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Experiments on Expanded Liquid Metals at High Temperatures

A. I. Savvatimski[1]

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The electrical resistivity of liquid tungsten was measured using electric pulse heating of the wires inside capillary tubes. Under "fast" heating (10 μs) or "slow" heating (50 μs), the wire expands and fills the inner cavity of the capillary. On the oscillogram traces of the voltage drop across the wire, one can see the phases solid, liquid, fast expansion, and then the moment when the cavity is filled with the metal. Using the voltage drop, current, and volume of the capillary cavity, one can calculate the electrical resistivity, ρ, of the expanded metal. Tungsten densities from 7.5 to 1 $g \cdot cm^{-3}$ (3×10^{22} to 0.5×10^{22} atoms $\cdot cm^{-3}$) were investigated at temperatures from 10×10^3 to 14×10^3 K. For these densities, the electrical resistivity increased from 0.5 to 5m$\Omega \cdot$ cm.

KEY WORDS: density; electrical resistivity; high temperatures; liquid metal; metal vapor; niobium; pulse heating; tungsten.

1. INTRODUCTION

Studies of expanded liquid metals began with low-boiling metals, such as mercury and cesium. Under steady-state conditions, it is easier to perform the experiments when the temperature is not too high. This subject is discussed in the literature [1–3]. In addition to Hg and Cs, Likalter [3] treats the electrical conductivity of expanded liquid copper (at density of the order of 1 $g \cdot cm^{-3}$) and uses the term "gaseous" metal as an equivalent to "expanded" metal.

Dynamic methods appear most attractive for the investigation of liquid metals at high temperatures. This is especially true of the dynamic

[1] High Energy Density Research Center (HEDRC), United Institute IVTAN, Russian Academy of Sciences, Izhorskaya 13/19, Moscow 127412, Russia.

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methods which involve heating by high-density electric current. Major contributions to the experimental investigations of metals under conditions of pulsed electric heating were made by several investigaters [4–20].

A major difficulty in pulsed heating of refractory metal wires in a gaseous environment at atmospheric pressure is that, after melting, the wire is shunted by an electric discharge because of low external pressure, high temperature, and the presence of a high electric potential difference along the wire. To avoid the shunting discharge, the metal wire is placed in a medium of high dielectric strength, for instance, in water, including pressurized water [4, 8, 9, 16], or in a high-pressure gas [7, 10–12, 14, 15, 18].

In 1978, Lebedev and Savvatimski [21] pointed to the possibility of precluding the emergence of shunting discharge during pulsed heating of refractory metals in an atmospher of air. It will suffice to place the wire in a restricted volume, for example, in an insulating capillary with the diameter exceeding the wire thickness. In this case, pulsed heating causes the gap between the capillary and the wire to be filled with metal vapor whose pressure inhibits the emergence of shunting discharge. This makes possible investigations of heated expanding liquid metal at high temperatures.

This technique enabled one to measure the thermal expansion of liquid metals [22] (the specific volume is recorded at the moment the capillary space is filled with metal), as well as to determine the dependence of electric resistance on pressure of tens of kilobars [23, 24] (from the moment of filling the space of the capillary to the moment of destruction of the latter, liquid metal is heated under conditions of rising pressure). The results reported in Refs. 22–24 are discussed by Lebedev and Savvatimski [25].

2. EXPERIMENTS

Tungsten wires 0.08 to 0.15 mm in diameter in glass and quartz capillaries with an inner diameter of 0.2 to 0.45 mm were subjected to pulsed heating by electric current. The capacitors were equipped with ballast resistors to provide quasi-stationary current (Fig. 1). Oscillogram recordings of the voltage drop U (solid line) and current I were used in the calculations.

