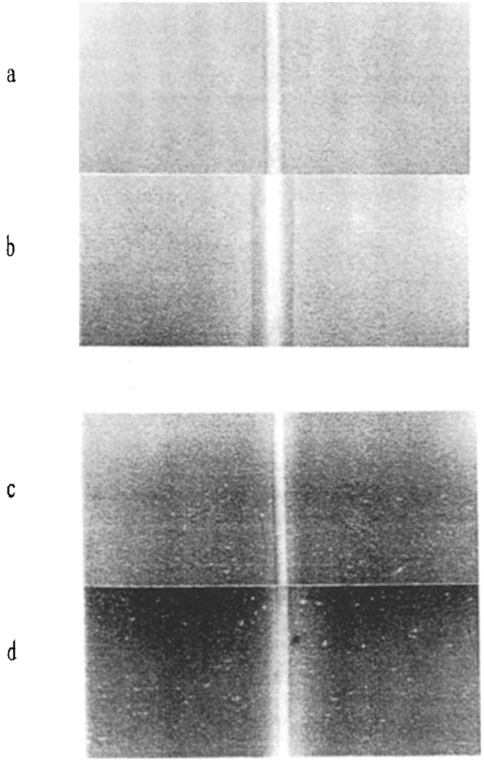

Fig. 3. Short-time picture of the expanding liquid tantal wire. Exposure time, 30 ns. (a) One bar, 4 μs after start of experiment; (b) 1 bar, 20 μs after start of experiment; (c) 2 kbar, 4 μs after start of experiment; (d) 2 kbar, 20 μs after start of experiment.

volume expansion in the liquid phase. This might be due to a change of the refractive index of the water near the wire as reported in Ref. 10, as the inner bright area of the hot sample is surrounded by a relatively broad dark edge. The picture in Fig. 3a was taken 4 μs after the start of the experiment, which is the end of the investigated time interval. The picture in Fig. 3b was taken after 20 μs only to prove the increase in the sample expansion. At that time, generally no more measurements are performed.

At a surrounding pressure of 2 kbar, the pictures do not show such a strong volume expansion of the wire (Figs. 3c and d). The bright area of the sample is in the same dimension as in Figs. 3a and b, but the surrounding darker areas are very thin. Using this pressure, one is definitely above the critical pressure of the surrounding water (220 bar) and a strong change in the refractive index of the water does not occur.

Concluding, we can say that our optical expansion measurements in water with the help of the shadograph technique are a good approach for expansion measurements of pulse-heated wire samples, but one should use a surrounding pressure of at least 1 kbar. The difference between expansion results at pressures of 1 bar and of 2 kbar can be seen in Fig. 4. At 1 bar, too strong volume expansions are observed. This means that our earlier expansion measurements, e.g., on rhenium [11] (at that time the pressure vessel was not yet operating), have to be corrected to lower values.

Figure 5 presents electrical resistivity without volume correction ρ_0 as a function of enthalpy H obtained in this work (2 kbar), as well as the values of Berthault et al. [6], Cezairliyan et al. [12], and Hixson [9]. Our corresponding polynomial fit for ρ_0 (in $\mu\Omega \cdot m$) the range $0.35 \leqslant H \leqslant 1.75\ \mathrm{MJ \cdot kg^{-1}}$ is:

$$\rho_0 = +0.19492 + 2.7409H - 2.4685H^2 + 0.70920H^3 \tag{5}$$

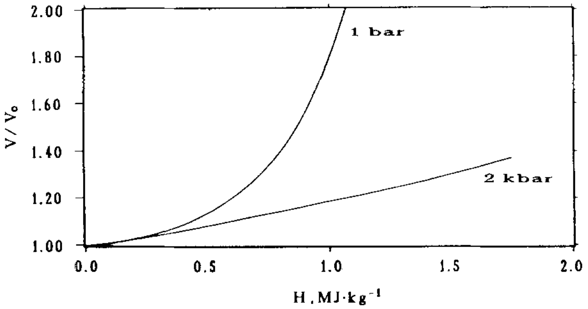

Fig. 4. Volume expansion V/V_0 versus enthalpy H for tantalum at two pressures.

