

Technique for Measuring Thermophysical Properties of Refractory Metals at Supercritical Temperatures¹

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A foil confined in a sapphire cell is heated by an electrical current pulse at rates from 10^{10} to 10^{11} K · s⁻¹. Temporal evolutions of the temperature and pressure distributions in the foil is investigated by means of numerical modeling. It is demonstrated that such a technique allows reaching temperatures far above 10,000 K and pressures up to 50 kbar and provides a uniform temperature and pressure in the foil. The technique has been applied for investigation of liquid tantalum. Experimental results on temperature measurement are discussed. Data on specific heat of liquid tantalum at temperatures up to 11,000 K are reported.

KEY WORDS: critical point; exploding wire technique; high-pressure cell; numerical modeling; specific heat; tantalum; temperature.

1. INTRODUCTION

Critical temperatures of refractory metals are estimated to be above 10,000 K, and critical pressures to exceed 10 kbar. To date the thermophysical properties of refractory metals are known at temperatures up to 7,000 K and at pressures up to 4 kbar [1–3]. Hence, for any refractory metal, thermophysical data are available in a narrow region of its phase diagram far from the critical point. This paper focuses on a technique providing, in principle, thermophysical data up to supercritical temperatures and pressures. A modification of exploding wire technique is used. A sample, heated by an electrical current pulse, is a thin foil confined in a sapphire

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cell. Temporal evolutions of the temperature and pressure profiles in the foil are investigated by means of numerical modeling. To perform this modeling, the one-dimensional magnetohydrodynamic model [4] has been adjusted to flat geometry. We show that to reach the supercritical region of the phase diagram of tantalum and to ensure uniform temperature and pressure in the foil, the thickness of the foil should be of the order of $10\ \mu\text{m}$ and the heating rate of 10^{10} to $10^{11}\ \text{K}\cdot\text{s}^{-1}$. On the one hand, thin foils provide low velocities of expanding metal, and therefore, uniform temperature and pressure distributions are not disturbed when the foil surface comes into collision with the wall of the cell. On the other hand, this uniform thermodynamic state in the sample is possible to maintain up to supercritical temperatures, as vaporization (boiling) is suppressed due to the high pressure generated in the cell.

In Section 2, we discuss the physical background of the technique. In Section 3, we describe a theoretical model of the heating process and present numerical modeling results demonstrating the evolution of the thermodynamic state of the foil during such a process. In Section 4, we present experimental results on liquid tantalum and their interpretation.

2. PHYSICAL BACKGROUND OF THE TECHNIQUE

In isobaric expansion experiments, a wire is heated in a gas-filled pressure vessel at 2–4 kbar [1–3] or in a water-filled vessel at 5 kbar [6]. When a pressure-vessel limitation of approximately 10 kbar is imposed [1], the critical points of refractory metals cannot be achieved by means of this technique. A solution of this problem could be a technique using the dynamic pressure generated in a sample under conditions of rapid resistive heating. This pressure results from several effects: the inertia effect in surrounding medium, the inertia effect in the sample, and the pinch effect [5]. Both the inertia effect in the wire and the pinch effect give nonuniform pressure distributions. Only the inertia effect in the surrounding medium can provide uniform pressure in the sample.

The pressure P_2 at the surface of a cylindrical piston moving with a constant velocity U in a medium is given by [7]

$$P_2 = \rho_2 U^2 \left[\ln \left(\frac{2c_2}{U} \right) - \frac{1}{2} \right] \quad (1)$$

where ρ_2 and c_2 are the density and the sound velocity in the medium, respectively. For Ar gas at $P=3$ kbar and $T=300$ K, the pressure P_2 reaches 10 kbar when the velocity U exceeds $1\ \text{km}\cdot\text{s}^{-1}$. For Ar at $P=3$ kbar and $T=300$ K, we used the values of $\rho_2=1.3\ \text{g}\cdot\text{cm}^{-3}$ and $c_2=1.6$

