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J. W. Shaner and G. R. Gathers

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MASTER

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THERMOPHYSICAL AND TRANSPORT PROPERTIES OF
METALS AT HIGH PRESSURE AND VERY HIGH TEMPERATURE

J. W. Shaner and G. R. Gathers

University of California, Lawrence Livermore Laboratory
Livermore, California 94550

INTRODUCTION

High pressures are conventionally applied to investigate the properties of materials at higher than normal density (i.e. density at room temperature and one atmosphere). Pressure can also be used to elevate the boiling temperature and thereby stabilize the liquid phase at densities several times less than normal. We have recently described the use of such a technique to obtain equation of state and transport data for liquid metals up to 0.4 GPa, 7000 K, and two- to fourfold expanded^[1,2]. Such information is necessary, for example, for accurate predictions of release processes following shock loading, intense laser pulse absorption, or any other rapid energy deposition.

Our technique consists of resistively heating a one mm diameter by 25 mm long metal rod, which has been placed inside a pressure vessel with optical access. The current pulse parameters, typically 20-30 kA and 10-50 μ s long, have been chosen so that samples are heated to 5000-10,000 K rapidly compared with hydrodynamic instability growth and radiative cooling rates but slowly compared with magnetic field diffusion rates. The latter constraint insures that the radial variation of energy density due to the skin effect has become negligible. The pressure vessel can be pumped up to 0.5 GPa with He or Ar to provide an isobaric environment during the expansion, to prevent chemical contamination of the hot liquid metal surface, and to increase the boiling temperature. The details of the electrical system and pressure vessel are given elsewhere^[3].

In addition to current and voltage drop measurements which provide the enthalpy data, several optical measurements are made during expansion. These optical diagnostics are shown schematically in figure 1. A CW Ar laser is used as a back lighting source to form an image of the sample on a slit plane. The slit image is then streaked to obtain a diameter vs time record. Since the sample is constrained by clamping jaws at the ends, the expansion can occur only in the radial direction. As a result, the square of the diameter is proportional to the specific volume. A Q-switched ruby laser is also used as a back lighting source to take a single snapshot at the end of the current pulse. Both optical lines of sight contain interference filters to discriminate against the intense black body radiation of the sample. An acceptable shot must show no necks or kinks indicative of magnetohydrodynamic instabilities.

The image at the slit plane can also be passed through a trifurcated fiber-optic bundle to a three color optical pyrometer for surface temperature measurements. The pyrometer, based on Si photodiodes and low-noise logarithmic preamplifiers, was designed to respond to the 2000 K to 10,000 K temperature range occurring in our most energetic shots^[3].

Since the current, voltage drop and diameter of the wire are all measured as a function of time, the electrical resistivity is also obtained during the heating of our samples. This parameter for high temperature liquids is necessary for the calculation of the dynamics of metal pieces moving in high magnetic fields, such as those found in explosive driven flux compression experiments^[4,5].

To summarize, we make simultaneous measurements of enthalpy, specific volume, temperature, and electrical resistivity. The accuracy of these measurements is $\pm 3\%$ in enthalpy, $\pm 2\%$ in specific volume, $\pm 5\%$ in temperature, and $\pm 3\%$ in resistivity.

RESULTS AND DISCUSSION

Detailed equation of state data have already been presented for Ta and Mo^[6], and for Nb and Pb^[2]. Since both the thermal expansion coefficients and the heat capacities at constant pressure are nearly constant for these liquid metals over a temperature range of several thousand degrees, the values near the melting point are quoted in the table.

For both thermal expansion and heat capacity the most difficult quantity to measure experimentally is the temperature. For the refractory transition metals, the melting point was clear as a region of constant light intensity vs enthalpy in the pyrometric records. Since the spectral emissivities of the liquid metals are generally unknown even over a limited temperature range, the first assumption made was that the emissivity was constant and equal to the value at the melting point. For Pb and U, however, the melting points are below the response threshold of the pyrometers, so another calibration was necessary. The calibration chosen involves the melting point of Ta as a standard, since the spectral emissivity of Ta near the melting point has been measured well^[7]. We then require that our data in each color channel either overlap (U case) or extrapolate into (Pb case) lower temperature data obtained by more conventional means. This technique is equivalent to arbitrarily choosing emissivities in the 2000 K region so that our data matches the data of the more conventional techniques.

Finer adjustments are then made in the unknown emissivities so that the color temperatures, based on the ratio of light intensities in two different color channels, agree with the one channel temperatures over as wide a temperature range as possible. So far, we have continued to use the temperature independent emissivity approximation.

The results of these procedures are shown for uranium at 0.2 GPa in figure 2. One channel temperatures for 700 and 450 nm are shown there to agree well

up to temperatures above 5000 K. The color temperature based on the ratio of the signals in these two channels is shown to agree well to ~ 4200 K. This behavior is similar to that reported previously for Pb^[2].