The electrical resistance of liquid tungsten at the melting point ρ_{liq} was measured by several investigators. Disregarding thermal expansion of the metal, $\rho_{liq} = 127$ [26], 132 [7], and 121 [19] $\mu\Omega \cdot$ cm and, considering thermal expansion, $\rho_{liq} = 137$ [27], 138 [12], and 146 [15] $\mu\Omega \cdot$ cm. In Fig. 1, the ordinate of voltage U, marked ρ_{liq}, corresponds to 127 $\mu\Omega \cdot$ cm. The rest of the characteristic ordinates in this figure may be measured on

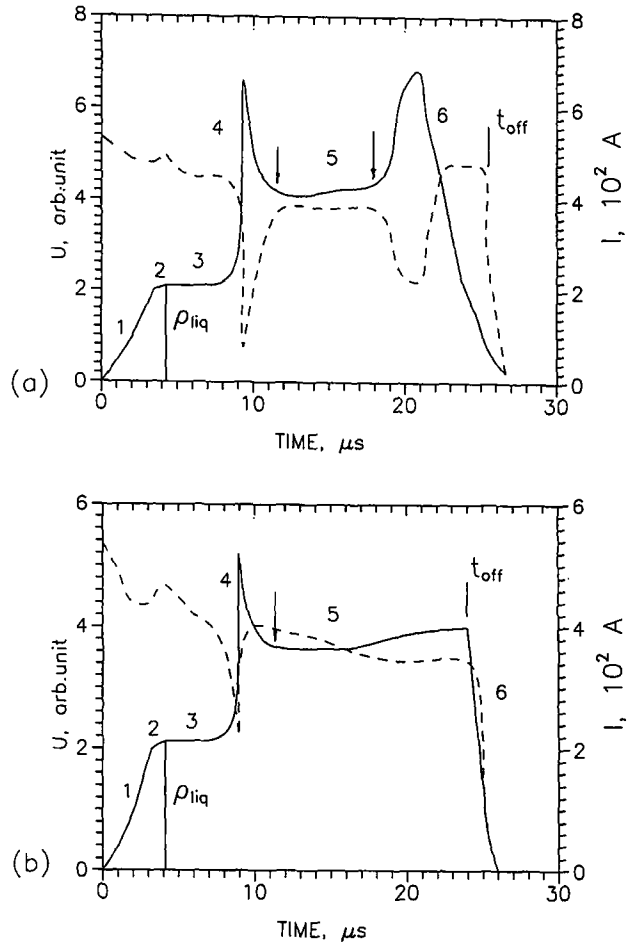

Fig. 1. "Fast" electrical explosion of a tungsten wire in a glass capillary in air at initial atmospheric pressure. (a) Capillary is destroyed after the experiment. Wire: diameter $\phi = 0.08$ mm, and length $l = 16.5$ mm. Capillary inner diameter $d = 0.23$ mm, and outer $D = 4$ mm. Numbers on the oscillogram trace of voltage drop U (arbitrary units): 1, solid state; 2, melting; 3, liquid state; 4, loss of conductivity; 5, the capillary cavity is filled and the capillary itself is safe; 6, the origin or electric disharge in the destroyed capillary. Left-hand arrow (in region 5), the moment the cavity is filled; right-hand arrow, the moment of destruction of the capillary; t_{off}, the moment of forced switching off the current (dashed line); ρ_{liq} (vertical line), the resistivity of liquid tungsten at the melting point ($\rho_{liq} = 0.127$ m$\Omega \cdot$cm [25, p. 18], [26]. For this point current $I = 460$ A, so current density $j = 9 \times 10^6$ A $\cdot$ cm^{-2}. (b) The capillary is safe after the experiment. Tungsten wire: $\phi = 0.08$ mm, and $l = 16$ mm. Capillary: $d = 0.34$ mm, and $D = 5.5$ mm. After the experiment $d \cong 0.4$ mm with the small cooled drops of metal at the inner cavity. Numbers 1–5 and t_{off}, near voltage drop U, are the same as in a.