$\text{km} \cdot \text{s}^{-1}$ obtained from the Van der Waals equation of state with the critical point parameters from Ref. 8. When we take quartz ($\rho_2 = 2.2 \text{ g} \cdot \text{cm}^{-3}$ and $c_2 = 5.9 \text{ km} \cdot \text{s}^{-1}$) as the surrounding medium, a pressure $P_2 = 14 \text{ kbar}$ is achieved for $U = 0.5 \text{ km} \cdot \text{s}^{-1}$. Since sound velocities of liquid metals are of the order of $1 \text{ km} \cdot \text{s}^{-1}$, the inertia effect in the wire will play an important role at such velocities of expansion. This inertia pressure in the wire is of the order of $\rho_1 U^2$, where ρ_1 is the liquid metal density. For liquid tantalum ($\rho_1 \approx 10 \text{ g} \cdot \text{cm}^{-3}$), the pressure is of the order of 10 kbar . In addition, at these velocities the electrical field strength becomes nonuniform due to the term $\mathbf{E}' = [\mathbf{u} \times \mathbf{B}]$, where \mathbf{u} is the velocity of metal and \mathbf{B} is the magnetic field strength.

For a flat geometry the pressure at the interface between the foil and the surrounding medium in the acoustic limit ($U \ll c_2$) is

$$P_2 = \rho_2 c_2 U \quad (2)$$

For a tantalum foil of $20\text{-}\mu\text{m}$ thickness and for a heating rate of $10^{11} \text{ K} \cdot \text{s}^{-1}$, we obtain $U = 5 \times 10^3 \text{ cm} \cdot \text{s}^{-1}$ (at $T = 6000 \text{ K}$ [2, 3]). In sapphire, $\rho_2 = 3.97 \text{ g} \cdot \text{cm}^{-3}$ and $c_2 = 11 \text{ km} \cdot \text{s}^{-1}$ [8], and therefore $P_2 \approx 22 \text{ kbar}$. Hence, even for a small velocity of the expanding foil, it is possible to reach supercritical pressures. The inertia effect in the foil gives a value of pressure of about 0.25 kbar . Thus, the main contribution to the pressure is caused by the inertia effect in the cell, which provides uniform pressure in the sample. From a general point of view, it is reasonable to choose a sample with a small thickness. In the case of a wire, this leads to a very small area of the visible surface of the wire [4]. In contrast to wires, foils are more convenient for the optical measurements because it is possible to get a large diameter of the spot on the metal surface from which thermal (or reflected) radiation is captured.

3. THEORETICAL MODEL

We consider pulse resistive heating of the foil confined in the sapphire cell (Fig. 1) as a result of the discharge of a capacitor bank through the foil. The dimensions of the foil and the arrangement of the cell are chosen to ensure one-dimensional expansion of this system. In particular, the thickness of the foil should be much smaller than its width and length. In this case, movement of substance and distributions of electric and magnetic fields can be described by a one-dimensional magnetohydrodynamic model. In Cartesian coordinate system, with the x axis perpendicular to the plane of the foil, the velocity of substance has only a component along the x axis, the electrical current density has only a z component, and the magnetic

field strength has only a y component. The system of equations describing the movement of substance consists of the local laws of conservation of mass, momentum, and energy. The distributions of electric and magnetic fields are given by Maxwell's equations. The total current in the circuit is the solution of the equation for the entire RCL circuit containing the capacitor bank, the sample, and a series ballast resistor.

In these calculations we used an equation of state [9]. The fitting parameters in this equation were adjusted to data for the thermodynamic properties of liquid tantalum [1–3]. The procedure of fitting is discussed in Ref. 4. As a result, this equation of state gives an equilibrium line and a critical point. For the lack of a first principle model, the electrical conductivity of tantalum as a function of the temperature and density was obtained on the basis of theoretical models and approximations of experimental data. In the solid and liquid states an approximation of experimental data [1–3] was used. In the region of classical weakly non-ideal plasma, collisions of electrons with ions and atoms were taken into account to calculate the conductivity. In the supercritical region, we constructed an interpolation formula, which gives as asymptotes the dependences for the liquid and ideal plasma. The dependence for the liquid phase was extrapolated to a value of the density ρ^* (a threshold for the metal–nonmetal transition). The value of this threshold was estimated to be 10–30 times less than the normal solid density. For $\rho < \rho^*$ a spline approximation was used to connect the dependence with those for plasma.