At the low temperature end of our data, the slope of T vs enthalpy (i.e. the heat capacity at constant pressure, Cp) is seen to agree well with the drop calorimetry measurements of Stephens^[8]. Our quoted value of Cp, 220 J/kgm K is somewhat higher than Stephens value of 200 J/kgm K, but the accuracy of slopes determined from our data is probably not better than $\pm 15\%$. Our value is a best fit of the data between 2000 and 3000 K.

In the intermediate temperature range, from 3000 to 4000 K, our data show a consistent trend toward a broad shallow peak in Cp of about 300 J/kgm K. More experiments are needed to positively identify this feature. One might expect such behavior arising from the narrowing and splitting of 5-f bands in the expanded uranium.

At the high temperature end (>4500 K) the color temperature and one channel temperatures deviate significantly. This behavior is to be expected as a result of a breakdown of the constant emissivity assumption. At these temperatures the equilibrium vapor pressure of U is close to one atmosphere, and the boundary layer of hot U vapor and Ar must be expected to play a role in the thermal emission properties of the wire. We are currently working on a technique, similar to that reported by Lincoln^[10], to find an optimal wavelength and temperature dependent emissivity which will give agreement among all the temperatures calculated from three channel pyrometry^[11].

NEW EXPERIMENTS

From the data we measure, it is possible to make reasonable estimates of the bulk modulus and sound velocity of expanded liquid metals. Since the isothermal bulk modulus, B_T , is related to the heat capacity by

$$B_T = \frac{C_p - C_v}{\alpha^2 V T}, \quad (1)$$

where V is the specific volume, an estimate of C_v in addition to our measurements gives B_T . The same procedure gives the bulk sound velocity, v_s , from

$$v_s = (B_T / \rho)^{1/2}. \quad (2)$$

For example, the Grover scaling law^[12] for the normal liquid part of C_v with a free electron gas added has been shown to reproduce v_s well for liquid Pb^[C]. However, for the transition metals and Uranium, the free electron gas model is unreliable. An independent measurement of B_T or v_s would be valuable.

We have previously reported on a non-contacting technique to measure acoustic velocities in very high temperature liquid metals^[13]. This technique consists of launching a stress wave across the diameter of the wire by focusing a Q-switched Nd-glass laser to a 0.3 mm spot on one side of the wire. The first surface motion of a diametrically opposed point on the wire should represent the arrival of an acoustic pulse travelling across the wire diameter at the bulk sound velocity. This surface motion is detected by making the wire the reflector of one leg of a Michelson interferometer.

This technique has now been applied to a lead wire heated to 1100 ± 100 K in a mock-up of the isobaric expansion experiment operated with one atmosphere of argon as confining gas. Figure 3 shows typical interferometer records for two shots. A portion of the Nd glass laser pulse has been mixed in the detector to provide a fiducial mark. This pulse was timed to occur 10 μ s after the main heating current was stopped in order to prevent confusion in the interferometer signal by late time expansion of the wire. A precursor of undetermined origin is seen in both records. This feature appears in most shots, but with varying shape. The main stress wave arrival is signalled by

the rapid surface motion indicated in the figure. The shape of this feature is the same for all shots. The transit time can be determined to $\pm 3\%$, with most of the uncertainty arising from which point on the laser pulse rise time one uses for the start of the stress pulse. The wire diameter at 1100 K is known to $\pm 2\%$ from our previous work^[2]. The acoustic velocity measured was 1.80 ± 0.1 km/sec, with a total scatter less than the estimated uncertainty. This value is higher than the value ~ 1.68 obtained by more conventional techniques^[14].

The reason for our slightly high velocity may be that we have a weak shock over at least part of the path. If the scaling law proposed by Kidder^[15] is used to estimate the stress generated by our 60 Gw/cm² laser pulse, the prediction is 9 GPa. Such a peak stress would be rapidly reduced by release waves after the laser pulse. However, even a 1 GPa average stress could increase the apparent acoustic velocity by 5%. Calculations are presently underway to determine quantitatively the corrections necessary for laser pulses intense enough to give a clear interferometer signal.

Although this technique will never be capable of high precision, even a $\pm 10\%$ acoustic velocity measurement will be useful, since the velocity may change by a factor of 3-4 over the experimentally accessible temperature range.

As a final illustration of the high temperature capabilities of the isobaric expansion experiment, it can be used to measure the surface tension of hot pressurized metals. Since the surface tension plays a central role in the boiling dynamics of pressurized liquid metals, this parameter is of practical importance.