the scale of ρ_{liq} with due regard for the variation of voltage, current, and cross-sectional area of the conductor. For instance, the electric resistance of liquid metal which completely fills the capillary space (left-hand arrow in region 5, in Fig. 1), $\rho_5 = U_5 I_5^{-1}(S_5 l_5^{-1})$ may be calculated by the equation

$$\rho_5 = \rho_{liq}(U_5 U_{liq}^{-1})(I_5^{-1} I_{liq})(S_5 S_{liq}^{-1})(l_5^{-1} l_{liq}) \tag{1}$$

where $\rho_{liq} = 127\ \mu\Omega \cdot \text{cm}$ [26] is used as the scale for Fig. 1. $l_5 = l_{liq}$, because the capillary length is unchanged. S_5 is the initial cross section of the inner space of the capillary, that is, assumed unchanged until destruction (up to the region 6). S_{liq} is the wire cross section at the melting point in the liquid state, that is, assumed equal to the initial cross section of the wire.

According to our measurements [28], the total imparted energy in regions 1–4 reaches 0.75 of the sublimation energy ($851\ \text{kJ} \cdot \text{mol}^{-1}$ [29]) for tungsten.

Therefore, in region 5 the imparted energy is comparable with the sublimation energy, and the electric resistance is finite.

It is assumed in the calculations that, at the beginning of region 5:

- there is no loss of mass (for control, the capillary was clamped in a vise at the ends with lead pads);
- the volume of the internal space of capillary is constant until destruction; and
- there is no electric discharge inside the space.

The latter assumption is very important; if it is not valid, nonuniform heating of expanded metal is possible. The effect of possible discharge in region 5 on light radiation was verified. If the discharge results in high local temperature, we should see a sharp increase of glow.

Figure 2 illustrates the tungsten wire glow for a wide range of the visible spectrum. One can see a light pulse at the moment of sharp expansion of the wire (region 4). After region 4, light radiation decreases, which is followed by a slight increase during subsequent heating in region 5. In some experiments, this increase was sharper. After the moment t_{off} (the moment of switching off the current), light radiation F decreases slowly as if it was radiation of heated metal rather than that of discharge. However, because the glow was recorded through the capillary tube wall, it is possible to shield radiation during condensation of vapor on the cold wall inside the capillary.

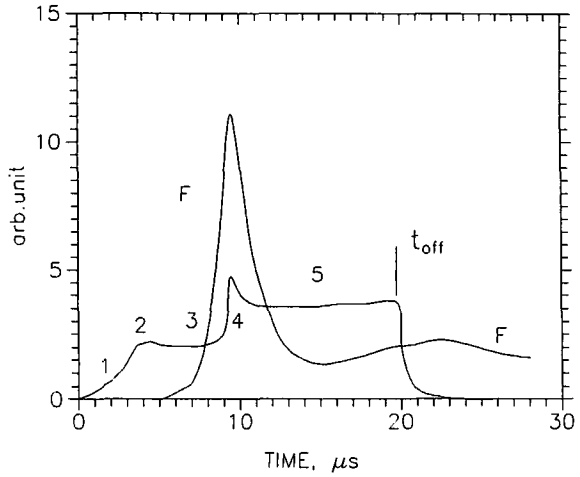

Fig. 2. Light emission F of the tungsten wire through the glass wall of the capillary. Wire: $\phi = 0.08$ mm, and $l = 15$ mm. Capillary: $d = 0.34$ mm, and $D = 5.5$ mm. The capillary is safe after the experiment. The wire and the capillary are shielded by the mask with a transversal slit, so the signal F is from the central part of the wire. Numbers 1–5 near the curve of voltage drop U and t_{off} are the same as in Fig. 1a.

One may note yet another fact that proves the absence of discharge in the right-hand part of region 5. Oscilligrams of U and I demonstrate the metallic properties of expanded liquid tungsten between region 5 and region 6. When the capillary tube deteriorates (right-hand arrow in region 5 of Fig. 1), the voltage U starts increasing simultaneously with the drop of current. In the case of discharge, the pattern would have been quite the reverse.

The rise and fall of the voltage between region 4 and region 5 (Fig. 1), which are outwardly similar with the emergence of discharge, produce additional uncertainty associated with the magnitude of the inductive component $L(dI/dt)$. Experiments with different heating rates revealed a rate of heating of wires in capillaries that does not lead to a sharp drop of current and produces no drop of voltage between region 4 and region 5.