Numerical modeling results for a shot with a heating rate of $6 \times 10^9 \text{ K} \cdot \text{s}^{-1}$ are presented in Fig. 2. The trajectories of five fixed Lagrangian particles (thin layers containing a fixed mass) in the (P, V) and (T, V) planes are shown. Here $V = \rho^{-1}$, and V_0 is the specific volume at normal conditions. These five curves correspond to the plane of symmetry of the system, to 20, 40, 60, and 90% of the total mass of the foil. In these calculations the vaporization of tantalum from the surface of the sample is neglected. Initially the space between the sample and the wall of the cell is filled with air under normal conditions. The ratio of the volume of the cell to the initial volume of the sample was 1.26. From the beginning of the process to point a the pressure increase is due to sample heating only. At point b the pressure is due to pressurized air in the space between the sample and the wall. Point c designates the moment when the cell is filled and point d corresponds to a weak rise in pressure due to the rise in the Joule heating. As one can see, the practically uniform distributions of the pressure and the temperature in the sample are not disturbed after the cell is filled, and it is possible to maintain this uniform thermodynamic state in the supercritical region of the phase diagram. The temporal measured and calculated dependences for the shot are presented in Fig. 3.

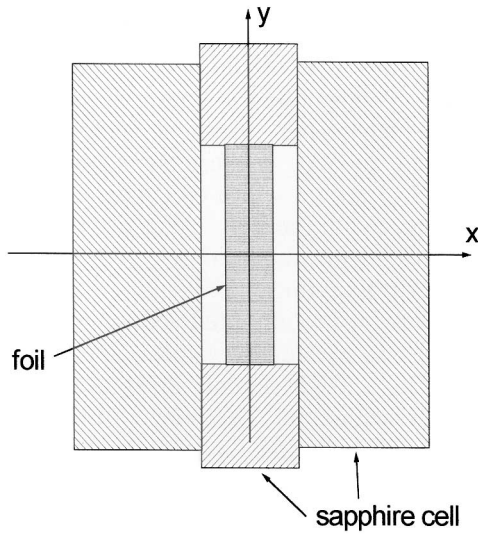


Fig. 1. Schematic diagram of composition of the foil in sapphire cell (in the cross section perpendicular to the current direction).

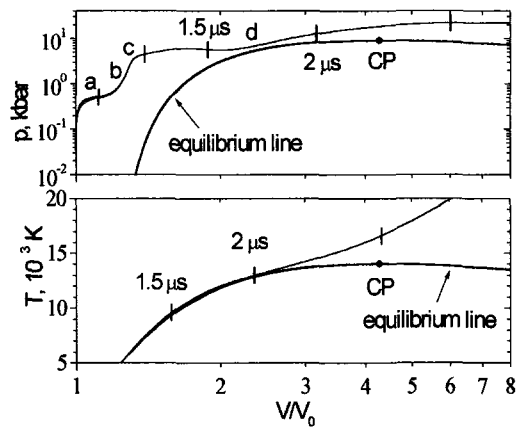


Fig. 2. Temporal evolution of thermodynamic state of five Lagrangian particles for a shot with a heating rate of $8 \times 10^9 \text{ K} \cdot \text{s}^{-1}$ in the (P, V) and (T, V) planes. Times from the beginning in $0.5\text{-}\mu\text{s}$ increments are indicated by the vertical bars.

4. EXPERIMENTAL RESULTS AND THEIR INTERPRETATION

Our experiments were carried out with Ta foils having a width of 1 mm, a thickness of 12–36 μm , and a length of 10 mm. The sapphire cell consisted of two thick plates 5 mm thickness, 10 mm in length, and 10 mm in width. Two thin plates placed between these plates form a rectangular cover with a length of 10 mm, a width of 1 mm, and a thickness of 20 to 100 μm . The foil was placed in this cover (Fig. 1). The ends of the foil were connected with massive clamps made of WC + Co alloy. The heating was accomplished by discharging through the foil a capacitor bank with a capacitance of 72 μF , charged to an initial voltage of 10 to 18 kV. The voltage across the sample was measured by means of a resistive divider, and the current through it by a Rogovski coil. Both measurements were carried out with a TDS640A oscilloscope. The temperature of the foil was determined from the thermal radiation emitted in the wavelength range of 900 ± 8 nm. The radiation from a spot with a diameter of 0.5 mm on the foil surface was captured by means of optical system, transferred through an interference filter, and then displayed on the entrance of a photodiode. In the photodiode signal, the melting plateau was identified and used for calibration. The temperature was calculated using Planck's law assuming the emissivity of liquid tantalum is constant.

