

Thermophysical Properties of Solid and Liquid Tungsten¹

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The thermophysical properties of solid and liquid tungsten have been measured up to an enthalpy of $H = 1.4 \text{ MJ} \cdot \text{kg}^{-1}$ using an isobaric expansion technique. These measurements give the pressure, temperature, volume, enthalpy, electrical resistivity, and sound velocity as fundamental quantities. From these, other properties may be calculated, such as specific heat at constant volume and pressure, heat of fusion, isothermal and adiabatic bulk moduli and compressibilities, and thermodynamic γ . Results of these calculations are presented for liquid tungsten and compared with literature values where such data exist. These data will help in understanding liquid-metal phenomenology theoretically and in the design and modeling of exploding wires, foils, and fuses.

KEY WORDS: electrical resistivity; enthalpy; heat of fusion; high temperatures; liquid metals; pulse-heating method; sound velocity; specific heat; tungsten.

1. INTRODUCTION

The thermophysical properties of liquid metals are difficult to measure but are of interest for several reasons. Such data are needed for understanding liquid metal phenomenology and for the empirical design and modeling of exploding wires, foils, and fuses, for example.

Equation-of-state property measurements of liquid metals are made difficult because of the generally high melting points of metals and because the critical points are at high pressure and temperature. The melting and critical points are low enough for only a few metals (mercury, cesium,

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rubidium, and potassium, for example) to allow the use of conventional static high-pressure techniques. Dynamic techniques such as pulse heating and shock-wave compression are typically used to study other metals.

We use a resistive pulse-heating method to reach high-temperature expanded liquid-metal states at a constant high pressure (up to 10 kbar). This technique can be used for many metals and allows data to be taken both above and below the thermodynamic critical point. The metallic wire-shaped sample (1×25 mm) is resistively heated in an inert gas atmosphere for a period of about 10^{-4} s by a square voltage pulse at high current ($\sim 15 \times 10^3$ A). Since the sample expands along an isobaric path the technique is called the isobaric expansion experiment (IEX) [1].

During the course of an experiment, enthalpy, temperature, and specific volume are measured as a function of time, and sound velocity is measured at the end state. From these quantities the specific heat at constant pressure, thermal expansion coefficient, Grüneisen's gamma, adiabatic and isothermal bulk moduli and compressibilities, specific heat at constant pressure, and specific heat at constant volume may be calculated. In this paper we discuss such measurements and calculations for solid and liquid tungsten.

2. EXPERIMENTAL DETAILS

2.1. Temperature Measurements

Temperatures are measured using a fast optical pyrometer. This pyrometer used pin photodiodes as detectors, interference filters to select wavelengths, and fast operational amplifiers in conjunction with transient digitizers to record data. The operational amplifiers are linear to better than 0.5%, and several gain levels may be used to cover the several-order-of-magnitude range of light emitted by the sample. The pyrometer presently consists of three channels with wavelengths of 500, 600, and 700 nm and a bandwidth of 80 nm. Frequency response of each channel is limited by the transient digitizers to 125 MHz.

Temperatures have been measured for tungsten at several wavelengths, and the results were found to be independent of wavelength to within the experimental precision of $\pm 5\%$.

2.2. Volume Measurements

Our method of recording volume as a function of time is similar to that described by Gathers et al. [1]. An argon-ion CW laser backlights the sample, forming a shadow, which is imaged onto a slit. An image-converter

streaking camera is focussed onto the slit plane with a streak duration of 100 μ s. The camera is protected from the self-illumination of the sample by a laser line filter at 5145 Å.

The tungsten (99.99%) samples are prepared by chemically cleaning and then polishing. Tungsten is a very granular material, and edge boundaries may still be observed after polishing. This granularity may cause the larger than normal error bars on volume measurements.

2.3. Enthalpy Measurements

During the pulse heating of our sample we simultaneously monitor the current through the sample with a precision transformer and the voltage along the sample between two probes. The voltage is measured with a current shunt circuit using a precision resistor and wide-band current transformer. An inductive contribution [$\approx L(\partial I/\partial t)$] is subtracted from the voltage signal, and the enthalpy is calculated by multiplying the current and voltage curves together and integrating. The enthalpy is estimated to be accurate to $\pm 3\%$.

