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Determination of Thermophysical Properties of Fluid Metals by Wire-Explosion Experiments[1]

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Wire explosion experiments were used to obtain thermophysical properties of tungsten. These properties are specific heat, thermal expansion coefficient, compressibility, and electrical and thermal conductivities and their dependences on temperature. There are strong indications that the emissivity in the liquid range should be dependent on temperature. A proposal has been made for determining the emissivity from comparison of experiments with model calculations. The temperature range is extended up to 10,000 K, and first indications for the near-critical-point behavior were obtained. Other properties such as diffusivity, viscosity, and surface tension may possibly also be derived in this range.

KEY WORDS: compressibility; electrical conductivity; emissivity; specific heat; thermal conductivity; thermal expansion coefficient; tungsten.

1. INTRODUCTION

Thermodynamic, optical, and transport properties at higher temperatures are usually designated together as thermophysical properties. At higher temperatures, the corresponding state of matter is often the liquid. The highest temperature in the liquid range is the critical temperature which is of the order of 10,000 K for higher melting metals, whereas the critical pressure is about 10 kbar. To obtain such state parameters, steady-state experiments are unsuitable; dynamic experiments have to be carried out with the help of lasers or fast electrical discharges.

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Intense research on wire explosions started in the 1960s [1]. In the 1970s and continuing up to now, such research has been carried out with a goal of studying thermophysical properties of liquid metals at higher temperatures and pressures [2, 3].

The results obtained so far include thermodynamic properties (enthalpy at the onset and at the end of melting (enthalpy of melting), specific heat at constant pressure, thermal expansion coefficient, sound speed, compressibility, and, in some cases, critical data for the liquid–gas transition). Further, there are results for transport properties (electrical and thermal conductivity, diffusivity, and viscosity). For the optical properties, the spectral emissivity is the most important because it describes the deviation of the emitted radiation from blackbody radiation and is, therefore, crucial for the temperature determination. There are only few measurements of the emissivity for higher melting metals at temperatures slightly higher than the melting temperature. Thus, the emissivity of the liquid at melting is often used to describe the complete liquid range.

The current study involves thermophysical measurements on tungsten, the highest melting point metal. Compared with earlier results on this material [4–7], which are given at temperatures up to 7500 K, the experiments reported here extend to at least 10,000 K.

2. MEASUREMENTS

The tungsten samples used for measurements were wires (Goodfellow) 5 mm long with a diameter of 200 μm with a polytetrafluoroethylene (PTFE) coating of 50 μm. The wires were heated in a fast discharge circuit ($C \approx 0.345\,\mu$F, $L \approx 50$ nH). Melting is finished after 100 to 200 ns; due to the fast heating, the pressure reached some 10 kbar although the environment was at atmospheric pressure (air). The current through the probe and the voltage across it were measured, and the voltage was carefully corrected for the inductive part to obtain only the ohmic voltage. From the current and voltage, the power and the energy dissipated in the material as well as its resistance can be derived. The expansion of the wire under the influence of heating was measured with a streak camera (see Ref. 8), whereas the maintenance of the cylindrical shape and the homogeneity of the wire material over its entire length were monitored by a framing camera.

The temperature was derived from the measured radiation intensity at a wavelength of 650 nm. Usually, the radiation is calibrated at the melting plateau which is assumed to correspond to the melting temperature at atmospheric pressure (that means that a pressure dependence of the melting temperature has been neglected). There are, however, strong indications that the melting temperature (and, consequently, also the pressure) for coated wires increases with the energy-input rate (see Ref. 9) what has to be taken into account.

At temperatures above 8000 to 10,000 K (dependent on the energy input rate), however, the calculated temperature plotted versus the energy shows a sudden increase which would correspond to an unphysical decrease in the specific heat. As has already been assumed in Ref. 10, this temperature increase could be explained by an increase in the emissivity due to the generation of small bubbles (boiling) in a thin layer near the surface. Each of these bubbles acts as a cavity. A quite reasonable behavior of the specific heat can be achieved when in this range (above 8000 K), the emissivity is increased by about 50% compared with that in the liquid at lower temperatures [11, 12]. A more detailed description of the experiments can be found in Refs. 8 and 9.