We have observed that the MHD instability which usually appears first during heating is the Z-pinch. If the current is crowbarred before the instability destroys the liquid column, we are left with a sample of varying radius along

the axis. The subsequent dynamics of the wire will be determined by the surface tension. In the simplest case, a sinusoidal modulation of the surface will propagate as a train of capillary waves with a dispersion relation

$$\omega^2 = \sigma k^3 v, \quad (3)$$

where σ is the surface tension^[16]. If the σ for liquid uranium, for example, is taken to be 1.5 N/m ^[17], the phase velocity will be $\sim 700/\lambda^{1/2} \text{ mm/sec}$. If the solid wire is prepared with surface ripples to stimulate upon melting a capillary wave train of wavelength 10^{-1} mm , the phase velocity should be $\sim 2 \text{ m/sec}$, which could be an easily measurable quantity with fast streaking and framing cameras. Since we can explore specific volume ranges up to 3 or 4 fold expanded, this experiment in conjunction with our acoustic velocity measurement will represent a valuable test of the often used empirical relation $\sigma/B_T \sim \text{const}$ ^[18].

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REFERENCES

1. J. W. Shaner, G. R. Gathers, and C. Minichino, High Temp.-High Pres., **8**: 425 (1976).
2. J. W. Shaner, G. R. Gathers, and W. M. Hodgson, Proc. Seventh Symposium on Thermophysical Properties, Gaithersburg, Md., May 10-12, 1977.
3. G. R. Gathers, J. W. Shaner, and R. L. Brier, Rev. Sci. Instr., **47**: 471 (1976).

4. C. M. Fowler, W. B. Garn, P. S. Caird, J. Appl. Phys., 31: 588 (1960).
5. R. W. Hawke, D. E. Duerra, E. G. Huebel, H. Klapper, D. J. Steinberg, and R. M. Keeler, J. Appl. Phys., 43: 2734 (1972).
6. J. W. Shaner, G. R. Gathers, and C. Minichino, to be published in High Temp.-High Pres.
7. A. Cezairliyan, J. L. McDure, L. Coslovi, F. Righini, and A. Russo, High Temp.-High Pres., 5: 193 (1975).
8. H. P. Stephens, High Temp. Sci., 6: 156 (1974).
9. W. L. Thayer and I. L. Robbins, in Plutonium 1975 and Other Actinides, H. Blank and R. Lindner, eds., North Holland, Amsterdam, 1976, p. 85.
10. R. C. Lincoln and R. B. Pettit, High Temp.-High Pres., 5: 421 (1973).
11. W. M. Hodgson and J. W. Shaner, to be published.
12. R. Grover, J. Chem. Phys., 55: 3435 (1971).
13. G. R. Gathers, J. W. Shaner, C. A. Calder, and W. W. Wilcox, Proc. Seventh Symposium on Thermophysical Properties, Gaithersburg, Md., May 10-12, 1977.
14. M. B. Gitis and I. G. Mikhailov, Sov. Phys. Acoustics, 11: 372 (1966).
15. R. E. Kidder, in Physics of High Energy Density, P. Caldirola and H. Knoepfel, eds., (Academic Press, New York, 1971), p. 306.
16. A. Sommerfeld, Mechanics of Deformable Bodies, (Academic Press, New York, 1950), p. 180.
17. L. J. Wittenberg, in Plutonium 1975 and Other Actinides, H. Blank and R. Lindner, eds., North Holland, Amsterdam, 1976, p. 71.
18. R. D. Present, J. Chem. Phys., 61: 4267 (1974).

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FIGURE CAPTIONS

Fig. 1. Schematic of the optical diagnostics. The image formed at the slit may be used either for the streak camera or the optical pyrometer.

Fig. 2. Temperature vs enthalpy for Uranium at 0.2 GPa. Temperatures were calculated from the light intensity in the 700 and 450 nm channels, and from their ratio (color temp.). Low temperature data for comparison is from:

a. Ref [8]

b. Ref [9]

Fig. 3. Interferometer signal for detecting acoustic pulse arrival. Some pulsed laser power is mixed in at the beginning of the record to serve as a time origin. The time base is 100 μ s/cm.

TABLE I

Volume Expansion Coefficient, Heat Capacity, and
Electrical Resistivity of Liquid Metals

<u>Property</u>	<u>Nb</u>	<u>Mo</u>	<u>Ta</u>	<u>W</u>	<u>U</u>	<u>Pb</u>
$\alpha(\text{liquid})\text{K}^{-1}$	7×10^{-5}	9×10^{-5}	9×10^{-5}	---	1×10^{-4}	1.25×10^{-4}
$C_p \text{ (J/kg-K)}$	610	730	380	282	220	157
(R)	(6.8)	(8.5)	(8.3)	(6.2)	(5.9)	(3.9)
$\rho(\mu\Omega\text{m})$	1.09	0.99	1.32	1.32	0.69	---

Fig. 1

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Fig. 2

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Fig. 3

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