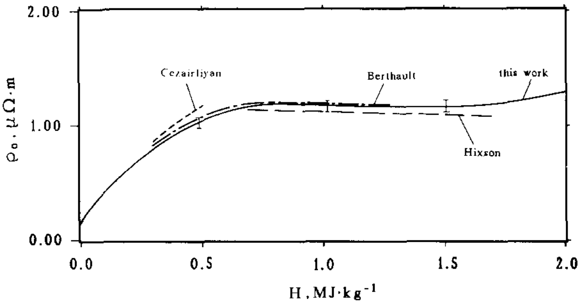

Fig. 5. Electrical resistivity ρ_0 (without volume correction) versus enthalpy H for tantalum.

The resistivity starts at room temperature with a value of about 0.135 $\mu\Omega\cdot$m and has a strong increase while heating up to melting. For the liquid metal the resistivity without volume correction stays nearly constant at a value of 1.17 $\mu\Omega\cdot$m. The values of Cezairliyan are obtained with a millisecond pulse-heating experiment and lie somewhat higher than our values; those of Berthault and Hixson are obtained with a submicrosecond pulse-heating experiment similar to ours and show a good agreement.

Figure 6 presents electrical resistivity with volume correction ρ as a function of enthalpy H for this work, as well as the values of Berthault et al. [6], Desai et al. [13] and Hixson [9], which are all in good agreement. Our corresponding polynomial fit for ρ (in $\mu\Omega\cdot$m) in the range $0.35 \leqslant H \leqslant 1.75$ MJ·kg^{-1} (2 kbar) is

$$\rho = +0.11837 + 3.1730H - 2.6952H^2 + 0.79722H^3 \tag{6}$$

In Table I, also a summary of electrical resistivities at the beginning and at the end of the melting transition of tantalum is given, reported by

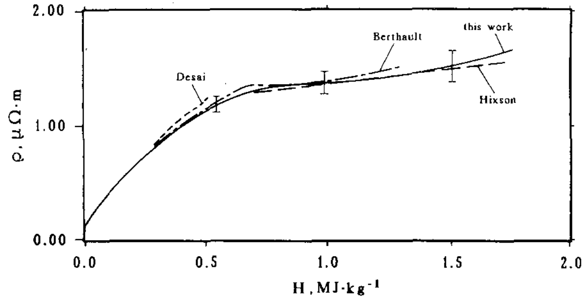

Fig. 6. Electrical resistivity ρ (with volume correction) versus enthalpy H for tantalum.

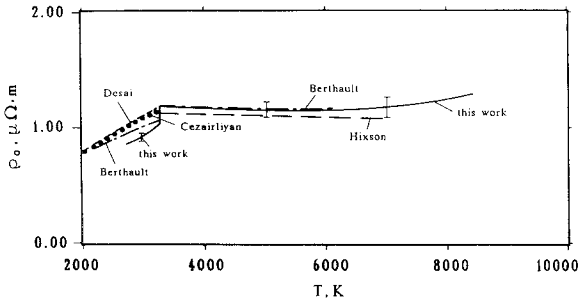

Fig. 7. Electrical resistivity ρ_0 (without volume correction) versus temperature T for tantalum.

different authors. The resistivity values, obtained with pulse experiments, agree within 4% for the beginning and the end of melting. Table II also gives the difference and the ratio of these resistivities, reported by different authors.

For a better comparison with the values given in the literature, Fig. 7 shows the dependence of the electrical resistivity without volume correction ρ_0 (in $\mu\Omega \cdot \mathrm{m}$) versus temperature T. Our corresponding polynomial fit for the range $2.75 \leqslant T \leqslant 3.25$ kK (2 kbar) is

$$\rho_0 = -8.5504 + 9.5882T - 3.3357T^2 + 0.39764T^3 \qquad (7)$$

and for the range $3.3 \leqslant T \leqslant 7.5$ kK (2 kbar),

$$\rho_0 = +1.0070 + 0.1387T - 3.5881 \times 10^{-2}T^2 + 2.7943 \times 10^{-3}T^3 \qquad (8)$$

