Experimental results and constitutive modelling for tungsten and tantalum at high strain rates and very high temperatures✩

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Abstract
Recently reported results of the high strain rates, high temperature measurements of the yield stress of tungsten and tantalum have been analyzed. The highest temperature reached in the experiment, based on heating and stressing a thin wire by a fast, high current pulse, was 2250 °C and 2450 °C, for tantalum and tungsten, respectively. The strain-rates in both the tungsten and tantalum tests were in the range from 500 to 1500 s⁻¹. The parameters for the constitutive equation developed by Zerilli and Armstrong have been determined from the experimental data and the results have been compared with the data obtained at lower temperatures.

Keywords: tungsten, tantalum, yield strength, high temperature, high strain rate

1. Introduction
Tungsten, tantalum and their alloys are the preferred candidates for many high strain rate applications, for example kinetic energy penetrators [1, 2]. Tungsten has been used for many decades in high temperature applications. Even so, there is a lack of experimental data on the thermomechanical response of refractory metals to high strain rate effects at temperatures higher

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The behaviour of materials under these conditions is of great interest from phenomenological point of view, for example for testing the consistency of the different constitutive models. In addition to this, the materials used in the target systems in the next generation of high power particle accelerators will be more and more frequently exposed to the combination of high stresses, high strain rates and high temperatures. In order to estimate the lifetime of the target and target system components in this environment, the candidate materials must be tested under these extreme conditions.

As part of the UK programme of high power target developments for a Neutrino Factory, a new dynamic method for thermomechanical characterization of the candidate materials (tungsten and tantalum in this case) has been developed. The method is based on heating and stressing a thin wire (less than 1 mm diameter) of the candidate material by a fast (\( \sim 1\mu s \)) pulse. This paper is a continuation of the study presented in where the yield strength of tungsten and tantalum has been measured at strain rates from 500 to \( 1500 \, s^{-1} \) and at temperatures much higher than previously recorded in the literature. The highest temperature reached in the experiment was 2250 °C and 2450 °C, for tantalum and tungsten, respectively. It should be noted that reference has a different aim and scope and that this paper is focused on the analysis of the new yield strength data, constitutive modelling and comparison with previous results for tungsten and tantalum at elevated temperatures.

The rest of the paper is organized as follows. The description of the experimental procedure and results are given in Section 2. Section 3 describes the constitutive modelling procedure with special attention paid to the Zerilli and Armstrong model. The results are discussed in Section 4, followed by the summary in Section 5.

2. Experimental procedure and results

A thin wire is necessary to allow the current to diffuse into the centre of the wire in a sufficiently short time to generate the required thermal stress. The tungsten and tantalum wires with diameters from 0.5 mm to 0.8 mm have been made by standard powder metallurgy methods - pressing, sintering, forging and finally drawing. The purity of the wires used in the tests was at least 99.9%. The tungsten wires have been additionally stress relieved at about 400 °C.
To determine the yield strength of the tantalum and tungsten wires the pulse amplitude of the current in the wire was increased in steps at a fixed temperature. The current was kept constant for 3-5 minutes before taking the next increase. This was continued until the wire was observed to start bending or kinking. The surface motion of the test wire during the pulsing is measured by a laser Doppler vibrometer (LDV) from Polytec [4]. The radial surface velocity measured by the LDV (see, for example, Figure 1), is used to extract the strain rate during the test. The optical sensor head of the LDV includes an integrated fast camera that is used to visually monitor the strain of the wire. In addition, it was noticed that the LDV radial velocity signal would become very noisy as one approached the first signs of plastic deformation, eventually resulting in the loss of any coherent vibration signal. This monitoring and the change of the quality of the LDV signal were the main indicators that the wire is near or at the bending point.

The bent wire was then replaced with a new, straight sample and the experiment was repeated at a new temperature and current. In some cases, the procedure was changed so the current was kept constant and the temperature was changed by adjusting the pulse repetition rate. In order to estimate the systematic uncertainties, at some temperatures there were several tests and it was found that the results were repeatable to within ±5% of the current. An optical pyrometer was used to measure the temperature of the wire at the same point measured by the vibrometer.

