Electrical resistivity of high melting metals up into the liquid phase (V, Nb, Ta, Mo, W)

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Abstract. Objective of the collaboration of TU Graz and DLR Cologne is the measurement of specific electrical resistivity comparing pulse-heating and levitation results. The TU Graz measurements on V, Nb, Ta, Mo and W will be presented and discussed with regard to volume expansion and resistivity.

1. The ASAP Project
The Subsecond Thermophysics Group in Graz has a long tradition in performing pulse-heating experiments (see TU Graz Experimental Setup). These deliver thermophysical properties of metals up into the liquid phase as a function of temperature [1]. We started to team up with the Institute of Materials Physics in Space at DLR (German Aerospace Center) in Cologne, which also performs experiments on liquid metals by means of electromagnetic levitation [2]. On one hand both setups, at TU Graz and DLR Cologne, have the same field of research, but on the other hand our investigations are very different: Pulse-heating experiments have a time-scale of microseconds (typically 50 µs for the transit from room temperature to 4000 K) whereas levitation can be seen as a quasistatic method.

This cooperation covers the valuable possibility to create a compilation of electrical resistivity data with the excellence of being verified in two completely different approaches, pulse-heating and levitation. The latter is further planned to be carried out under micro gravity (µg) conditions at the ISS. The prearrangements to this more precise experiment in space e. g. material selection, also have to be done in terms of comparability. The ensure TU Graz’s ability to take part in these spaceflight networks the Austrian Space Applications Program (ASAP) delivers financial support (see last paragraph).

2. TU Graz Experimental Setup
A wire shaped specimen is assembled into a discharge circuit. A capacitor bank of 500 µF is charged with a high voltage power supply and then discharged through the wire. Its ohmic resistivity leads to a rapid increase in temperature. Heating rates of $10^8$ K/s eliminate chemical reactions between the wire and the surrounding atmosphere and the cylindrical sample maintains its shape until the end of the liquid phase. This leads to an experimental duration of about 50 µs.

The current is measured with an induction coil. The voltage drop is measured with two knife edges placed on the wire. Temperature has to be detected pyrometrically using Planck’s law of black-body radiation. These records deliver the electrical resistivity with initial geometry $\rho_G$ as a function of temperature. As thermal expansion leads to smaller densities, which affects the calculation of the
specific electrical resistivity $\rho$, the volume expansion has to be taken into account. At TU Graz the change of the diameter $D$ can be measured with a fast CCD camera as a shadowgraph when the wire is backlit with a photoflash.

3. Results

For each material, the results are given as a function of temperature $T$ in polynomial fits, split into solid and liquid phase (see table 1). Melting temperatures and references: V: 2201 K, [3], Nb: 2745 K, [4, 5], Ta: 3270 K, [6, 7], Mo: 2895 K, [8, 9], W: 3687 K, [4, 10]. Plots see figure 1 and figure 2.

Table 1: Polynomial least-squares fits ($\text{Value} = a + bT + cT^2 + dT^3$). $D$: diameter, $D_0$: diameter at room temperature, $\rho_{IG}$: specific electrical resistivity with initial geometry, $\rho$: specific electrical resistivity, $\rho_{IG}$ and $\rho$ given in $\mu \Omega \cdot m$, decimal places according to references.