The surface temperatures as functions of the specific imparted energy for different shots are given in Fig. 4. Shown for comparison are also the

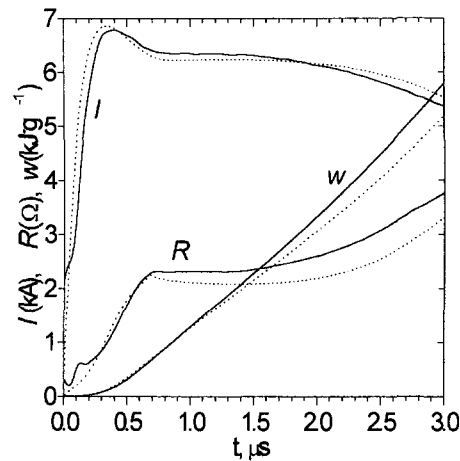


Fig. 3. Temporal dependences of the electrical current through the sample (I), its resistance (R), and the total energy (w) measured (solid lines) and calculated (dotted lines) for a shot with a heating rate of $8 \times 10^9 \text{ K} \cdot \text{s}^{-1}$.

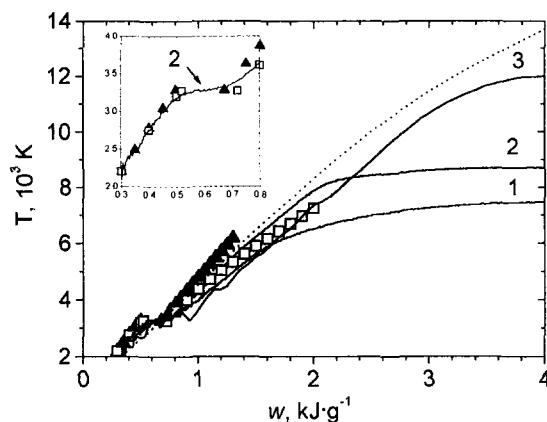


Fig. 4. Measured temperatures (solid lines) and calculated temperature (dotted lines) as a function of the specific energy. (1) Shot with a heating rate of $8 \times 10^9 \text{ K} \cdot \text{s}^{-1}$. (2) Shot with a heating rate of $8 \times 10^9 \text{ K} \cdot \text{s}^{-1}$ for a cell with a thin quartz plate placed on its inner wall (the calculated curve corresponds to this shot). (3) Shot with a heating rate of $6 \times 10^{10} \text{ K} \cdot \text{s}^{-1}$. Symbols indicate literature data: squares [2]; triangles [3].

results of Gathers [2] and Berthault et al. [3]. As one can see, our measurements on the submicrosecond time scale (the duration of the melting plateau was less than $0.1 \mu\text{s}$) give an explicit melting plateau and a reasonable value of the specific heat of liquid tantalum. The temperature measurements for shot 1 were limited to a temperature around 6000 K. Analysis shows that this limitation to 6000 K can be caused by heat conduction into the sapphire after the cell is filled (for this shot the moment of filling of the cell corresponds to the specific energy of $1.2 \text{ kJ} \cdot \text{g}^{-1}$). After this condition, the surface of the sample had good thermal contact with the sapphire wall for about $0.5 \mu\text{s}$ to the moment when the temperature measurement was terminated, i.e., when the temperature reached the value of 6000 K.

To interpret this result, the temperature profiles in the sample and in sapphire were calculated. The corresponding system of equations consists of the equation of heat conduction with the Joule heating term (in the region of the sample)

$$\frac{\partial T}{\partial t} = \chi_1 \frac{\partial^2 T}{\partial x^2} + \frac{\sigma_1 E^2}{\rho_1 C_{p1}} \quad (3)$$

and the heat conduction equation (in sapphire)

$$\frac{\partial T}{\partial t} = \chi_2 \frac{\partial^2 T}{\partial x^2} \quad (4)$$

where E is the electrical field strength, σ is the electrical conductivity, C_{p1} is the specific heat at constant pressure, and χ is the thermal diffusivity; subscript "1" refers to the sample and subscript "2" to the cell. Boundary conditions are set at the plane of symmetry of the system, at the outer surface of the wall of the cell (no heat fluxes), and at the interface (the continuity conditions for both the heat flux and the temperature). The numerical solution of these equations for shot 1 is presented in Fig. 5a. Temperature profiles in the sample ($x < a$) and in the cell ($x > a$) for different times are shown. The initial condition for this case was $T(x, 0) = 4000$ K; the time $t = 0$ corresponds to the moment of filling the cell.