Figure 3 shows a record of voltage U and current I for such "slow" heating: for tungsten (Fig. 3a), the current density $j = 3 \times 10^6$ A $\cdot$ cm^{-2}; and for niobium (Fig. 3b), $j = 3.6 \times 10^6$ A $\cdot$ cm^{-2}. These experiments were performed with longer tubes ($l = 4$ cm). As in the case of "fast" heating of tungsten, Fig. 3a shows the moment of filling the capillary space (left-hand arrow in region 5) and the moment of the beginning of tube destruction (right-hand arrow in region 5).

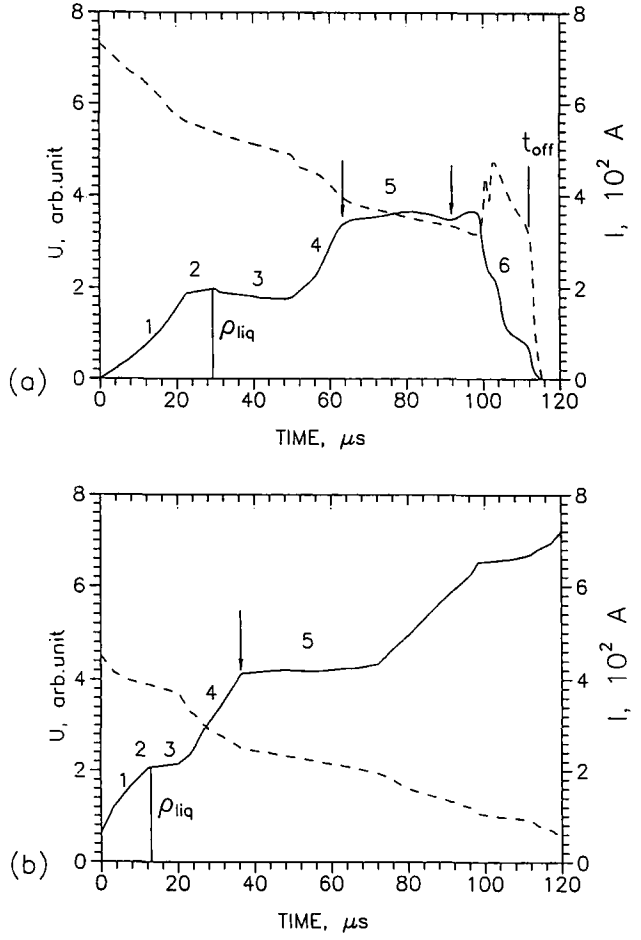

Fig. 3. "Slow" electrical explosion of a metal in capillary in air, at initial atmospheric pressure. (a) Tungsten wire in a quartz capillary. Wire: $\phi = 0.15$ mm, and $l = 40$ mm. Capillary: $d = 0.45$ mm, and $D = 4.5$ mm. Left-hand arrow in region 5 (near voltage drop U), the moment the cavity is filled; right-hand arrow, the moment of destruction of the capillary. Numbers 1–6, ρ_{liq}; t_{off}, the same as in Fig. 1a. For point ρ_{liq}, current (dashed line) $I = 550$ A, so current density $j = 3 \times 10^6$ A · cm^{-2}. (b) Niobium wire in a glass capillary. The capillary is safe after the experiment. Wire: $\phi = 0.12$ mm, and $l = 42$ mm. Capillary: $d = 0.34$ mm, and $D = 5.8$ mm. Arrow, the moment the cavity is filled. Numbers 1–5, near voltage drop U, the same as for Fig. 1a; ρ_{liq}, the resistivity of liquid niobium at the melting point ($\rho_{liq} = 0.108$ mΩ · cm [25, p. 18]). At this point current (dashed line) $I = 400$ A, so current density $j = 3.6 \times 10^6$ A · cm^{-2}.