3. DATA ANALYSIS

For the data analysis, a homogeneous thermodynamic state throughout the whole wire was assumed. A theoretical justification for this assumption up to the onset of strong vaporization at the surface and the formation of two-phase (liquid–gas) peripheral layers was given in Refs. 10 and 13. There, a magnetohydrodynamic description combined with the so-called soft-sphere model in a modified form [14] were successfully applied to describe the thermodynamics of liquid tungsten. According to these calculations carried out now for a coated tungsten wire, for temperatures above 8000 K, the pressure at the sample axis is about 10 kbar and it falls to several kilobars at the interface between the PTFE shell and the sample material.

Thus, the PTFE coating keeps the pressure high at the edge and the radial pressure profile more flat, and it prevents tungsten vaporization. The coating is assumed to be transparent for the observation wavelength. The remaining gradient of the pressure should not have a serious influence on the determination of the thermodynamic properties because the pressure dependence of these quantities is very weak and becomes essential only in the neighborhood of the critical point. Only at higher temperatures ($T > 10{,}000$ K) does the pressure at the edge become lower than the vapor pressure of tungsten and boiling starts. A two-phase layer develops [13], which keeps the pressure in the outer region high and the profile flat due to its low velocity of sound and which should also be responsible for the assumed increase in the emissivity [11].

In the present paper, the soft-sphere model has been used for the discussion of the reliability and the qualitative behavior of the thermodynamic functions extracted from the measured quantities. The soft-sphere model is based on Monte Carlo calculations for particles interacting with pair potentials of the form $\Phi(r)=\varepsilon(\sigma/r)^n$. The resulting configurational energy is multiplied by a factor $Q>1$; this takes into account the large electronic contribution in the thermodynamic functions for some liquid transition metals [14]. The density dependence of the van der Waals-like attractive energy has the form ρ^{m+1} ($m=1$ for a van der Waals gas). With the chosen fit parameters, $n=8$, $m=1.06$, $Q=1.4$, $\varepsilon=53.4$ eV, and $\sigma=2.03$ Å, the liquid density at melting ($T_m=3680$ K, $p=1$ bar) becomes $\rho_m=16.5\ \mathrm{g\cdot cm^{-3}}$ (as in the experiment) and the critical point for the liquid–gas phase transition is characterized by $T_c=12{,}500$ K and $\rho_c=4.52\ \mathrm{g\cdot cm^{-3}}$. The latter values correspond to Likalter's similarity relations for the critical point parameters [15–17]. The critical pressure from the model, however, is $p_c\approx 13$ kbar in comparison with 16 kbar from Likalter. For the enthalpy of vaporization ΔH_V as well as for the sound velocity c at the boiling point, reasonable values are given by the model ($\Delta H_V\approx 880\ \mathrm{kJ\cdot mol^{-1}}$ and $c\approx 3400\ \mathrm{m\cdot s^{-1}}$). Considering the uncertainty of the corresponding experimental values as well as that of the critical parameters, the agreement seems to be relatively good.

The thermal expansion coefficient and the specific heat at constant pressure are calculated using the temperature derived assuming a constant emissivity; otherwise (using the corrected temperature according to the proposed emissivity), just the soft-sphere model values would result. Further, the pressure is assumed to be constant. The isothermal compressibility has been estimated with a constant specific heat $c_v=3R_g$ (R_g the universal gas constant) according to the well-known thermodynamic relation.

4. RESULTS

In Fig. 1, the density versus temperature is plotted from the measurements. It is compared with measurements of other authors, who have, however, all used a constant value of emissivity in the liquid. In addition to the experimental values, results from the soft-sphere model are given. The two-phase region (liquid–gas) can be seen, and furthermore, some isobars are shown. As can be seen, the whole liquid range up to some 10 kbar is located in a very small range along the boundary of the coexistence region. Therefore, in this range due to the small value of the compressibility, small changes in the density lead to very large changes in the pressure. This means that the pressure in the liquid cannot be determined accurately by only rough density-temperature measurements.

Obviously, all the experimental values give a density too low at a certain temperature compared with the soft-sphere model and other relevant