It seems reasonable to expect that the effect of lowering the surface temperature can be suppressed when the heating rate is sufficiently high. More careful analysis shows that this conclusion is not correct. Indeed, for a time t the thickness of a heated layer in the wall is $\delta_2 \sim \sqrt{\chi_2 t}$. The thickness of a cooled layer in the sample is $\delta_1 \sim \sqrt{\chi_1 t}$. From the energy conservation law we obtain

$$T_1 - T_s \approx \frac{C_{p2}}{C_{p1}} \sqrt{\frac{\chi_2}{\chi_1}} T_s$$

where T_1 is the temperature in the sample far from the surface and T_s is the temperature at the surface; we have neglected the initial temperature in the wall in comparison to T_s . Thus, a lowering of the surface temperature

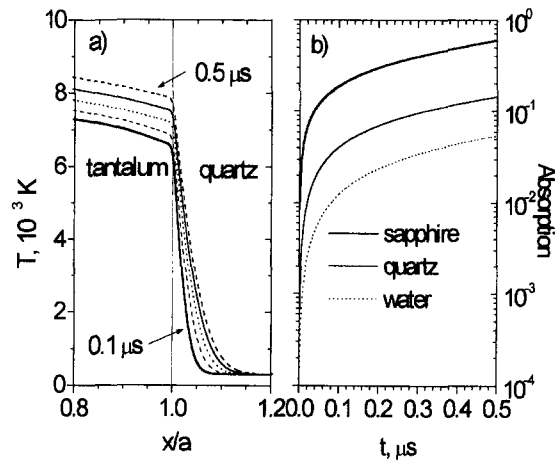


Fig. 5. (a) Temperature profiles in the sample and in the wall after filling the cell, and (b) absorption of radiation in different materials for the distance of 0.5 cm.

$T_1 - T_s$ is proportional to the surface temperature and does not depend on the heating rate. Nevertheless, the transparency of sapphire decreases with a rise in the temperature and this effect depends on the heating rate.

To estimate the effect of absorption of radiation, we calculated the amount of radiation absorbed in the wall, taking into account the temperature dependence of the absorption coefficient from Ref. 10. In Fig. 5b the ratio of the absorbed light to the incident light as a function of time is given. In these calculations, at each time we used the temperature profile in sapphire calculated according to Eqs. (3) and (4). Thus, to avoid effects of heat conduction and absorption, it is preferable to use both the cell with the low-heat conductivity walls and the higher heating rate. In Fig. 4, curve 2 corresponds to a shot with a heating rate of $8 \times 10^9 \text{ K} \cdot \text{s}^{-1}$ for a cell with a thin quartz plate placed on the inner surface of the wall. Curve 3 corresponds to a shot with a heating rate of $6 \times 10^{10} \text{ K} \cdot \text{s}^{-1}$. The maximum temperature measured for this shot was about 11,000 K. Because of the wide dynamic range of the pyrometer signal, the melting plateau is of the order of the noise. Calibration of the pyrometer signal for this shot was performed at 5000 K, where the signal-to-noise ratio is sufficiently high. As one can see, the calculated and measured temperatures are in reasonable agreement. This may confirm our assumption about the constant emissivity of liquid tantalum in the temperature range $T < 11,000 \text{ K}$.

4. CONCLUSIONS

We have demonstrated that the technique of thin foils confined in sapphire cells can be used to reach supercritical temperatures and pressures for refractory metals and ensures the uniform distributions of the temperature and the pressure in the foil under such conditions. The experimental results for the temperature measurements are in good agreement with isobaric expansion experiments at low temperatures. Data on the specific heat of liquid tantalum at temperatures up to 11,000 K have been obtained.

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