The stress in the wire is not directly measured, but it is proportional to the square of the current and can be modelled by using modern finite element codes. The first step is to measure the current pulse in the wire, then to calculate the current density as a function of time and radius using the solution to the diffusion equation [3]. Knowing the current density, the temperature rise is calculated and finally the finite element code LS-DYNA [9] is used to calculate the equivalent (von Mises) stress as a function of time and radius (more details can be found in [4, 5, 10]). As the stress in the wire is a result of calculation it is important to benchmark the results obtained. Figure 1 shows the comparison between measured radial surface velocity of the tungsten wire at different temperatures and the LS-DYNA results [5]. The agreement between experiment and simulation is very good and this can be taken as a proof that the corresponding calculated stresses are correct.

Figure 2 shows the stress at which the tantalum and tungsten wires reached the yield point as a function of temperature. The lower edge of the wide bands indicates the stress at which the wire still appeared unde-
formed and the upper edge the stress at which the wire started to bend. For tantalum, bands are shown for wires of 0.5 and 0.8 mm diameter. In both cases, the current was increased in the same, fixed steps. As the stress is inversely proportional to the square of the diameter, the band for the 0.5 mm diameter tantalum wire in Figure 2 is much wider than for the 0.8 mm diameter wire. In the case of tungsten, 0.5 mm diameter wires have been tested. The highest temperature reached in the experiment was 2250 °C and 2450 °C, for tantalum and tungsten, respectively. On the low side, it was not possible to go below 1150 °C (tantalum) and 1450 °C (tungsten) as the minimum temperature is limited by maximum available current.

The peak strain rate is determined by dividing the measured peak radial velocity (see Figure 1) by the wire radius. The peak strain rates for the measurements of both tantalum and tungsten cover the range (500 – 1500) s⁻¹. The characteristic values of the strain rate are indicated at various points in Figure 2. In this experiment the strain rates decrease at high temperatures. The strain of the wire during the pulsing was monitored by a fast camera. These observations were compared to the change in the quality of the LDV signal and it was found that the strain at the yield point was usually between 2% and 4%. Those values have been used also in the constitutive modelling (see Section 3).

Once a wire has begun to distort the stress in the wire increases due to the curvature of the wire. Also accompanying the bending, there is usually stretching and thinning of the wire at the hottest point, so the stress and temperature of the wire increases locally. This all happens within a few pulses and is soon followed by severe bending and/or breaking, so that it is difficult to measure the temperature, diameter and to calculate the true stress in the wire as the plastic strain increases. So, in this experiment it was not possible to measure the ‘complete’ stress-strain curve but only the yield point.

### 3. Constitutive modelling

Zerilli and Armstrong [6] proposed the following constitutive (Z&A) equation for the flow stress in body centred cubic (bcc) metals as a function of temperature $T$, strain $\varepsilon$ and strain rate $\dot{\varepsilon}$:

$$\sigma = C_1 e^{(-C_2T + C_3 T \ln \dot{\varepsilon})} + C_4 + C_5 \varepsilon^n + k d^{-1/2}. \tag{1}$$
$C_1$ to $C_5$, $n$ and $k$ are material parameters, while $d$ is the grain size. If the precise information about grain size is not available (as in our case), the usual procedure \cite{12, 2, 14, 15} is to incorporate the grain size term into the $C_4$ parameter because both are independent of temperature, strain and strain-rate:

$$\sigma = C_1 e^{(-C_2 T + C_3 T \ln \dot{\varepsilon})} + C_4 + C_5 \varepsilon^n.$$  \hspace{1cm} (2)

The first term in this equation combines effects of ‘thermal softening’ (reduction of yield strength with increasing temperature) and ‘strain-rate hardening’ (rise of yield strength with increasing strain-rate). This term does not depend on strain. The parameters $C_5$ and $n$ describe the strain hardening changes. In our case, $C_4$ and $C_5$ terms are practically constants (see the previous section) but they are left to vary during the fitting procedure.