2.4. Sound Velocities

Sound velocities are measured using a technique described previously [2]. Briefly, a ruby laser pulse (≈ 0.1 J, 25 ns) is focused onto one side of our cylindrical column of liquid metal. A low-amplitude stress wave is launched into the sample by the interaction of this laser pulse with the sample surface. This stress wave is unsupported and spherically diverging and, so, quickly attenuates into an acoustic wave. The arrival of this wave at the side of the sample opposite the source is seen from the disturbance it sends into the surrounding gas by a shadowgraph technique. This measurement gives the transit time across the sample, and by dividing this into the known sample diameter the average velocity through the sample is found.

This technique has been used on several materials to date [3, 4]. Many shots are required since only one data point per shot is measured. The sound velocity is estimated to be accurate to $\pm 5\%$.

3. RESULTS

The thermophysical properties of tungsten are shown in Table I. All values shown are for the 0.3 GPa isobar and are referenced to 300 K and 0.3 GPa. It is possible to extend the upper limit of these measurements by going to a higher-pressure isobar.

Table I. Thermophysical Properties of Solid and Liquid Tungsten

| H (MJ · kg ⁻¹) | T (K) | V/V_0 | ρ ($\mu\Omega \cdot m$) |
|------------------------------|---------|---------|--------------------------------|
| 0.35 | 2650 | 1.09 | 0.84 |
| 0.40 | 2900 | 1.10 | 0.95 |
| 0.45 | 3150 | 1.116 | 1.03 |
| 0.50 | 3350 | 1.125 | 1.13 |
| 0.55 | 3550 | 1.13 | 1.19 |
| 0.61 (S) | 3690 | 1.14 | 1.26 |
| 0.87 (L) | 3690 | 1.17 | 1.46 |
| 0.90 | 3790 | 1.18 | 1.46 |
| 0.95 | 3981 | 1.195 | 1.46 |
| 1.00 | 4172 | 1.215 | 1.46 |
| 1.05 | 4363 | 1.23 | 1.46 |
| 1.10 | 4554 | 1.24 | 1.46 |
| 1.15 | 4745 | 1.26 | 1.46 |
| 1.20 | 4935 | 1.28 | 1.46 |
| 1.25 | 5126 | 1.30 | 1.46 |
| 1.30 | 5317 | 1.31 | 1.46 |
| 1.35 | 5508 | 1.33 | 1.46 |
| 1.40 | 5699 | 1.36 | 1.46 |

Figure 1 shows our results for enthalpy plotted as a function of temperature for the solid and liquid phases. The best fit to our data for the solid phase ($2650 \leq T \leq 3690$ K) is

$$H = 0.59409 - 3.3025 \times 10^{-4}T + 9.0169 \times 10^{-8}T^2$$

and that for the liquid phase ($3690 \leq T \leq 5700$ K) is

$$H = -0.0932 + 2.62 \times 10^{-4}T$$

where H is in MJ · kg⁻¹ and T is in K. As may be seen from Fig. 1, the temperature varies linearly with enthalpy in the liquid phase. Our value of C_p for liquid tungsten is 262 J · kg⁻¹ · K⁻¹, in good agreement with the values reported by Berthault et al. [5] of 300 J · kg⁻¹ · K⁻¹, Seydel et al. [6] of 310 J · kg⁻¹ · K⁻¹, and Shaner et al. [7] of 282 J · kg⁻¹ · K⁻¹. Our value for the heat of fusion of tungsten is 0.26 MJ · kg⁻¹, also in good agreement with the values obtained by Berthault et al. [5] of 0.254 MJ · kg⁻¹, Seydel et al. [6] of 0.275 MJ · kg⁻¹, and Shaner et al. [7] of 0.250 MJ · kg⁻¹.