Figure 8 presents the volume-corrected resistivity ρ (in $\mu\Omega \cdot \mathrm{m}$) versus temperature T. The corresponding polynomial fit for the range $2.75 \leqslant T \leqslant 3.25$ kK (2 kbar) is

$$\rho = -12.018 + 13.249T - 4.6213T^2 + 0.55029T^3 \qquad (9)$$

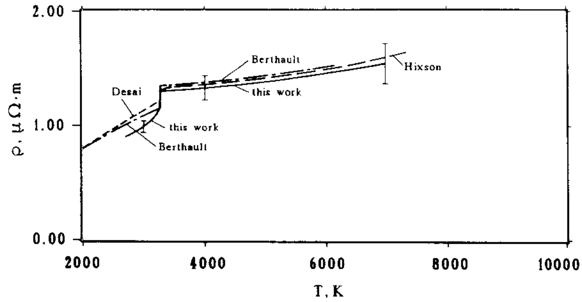

Fig. 8. Electrical resistivity ρ (with volume correction) versus temperature T for tantalum.

and for the range $3.3 \leqslant T \leqslant 7.5$ kK (2 kbar),

$$\rho = +0.92432 + 0.23263T - 4.6616 \times 10^{-2}T^2 + 3.8493 \times 10^{-3}T^3 \quad (10)$$

Much to our surprise, the resistivity values up to melting, obtained by fast pulse experiments, lie a little bit lower than those obtained by slower experiments, e.g., Cezairliyan et al. [12]. For the liquid metal there is a very good agreement between the values in this work and those of Berthault et al. [6] and of Hixson [9].

Table III. Summary of the Mean Values of Temperature, Relative Volume, and Volume-Corrected Electrical Resistivity as a Function of Enthalpy in the Measuring Interval of Our Experiments on Tantalum (s, Solid; l, Liquid)[a]

$H\ (\mathrm{MJ \cdot kg^{-1}})$	T (kK)	V/V_0	$\rho\ (\mu\Omega \cdot \mathrm{m})$
0.35	2.780	1.06	0.91
0.40	2.950	1.07	0.99
0.45	3.100	1.08	1.07
0.51 (s)	3.270	1.09	1.12
0.66 (l)	3.270	1.12	1.29
0.70	3.350	1.13	1.31
0.75	3.475	1.14	1.34
0.80	3.675	1.15	1.34
0.85	3.875	1.16	1.35
0.90	4.075	1.17	1.36
0.95	4.275	1.18	1.37
1.00	4.525	1.19	1.38
1.05	4.700	1.20	1.39
1.10	4.900	1.21	1.40
1.15	5.100	1.22	1.41
1.20	5.250	1.23	1.42
1.25	5.450	1.24	1.43
1.30	5.600	1.26	1.45
1.35	5.800	1.27	1.46
1.40	6.000	1.28	1.48
1.45	6.200	1.29	1.50
1.50	6.400	1.31	1.52
1.55	6.600	1.32	1.54
1.60	6.800	1.34	1.57
1.65	7.000	1.35	1.60
1.70	7.200	1.37	1.63
1.75	7.400	1.39	1.66
1.80	7.700		
1.85	7.900		
1.90	8.100		
1.95	8.400		

[a] Reference point: $T_0 = 293$ K; $p_0 = 2$ kbar.

The volume expansion versus temperature dependence $V/V_0(T)$ is not plotted here, but a least-squares fit has been made for the range $2.75 \leqslant T \leqslant 3.25$ kK (2 kbar):

$$V/V_0 = -1.3393 + 2.4883T - 0.86945T^2 + 0.1026T^3 \tag{11}$$

and for the range $3.3 \leqslant T \leqslant 7.5$ kK (2 kbar),

$$V/V_0 = +0.95634 + 5.6199 \times 10^{-2}T - 2.6656 \times 10^{-3}T^2 + 3.7798 \times 10^{-4}T^3 \tag{12}$$

To complete this presentation of thermophysical properties of liquid tantalum, a summary of mean values for temperature, electrical resistivity, and relative volume as a function of enthalpy up to 2.0 MJ · kg^{-1} is given in Table III.

4. ESTIMATE OF UNCERTAINTY

The data for each individual experiment lie within the uncertainty bars (shown in the figures only for three points at the beginning, middle, and end of the investigated range), which give the experimental uncertainty. An uncertainty of 5% has to be assumed for the enthalpy and of 9% for the electrical resistivity close to the melting transition, increasing for the highest values up to 13%. The uncertainties in the temperature measurements should not exceed 15% for the highest values. The uncertainty in the specific heat also should be lower than 15%.

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