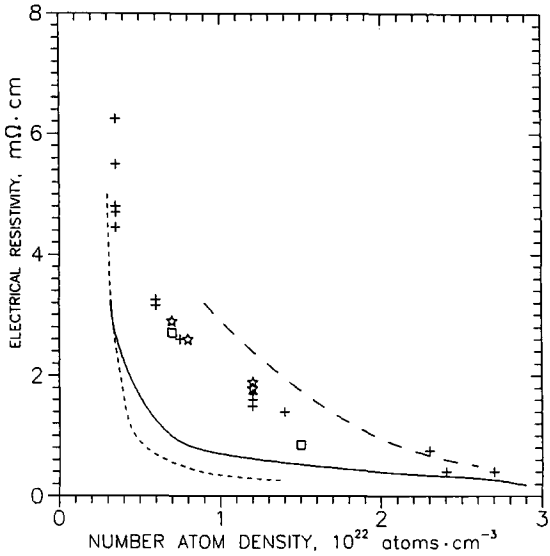

Fig. 4. Electrical resistivity of expanded metals plotted as a function of number atom density. Author's data: crosses, "fast" heating for tungsten; stars, "slow" heating for tungsten. Calculated temperature region 10×10^3 to 14×10^3; open squares, "slow" heating for niobium (no temperature region is known). Literature data for expanded copper: dashed line, electrical explosion copper wires in air [30] (no temperature measurements are given); solid line, electrical explosion copper wires in glass capillary tubes, $T = 10^4$ K for strongly coupled copper plasmas [31]; dotted line, calculated data [35] for copper at $T = 7600$ K.

"Slow" heating of niobium is illustrated in Fig. 3b. A new development in this case consists in that the current was not switched off and the capillary tube remained intact after the experiment. One can see that the current gradually decreases while the voltage continuously increases. The nonmonotonic rise of U may be interpreted as periodic outflow of metal from the ends of the capillary tube. The electric resistance was calculated for the moment indicated by the arrow in Fig. 3b (the moment when the expanding metal thrusts against the wall of the capillary tube). Prior to this moment, the outflow of vapor is unlikely.

Figure 4 gives the electric resistance of expanded liquid tungsten as a function of the number density of atoms n. The latter was calculated as

$$n = N\gamma A^{-1} \tag{2}$$

where N is the Avogadro number, γ is the specific density of the metal at the moment it thrusts against the capillary wall, and A is the atomic weight.

Given in the same figure are the experimental data for similar number densities of copper atoms, available in the literature [30, 31]. The theoretical data of Likalter [3] for low-density copper are close to the data of De Silva and Kunze [31] for strongly coupled copper plasma.

3. DISCUSSION

The temperature and pressure were not measured in the above-discussed experiments. However, it is possible to calculate the temperature level for which the electric resistance of expanded liquid tungsten was measured.

For the liquid phase of tungsten at the melting point, $T = 3690$ K and the enthalpy $H_{liq} = 870$ kJ kg^{-1} [15]. The heat capacity of liquid tungsten was measured by several investigaters to be (in J·kg^{-1}·K^{-1}) 400 [32], 282 [7], 310 [27], 300 [12], and 262 (constant from the melting point to the maximum temperature of measurements 5600 K) [15].

We will assume that heat capacity of liquid tungsten to be constant from the beginning of the liquid state to region 4 (to the maximum of voltage peak in Figs. 1 and 3). For the maximum and minimum values of heat capacity cited above, namely, 400 [32] and 262 J·kg^{-1}·K^{-1} [15], the temperature calculation yields 10,200 and 13,600 K, respectively, for the end of region 4.

Therefore, our measurements of the electric resistance of expanded liquid tungsten relate to the temperature level of 10×10^3 to 14×10^3 K. Unfortunately, it is still impossible to say anything about the uniformity of the metal in the restricted volume under investigation. This is especially true of the minimum investigated densities, for which the experimental points exhibit the maximum scatter (Fig. 4). It is in this case that the gap between the wire and the inside wall of the capillary is maximum and the probability of emergence of shunting discharge is likewise maximum.