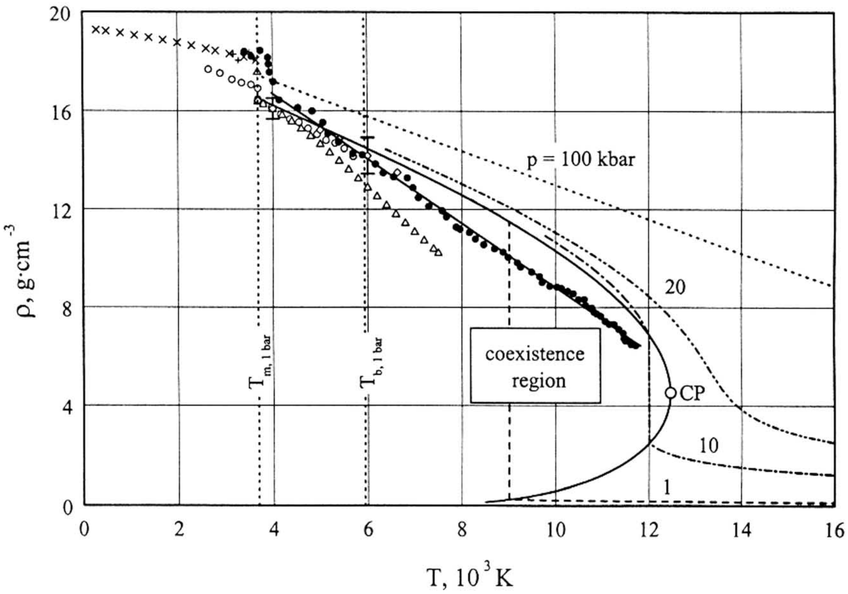

Fig. 1. Mass density versus temperature for tungsten. Experimental values: × [24]; + [25]; △ [4]; ◇ [26] with error bars; ○ [6]; ●, this work. The boundary of the liquid–gas coexistence region is given by a solid line. The labeled lines are isobars from the soft-sphere model (CP is the critical point [15, 16]), T_m, melting; T_b, boiling temperature.

results for the liquid boundary of the coexistence region [18] when it is assumed that the expansion is completely in the liquid. There can be more than one reason for this deviation, but the most convincing argument seems to be that the emissivity, unknown over most of the liquid range, is responsible. A lower value of the emissivity in the liquid will lead to higher temperatures at a certain density and will push the expansion curve out of the coexistence region. An upper limit of the emissivity results for a density-temperature curve coincident with the liquid boundary of the two-phase region. The resulting emissivity is shown in Fig. 2, which can be interpreted as an emissivity in liquid tungsten determined by a combination of measurements (density, radiation) and a model calculation (soft-sphere model). The behavior of the emissivity at high temperature is quite similar to those assumed in Section 2 from specific-heat considerations.

The data obtained so far have been used to derive some thermodynamic data. As already mentioned, the temperature used here is calculated under the assumption of a constant emissivity. In Fig. 3, the thermal expansion coefficient α_p is compared with results from other experiments and from model calculations using the soft-sphere model as a

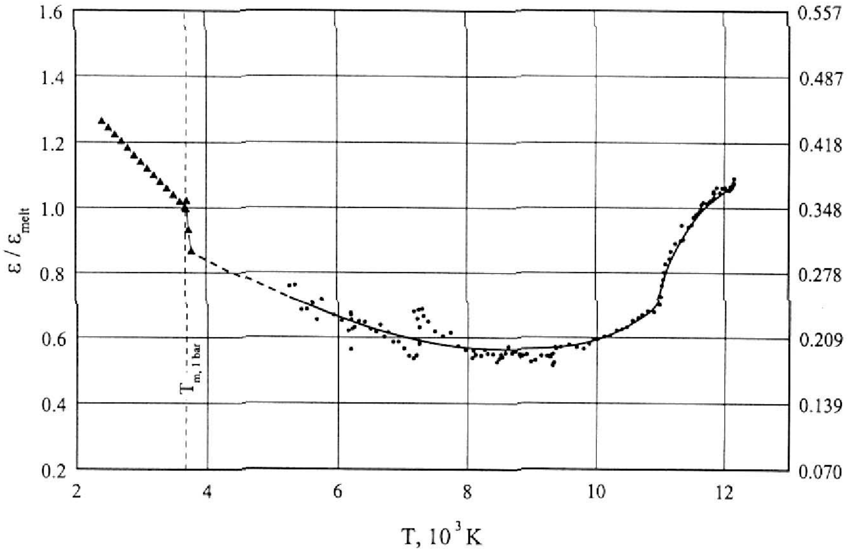

Fig. 2. Emissivity of tungsten versus temperature. ▲ [10]; ●, this work (procedure see text).