| Material | Range / K   | Value $a$ | $b \times 10^5$ | $c \times 10^6$ | $d \times 10^9$ |
|----------|-------------|-----------|-----------------|-----------------|-----------------|
| Vanadium: | 1800 - 2201 | 0.990     | 3.137           | 1.261           |
|          | 2201 - 2900 | 0.910     | 7.727           |                 |
|          | $\rho_{IG}$ | 1800 - 2201 | -0.22754 | 1.120 x 10^{-3} | -2.15791 x 10^{-7} |
|          | $\rho_{IG}$ | 2201 - 2900 | 1.486     | -3.305 x 10^{-4} | 5.188 x 10^{-8} |
|          | $\rho$      | 1800 - 2201 | -0.280     | 1.180 x 10^{-3} | -2.174 x 10^{-7} |
| Niobium: | 2745 - 3700 | 0.963     | -2.5 x 10^{-5} | 3.14 x 10^{-8}  | -1.17 x 10^{-12} |
|          | 473 - 1573  | 0.023     | 4.839 x 10^{-4} | -8.899 x 10^{-8} |
|          | 1790 - 2745 | 0.199     | 2.441 x 10^{-4} |                 |
|          | 2745 - 3700 | 0.972     | 5.527 x 10^{-6} |                 |
| Tantalum: | 2750 - 3250 | -1.3393   | 2.4883 x 10^{-3} | -0.86945 x 10^{-6} | 0.1026 x 10^{-9} |
|          | 3300 - 5000 | 0.95634   | 5.6199 x 10^{-5} | -2.6656 x 10^{-9} | 3.7798 x 10^{-13} |
|          | $\rho_{IG}$ | 2750 - 3250 | 0.2307     | 2.7007 x 10^{-4} |                 |
|          | $\rho_{IG}$ | 3300 - 5000 | 1.3401     | -6.243 x 10^{-5} | 5.503 x 10^{-9}  |
|          | $\rho$      | 2750 - 3250 | 0.2037     | 2.9027 x 10^{-4} |                 |
| Molybdenum: | 2895 - 3600 | 0.969     | 5.768 x 10^{-5} |                 |
|          | 400 - 2895  | -0.0384   | 2.8436 x 10^{-4} | -1.0970 x 10^{-8} | 3.6622 x 10^{-12} |
|          | $\rho_{IG}$ | 2895 - 3600 | 0.9535     | -1.1498 x 10^{-5} |                 |
|          | $\rho$      | 2895 - 3600 | 0.93106   | 3.95726 x 10^{-5} |                 |
| Tungsten: | 3680 - 5000 | 0.95062   | 6.344 x 10^{-5} |                 |
|          | 5000 - 6000 | 1.34989   | -1.0333 x 10^{-4} | 1.73957 x 10^{-8} |
|          | $\rho_{IG}$ | 423 - 1723 | -0.021     | 2.456 x 10^{-4}  | 1.201 x 10^{-8} |
|          | $\rho_{IG}$ | 2390 - 3687 | -0.059     | 3.166 x 10^{-4}  |                 |
|          | $\rho$      | 3687 - 5400 | 1.833      | -2.573 x 10^{-4} | 2.169 x 10^{-8}  |
|          | $\rho$      | 3687 - 5400 | 2.313      | -4.585 x 10^{-4} | 5.65 x 10^{-8}  |
Figure 1: Diameter expansions, $D^2/D_0^2$, as a function of temperature.

Figure 2: Electrical resistivities, $\rho$, as a function of temperature. Thin lines: values including volume expansion, dashed lines: melting transitions.

4. Discussion
The pulse-heating setup contains some peculiarities concerning the volume-correction of the electrical resistivity, which shall be considered first. In the formula for the specific electrical resistivity $\rho = U \cdot I^{-1} \cdot r^2 \pi \cdot l^{-1}$, with $U$: voltage drop, $I$: current, $r$: radius, $l$: length, one can see, that the consideration of thermal expansion requires knowledge of the change in radius (or diameter) and length. In our case, the wire can not change its length, because of the mechanical clamping. Due to improper experimental adjustments (e.g. unsatisfactory slow heating-rate) the wire will bend when its length increases leading to inoperative results. Hence the described monitoring of the cross-section is the best considered method: It delivers the thermal expansion of the diameter (later expressed as
and it will indicate, if the wire maintained its position during the heating process. The CCD camera delivers a picture of the diameter every 5 µs. The relation with the simultaneous temperature measurement yields the diameter \( D \) as a function of temperature.

As can be seen in figure 1, the five elements are not very different in their thermal expansions, except for niobium. The slope of Nb is steeper compared to the other metals. From this follows that the expansion of Nb in the solid phase has to be much lower than in the liquid phase as the ratio \( D^2/D_0^2 \) equals unity at room temperature. This common trend is approved by the two elements vanadium and tantalum, where solid state values are plotted too. They also show the typical increase at the melting-point.

The comparison of electrical resistivities exhibits a different behavior: Here vanadium is the element being outside of the bulk. It has the highest resistivity. W, Ta and Mo show a decreasing resistivity in the liquid state. This characteristics disappears when volume-expansion is taken into account.

It is easily noticeable from figure 2, that the temperature range is different for each material. Although pulse-heating would provide the possibility to measure resistivity values up to the end of the liquid phase, this additional experimental effort is barely appreciated within this project’s framework: No change in liquid phase behavior is expected at higher temperatures and the comparison with the results of our partners, obtained though levitation experiments, will be done some hundred degrees around the melting-point region.

Uncertainty consideration: The expanded relative uncertainty with a coverage factor of \( k = 2 \) of the electrical resistivity with initial geometry is 4%. The calculation for the thermal expansion in terms of GUM [11] is still missing, but even broad estimations of 0.04 (3.5%) for \( D^2/D_0^2 \) will only increase the uncertainty for specific resistivity to 6%.

5. Conclusion
We have presented our measurements of specific electrical resistivity on V, Nb, Ta, Mo, and W. The results shall be compared with the results obtained at our partner’s laboratory by means of levitation. This is one topic to be done in the course of our collaboration project.

TU Graz will proceed with the compilation of resistivity measurements. The improvement of density measurements with the pulse-heating method is the active research at TU Graz, because density values at high temperatures are sparse.

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