The data obtained for volumes of solid and liquid tungsten are shown

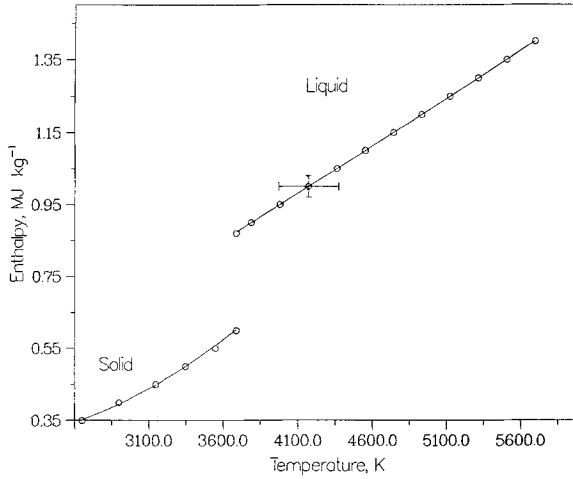


Fig. 1. Enthalpy as a function of temperature for solid and liquid tungsten. The linear relationship between enthalpy and temperature for the liquid yields a constant value of C_p .

in Fig. 2 as a function of enthalpy, and the best fit to this data is given in the solid phase by

$$V/V_0 = 0.94782 + 0.5249H - 0.3457H^2$$

with $0.35 \leq H \leq 0.61$ MJ/kg. The volume ratio is given by

$$V/V_0 = 0.86839 + 0.34388H$$

in the liquid phase with $0.87 \leq H \leq 1.40$ MJ · kg⁻¹.

Volume measurements for the shadowgraph technique we use are typically quoted to be $\pm 2-3\%$. For the tungsten measurements we see somewhat larger scatter than this, probably due to the crystalline nature of the samples, as well as the extremely small expansions observed. We have relaxed our error bars to $\pm 4\%$ for these measurements.

Electrical resistivities may be calculated from the electrical current, voltage, and volume data normally obtained for an experimental run. These calculations have been performed, and the results are shown in Fig. 3, plotted against enthalpy. It may be seen that the electrical resistivity of tungsten increases up to melt and then remains constant up to the maximum enthalpy obtained in these experiments. These results are in good agreement with those of Berthault et al. [5].

Our measured values for sound velocities are shown in Fig. 4 for solid and liquid tungsten plotted against density. Both the liquid and the solid

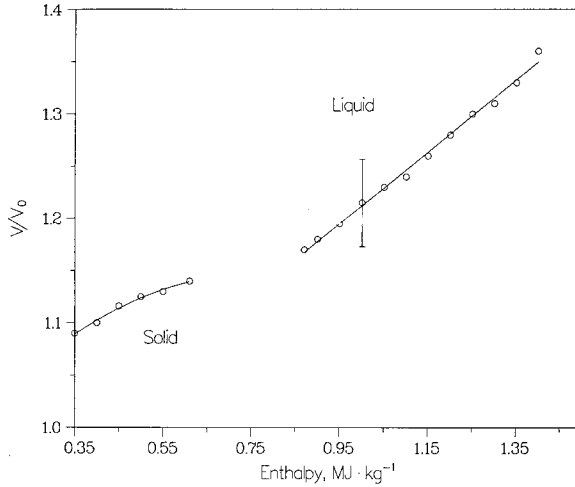


Fig. 2. Volume ratio V/V_0 for solid and liquid tungsten plotted versus enthalpy, where $V_0 = 5.181 \times 10^{-5} \text{ m}^3 \cdot \text{kg}^{-1}$.

segments of this curve may be described by a linear function of density; for the solid the best fit is given by

$$c = -6,704.85 + 0.61967\rho$$

and for the liquid

$$c = -3,695.75 + 0.42318\rho$$

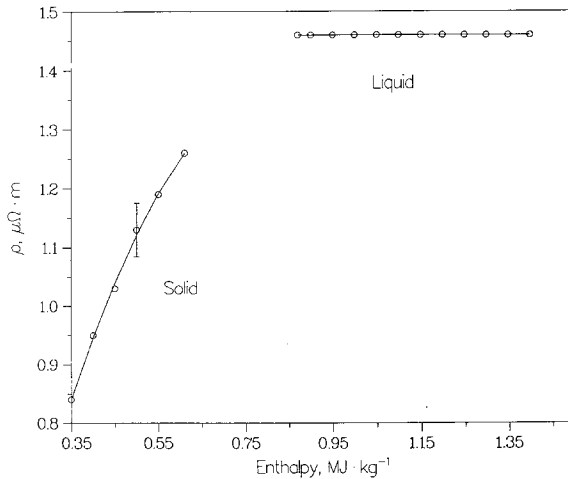


Fig. 3. Electrical resistivities for solid and liquid tungsten.