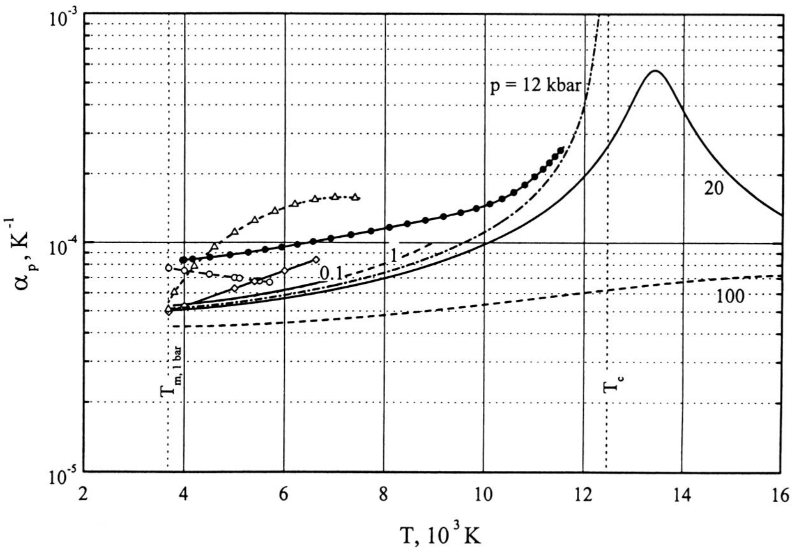

Fig. 3. Thermal expansion coefficient of tungsten versus temperature. Experimental values: △ [4]; ◇ [26]; ○ [6]; ●, this work. The labeled lines are isobars from the soft-sphere model. T_c, critical temperature.

guide. In the low-temperature range where the pressure dependence should be weak, the results differ by a factor of two and even the slope is different; this is also a hint for correcting the temperature as to the emissivities. In the high-temperature range, only the present values are available. They are higher than the model values (as can be seen in Fig. 1), but there is a qualitative agreement. At the critical point, the theoretical thermal expansion coefficient α_p shows—similar to other properties (compressibility χ_T, specific heat c_p)—a divergence [19].

In Fig. 4 the isothermal compressibility κ_T as derived here is compared with experimental results and with model calculations. Our results can only be a rough estimate because of the assumed constant specific heat c_v. There is, however, a relationship between the compressibility and the surface tension [27], which remarkably shows the best agreement with the model calculations.

In Fig. 5 the specific heat at constant pressure c_p is plotted versus temperature. The experimental values are compared with those of other authors and, again, with the results of the soft-sphere model. In the low-temperature range, there is good agreement with measured values, especially from Arpaci and Frohberg [20], which are thought to be the most reliable due to the method used. The deviation from the other results is less

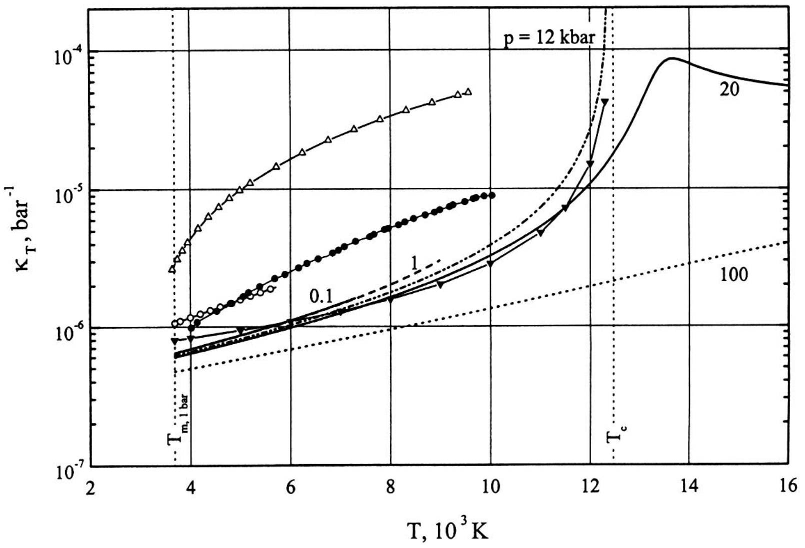

Fig. 4. Compressibility versus temperature. Experimental values: △ [4]; ○ [6]; ▼ from surface tension [27]; ●, this work, with $c_v = 3R_g$. The labeled lines again are isobars from the soft-sphere model.