The CERN computer package for function minimization MINUIT \cite{11} was used to vary the parameters $C_1$ to $C_5$ and $n$ and to minimise the sum of the squares of the deviations ($\chi^2$) between the values obtained from Z&A equation and experimental (fitting) points. The stress value, for each measured set of $(T, \varepsilon, \dot{\varepsilon})$, was chosen to lie in the middle between the corresponding upper and lower stress values (see Figure 2). The weighted least square fit was used, where the weight of the fitting point, $i$, is $w_i = 1/\sigma_i^2$, $\sigma_i$ being the half of the difference between upper and lower edge stress values at particular temperatures.

The best fit of the tantalum data is shown in Figure 3. One can note that the 0.5 and 0.8 mm data are combined into one set for fitting purposes. Due to the weighted fitting method the fitting curve has a characteristic shape in the region where the results overlap each others. Figure 4 shows the best fit for the tungsten data. In both cases the normalized $\chi^2$ value is close to 1.

The corresponding optimized parameters values for tungsten and tantalum are presented in Table 1. The values of the $C_1$, $C_2$ and $C_3$ parameters for tungsten and tantalum are very similar. This lead to the conclusion that the yield strength dependance on temperature and strain-rate (the slope of the curve) for tantalum and tungsten at very high temperatures and strain rates at around 1000 $s^{-1}$ is similar.

Finally, using an identical fitting procedure the experimental data was fitted with Johnson-Cook model \cite{16}, but this model was found to be inadequate for describing our experimental data. This is in agreement with previous conclusions that the Z&A model is much better equipped to capture strain-rate dependence and thermal softening behaviour of bcc metals.
4. Discussion

4.1. Tungsten

Table 2 shows the values of the parameters of the Z&A equation obtained in this work and previous experiments with tungsten samples.

A limited data set exists for pure tungsten behaviour at elevated temperatures and high strain-rates. Chen and Grey [12] have used commercially pure rolled tungsten plate for high strain-rate (0.001 – 4000 s\(^{-1}\)) tests at temperatures up to 1000 °C. Their fitting procedure resulted in a set of Z&A parameters denoted as 'CG' in Table 2 (and Figure 5 - see below).

Lennon and Ramesh [2] have measured and fitted ('LR' set in Table 2) the stress-strain curve of the samples made from heavily deformed extruded tungsten rod. The commercially obtained extruded rod had undergone about 80% reduction in area (about 60% effective strain). The samples were tested in the strain-rate range of (0.001 – 7000) s\(^{-1}\) and at temperatures ranging from 27 to 800 °C.

Tungsten samples for the tests performed by Dummer et al. [13] were subjected to different heat treatments. The as-received tungsten were made by pressing the tungsten powder at 1200 °C and then sintered at 2600 °C. The portions of the final samples were annealed at 1750 °C, 2600 °C and 2800 °C. The experiments were performed at room temperature only with the strain-rate ranging from 0.001 to 4000 s\(^{-1}\). The parameters of the Z&A equation obtained for as-received tungsten samples were practically the same as in the Chen and Grey experiment. In the case of annealed tungsten the yield stress was reduced by about 40%.

The common factor of these sets of tests is that the thermomechanical properties of tungsten were measured at temperatures well below the temperature region explored in this work. In an attempt to compare our new results with these, the tungsten yield stress was calculated using the Z&A constitutive equation for the strain rate of 1000 s\(^{-1}\) and strain of 3% (characteristic values in this experiment) and plotted in Figure 5 as a function of temperature. All parameter sets from Table 2 have been used and the results based on parameters from [12, 2] are extrapolated to cover the temperature range of our experiment. Visual inspection of Figure 5 reveals an excellent agreement between this work and the 'CG' parametrization of the Z&A equation while the 'LR' parametrization has a different slope.
4.2. Tantalum

Table 3 shows the values of the parameters of the Z&A equation obtained in this work and previous experiments with tantalum samples.