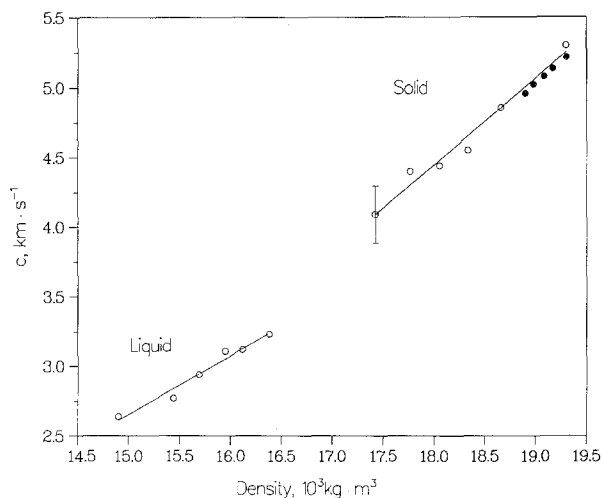


Fig. 4. Measured sound velocities for solid and liquid tungsten plotted against density. Open circles are the results of this work, and filled circles are taken from Ref. 9.

Here c is sound velocity in $\text{m} \cdot \text{s}^{-1}$, and ρ is density in $\text{kg} \cdot \text{m}^{-3}$. This linear variation of sound velocity with density has been observed by us previously for other liquid metals [2–4] and elsewhere in the literature [8]. Some data were found in the literature for sound velocities in hot solid tungsten [9] up to 1673 K for single-crystal samples, but no sound-velocity data have been found for liquid tungsten. The data points up to 1673 K have been used to estimate the polycrystalline sound speed at these temperatures and are shown in Fig. 4.

4. DISCUSSION

The data presented in the preceding section are unique in that they form a very complete set of the thermophysical properties of solid and liquid tungsten. These data are good to within the stated error limits, and we can use the various measured properties to calculate other quantities, as shown previously [3]. These calculated quantities will have uncertainties that depend upon how the uncertainties in the measured properties combine, but that can approach $\pm 20\%$ for the worst case. Such large-error bars mean that these calculations are not able to yield precise quantitative information but, rather, are useful to define trends and approximate quantitative results.

By combining the sound-velocity data with measured values of density, the adiabatic bulk modulus may be found:

$$B_S = \rho \left(\frac{\partial P}{\partial \rho} \right)_S$$

which may be rewritten as

$$B_S = \rho c^2$$

The adiabatic compressibility may be calculated from

$$K_S = 1/B_S$$

The specific heat at constant volume is calculated from

$$C_V = \frac{C_P^2}{C_P + \alpha^2 T c^2}$$

where

$$\alpha = \frac{1}{V} \left(\frac{\partial V}{\partial T} \right)_P$$

is the thermal-expansion coefficient. Once C_V is known the ratio of specific heats may be formed:

$$\gamma = C_P/C_V$$

The results of these calculations are shown in Fig. 5. Because C_P is a constant in the liquid phase, γ is just proportional to the inverse value of C_V as may be seen, and it can also be seen that to within our experimental error both C_V and γ are roughly constant in this density range.

When the values of γ are known, then the isothermal bulk modulus can be found from

$$B_T = \rho \left(\frac{\partial P}{\partial \rho} \right)_T = B_S/\gamma$$

and the isothermal compressibility from

$$K_T = 1/B_T$$

These quantities are shown plotted against density in Fig. 6.