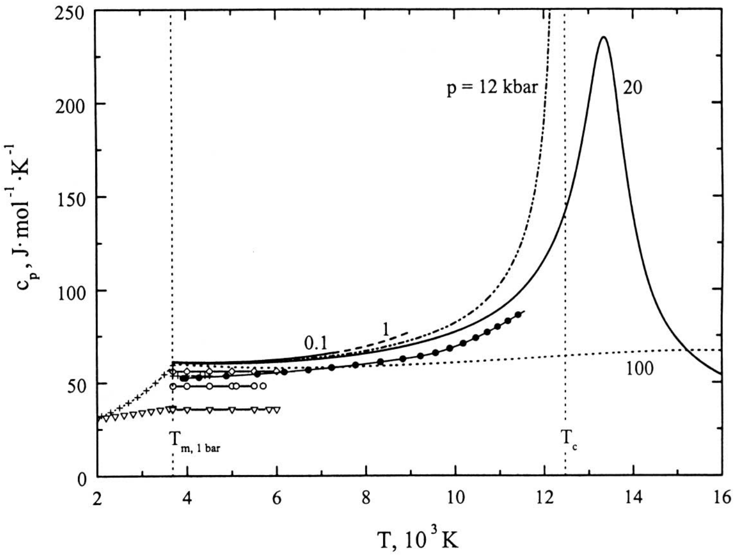

Fig. 5. Specific heat of tungsten versus temperature. ▽ [28]; ○ [6]; ◇ [7]; + [20]; ●, this work. The labeled lines are isobars from the soft-sphere model.

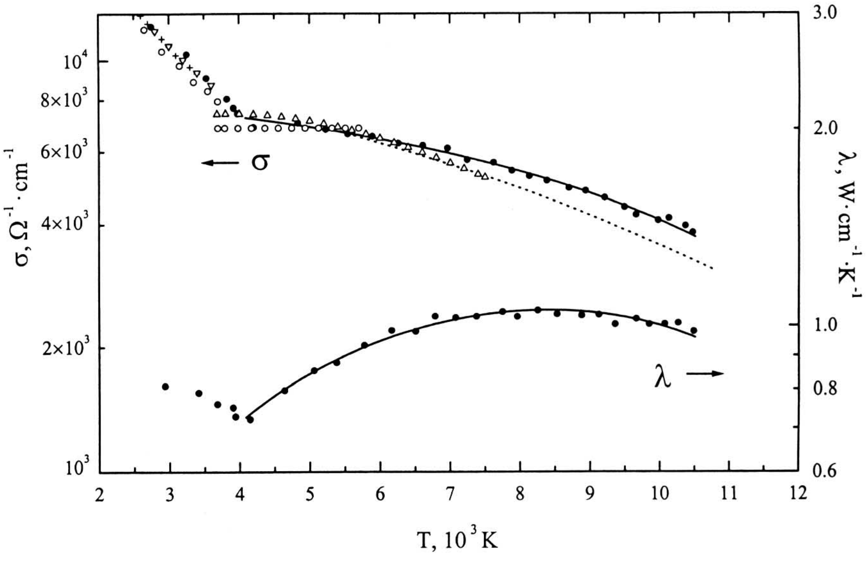

Fig. 6. Electrical and thermal conductivity of tungsten versus temperature. ○ [6]; + [23]; △ [7]; ▽ [24]; ●, this work; --- [21].

than 10%, and most important, the expected independence of specific heat with temperature is confirmed up to 5700 K. Above this temperature an increase is observed, that is expected due to the divergence of c_p at the critical point [19]. In this higher temperature range, a strong pressure dependence is also expected due to the neighborhood of the critical point.

In Fig. 6 the electrical and thermal conductivities are plotted as functions of temperature. According to Fig. 1, an increase in temperature is always connected with a decrease in density in the supercritical range. The electrical conductivity σ compares well with other measurements. After melting, there is a change in the conduction mechanism; the liquid part can be approximately described by the Ziman formula [21]. The general behavior of the electrical conductivity when a substance enters the two-phase region (liquid–gas) or expanding to supercritical pressures can be seen from alkali measurements [22].

Information about the thermal conductivity of liquid metals is very sparse [9]. Using the Wiedemann–Franz equation, the thermal conductivity λ can be calculated: $\lambda = LT\sigma$ with the constant $L \approx 2.45 \times 10^{-8}\ \mathrm{W} \cdot \Omega \cdot \mathrm{K}^{-2}$ [23]. The question is whether the Wiedemann–Franz relation is also valid near the critical point. From the thermal conductivity, the self-diffusivity D and the dynamic viscosity η may be determined [23].

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