For region 5 (Figs. 1 and 3), when the capillary space is filled with metal and the capillary preserves its integrity, the electric resistance varies but only slightly and over a relatively long period of time, in spite of the assumed temperature rise (the temperature may fail to increase in view of the liquid–vapor phase transition). An analogy may be drawn with other liquid metals such as mercury, for which it has been experimentally shown [33] that the increase in the electric resistance of liquid mercury is strictly related to the expansion of metal rather than to its temperature rise. The

data of Gubar and Kikoin [33] are supported by the experimental data of Bradley [34], who investigated mercury and gallium at temperatures in the neighborhood of 200°C. The condition of constancy of volume in Ref. 34 was satisfied by maintaining the pressure at about 8 kbar.

Our previous investigations of liquid tungsten revealed a marked increase in its electric resistance under conditions of restricted volume in the vicinity of the melting point (Ref. 25, p. 21), at the moment of liquid tungsten thrust against the capillary wall. Liquid carbon near the melting point behaves similarly. After a graphite sample fills the capillary, its electric resistivity at constant volume (disregarding the possible expansion of capillary prior to destruction) is doubled (from 1 to 2 $m\Omega \cdot cm$), with the injected energy varying from 16 to 36 $kJ \cdot g^{-1}$ (Ref. 25, p. 68).

It has been further observed by Lebedev and Savvatimski [25] that the electric resistance of liquid tungsten (far from the melting point) in restricted volume remains almost unchanged (or increases slightly) until the destruction of the capillary.

The data obtained by us for low-density tungsten (Fig. 4) may be compared only with the known experimental data for expanded copper. Such measurements were performed in 1969 for the number density of atoms of 1.22×10^{22} to 3.3×10^{22} cm^{-3} under conditions of electrical explosion of copper wire in an atmosphere of air [30]. The variation of volume was recorded by pulsed X-ray technique.

In 1994, De Silva and Kunze [31] performed an electrical explosion of copper wires in glass capillary tubes to find the electric resistance of a dense copper plasma for the number density of atoms in the range 0.28×10^{22} to 2.8×10^{22} cm^{-3} for temperature in the range 10×10^3 to 30×10^3 K.

The data of Ben-Yosef and Rubin [30] for copper (no temperature data are given) and of De Silva and Kunze [31] for copper at 10^4 K are given in Fig. 4 in number atom density, atoms $\cdot cm^{-3}$, for qualitative comparison with the data we obtained for tungsten. Calculated data of Likalter [3, 35] for copper densities in the range 0.3 to 1.5 $g \cdot cm^{-3}$(0.3×10^{22} to 1.4×10^{22}atoms $\cdot cm^{-3}$) at 7600 K are also given in Fig. 4.

4. CONCLUSION

The use of insulating capillary tubes in dynamic experiments with electric heating appears promising from the standpoint of investigation of expanded liquid metals. The measurements of the thermal properties of materials under these conditions may be succesful in the absence of shunting discharges inside the capillary. Obviously, more complete information will be obtained as a result of measurements of the metal temperature

inside the capollary, which should be made with the blackbody model for the liquid state (Ref. 25, p. 58).

The ways of realizing the advantages of using capillary tubes in dynamic experiments are as follows.

(a) During investigations of expanded liquid refractory metals under conditions of initial atmospheric pressure, the function of high external gas pressure or of liquid, which helps preclude the shunting discharge, is served by the layer of vapor of the metal in question in the gap between the capillary and the liquid metal.

(b) The use of capillaries enables one to define the modes of pulsed heating without a sharp loss of conductivity. This opens up new opportunities for further investigations of near-critical regions of state for metals.

(c) The use of capillary tubes helps obtain states with a high energy density (under conditions of fast pulsed heating) and, under conditions of relatively slow heating, when the vaporization of the material of the tube proper plays an important part [36–38], leads to development of technical devices for interruption and delay of high pulse currents at high voltages.

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