From the above calculations it is clear that a sound-velocity measurement is of value, since when combined with other thermophysical proper-

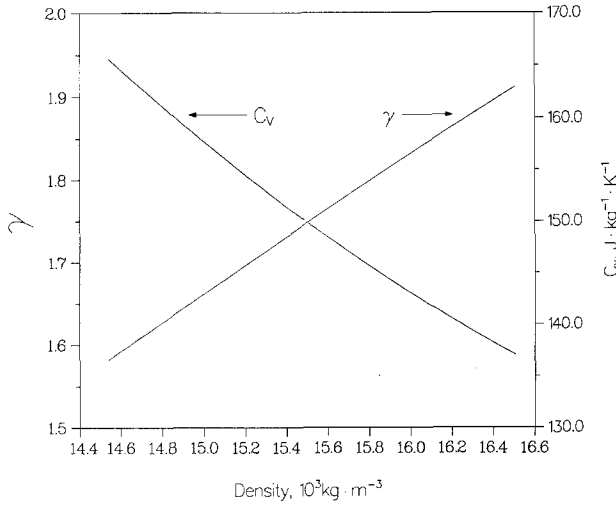


Fig. 5. Calculated values for specific heat at constant volume C_V , and $\gamma = C_P/C_V$ plotted against density.

ties many other quantities may be determined. Also, since the sound velocity is a derivative quantity, it may allow more accurate determination of other physical phenomena that are difficult to determine with more fundamental equation-of-state properties. For example, the location of the boiling line in pulse-heated liquid metals may be hard to find if some

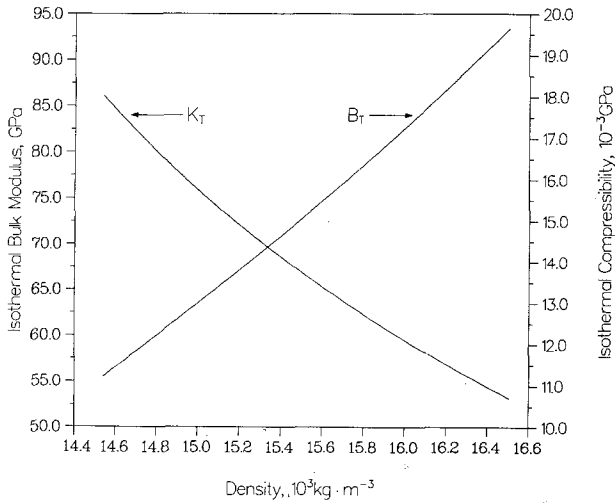


Fig. 6. Calculated values for isothermal bulk modulus (B_T) and isothermal compressibility (K_T) plotted against density.

amount of superheating is obtained, and the sound velocity is more sensitive to vapor nucleation than a simple volume measurement. The behavior of the sound velocity near a thermodynamic critical point is known for insulating fluids and so sound-velocity measurements may help to determine critical points for metals. These examples show the value of sound-velocity measurements for addressing basic physics issues, and work continues on these problems.

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REFERENCES

1. G. R. Gathers, J. W. Shaner, and R. K. Brier, *Rev. Sci. Instrum.* **47**:471 (1976).
2. R. S. Hixson, M. A. Winkler, and J. W. Shaner, *High Temp./High Press.* **17**:267 (1985).
3. R. S. Hixson, M. A. Winkler, and J. W. Shaner, *Physica* **139/140B**:893 (1986).
4. R. S. Hixson, M. A. Winkler, and J. W. Shaner, *High Temp./High Press.* **18**:635 (1986).
5. A. Berthault, L. Arles, and J. Matricon, *Int. J. Thermophys.* **7**:167 (1986).
6. V. Seydel, H. Bauhof, W. Fucke, and H. Wadle, *High Temp./High Press.* **11**:635 (1979).
7. J. W. Shaner, G. R. Gathers, and W. M. Hodgson, in *Proceedings of the Seventh Symposium on Thermophysical Properties*, A. Cezairliyan, ed. (ASME, New York, 1977), p. 896.
8. F. Birch, *Phys. Earth Planet. Inter.* **1**:141 (1968).
9. R. Lowrie and A. M. Gonas, *J.A.P.* **38**:4505 (1967).