One of the first applications of the Z&A model was in the analysis of tantalum experimental results when Zerilli and Armstrong [14] fitted Hoge and Mukherjee experimental data [17]. Hoge and Mukherjee have tested 99% pure, fully recrystallized, tantalum in the strain-rate range of \((0.00001 - 20000) \, s^{-1}\) and at temperatures ranging from \(-249\) to \(527^\circ C\). The corresponding set of obtained parameters is denoted as ‘ZA-HM’ in Table 3 (and Figure 6 - see below).

Chen and Grey have fitted the same experimental results [17] by optimizing the fit for the entire range of data (parameter set ‘CG-HM’ in Table 3). They have also performed their own experiment described in the same paper [12], where commercially pure tantalum was tested in the strain-rate range of \((1500 - 5000) \, s^{-1}\) and at temperatures up to \(1000^\circ C\). One of the conclusions of the experiment was that the new data are very different from the Hoge and Mukherjee data [17]. The set of Z&A parameters that corresponds to Chen and Grey experimental results [12] are shown in Table 3 (denoted as ‘CG’).

In another Chen and Grey experiment [15] commercially pure, triple electron beam melted, vacuum annealed tantalum plates were tested in the strain-rate range of \((0.001 - 4000) \, s^{-1}\) and at temperatures ranging from \(-196\) to \(1000^\circ C\). Tantalum samples in those tests were prepared by melting large ingots, then forging them into billets which were then annealed and cut prior to cross rolling. Finally, the plates were straight rolled in the final finishing passes. The fitting of corresponding experimental data resulted in a set of Z&A parameters denoted as ‘CG-AP’ in Table 3.

As in the tungsten case, the common thing for all those experiments is that the thermomechanical properties of tantalum were measured at temperatures well below those explored in this work. In order to compare our experimental results with previous ones, the tantalum yield stress was calculated using the Z&A constitutive equation with a strain rate of \(1000 \, s^{-1}\) and strain of 3%. All parameter sets from Table 3 have been used and the results based on parameters from [12, 14, 15] are extrapolated to cover temperature

\[1\] In Hoge and Mukherjee experiment a different experimental technique (tension) was used. In all other experiments cited in this paper a Split-Hopkinson pressure bar technique has been used [18].
range in our experiment (see Figure 6).

Unlike the tungsten case, there is a clear difference in Figure 6 between each of the extrapolated parametrizations and the new experimental results. On the other hand, the extrapolated results differ significantly between themselves (up to a factor of 2) in the temperature region explored in our experiment, but there is a common feature for all of them: a very weak temperature dependence of the yield strength. However, our experimental results show a much stronger temperature sensitivity, and it is interesting that they almost fully cover the range between the two extreme extrapolated curves shown in Figure 6.

5. Summary

An analysis of the extensive set of high strain rates measurements of the yield strength of tungsten and tantalum at the record high temperatures (in the range of 1350 – 2700 K) has been performed. The strain-rates in both the tungsten and tantalum tests were in the range from 500 to 1500 s\(^{-1}\). The parameters for the constitutive equation developed by Zerilli and Armstrong have been determined from the experimental data. Also, it has been found that the Johnson-Cook model is inadequate for describing our test results. The obtained parametrization of the Zerilli-Armstrong model has been compared with the extrapolated parametrizations obtained in the tests at lower temperatures. It has been found that in the tungsten case our results and the extrapolated Chen-Gray parametrization agree very well. However, our experimental results for tantalum show a much stronger temperature sensitivity than the extrapolated parametrizations obtained from the tantalum tests at lower temperatures.

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Table 1. Z&A model parameters for tungsten and tantalum.

| Parameter | Tungsten (W) | Tantalum (Ta) |
|-----------|--------------|---------------|
| $C_1$ [MPa] | 4711±261 | 4166±354 |
| $C_2$ [K$^{-1}$] | $(5.97±0.28)$x$10^{-3}$ | $(7.89±0.41)$x$10^{-3}$ |
| $C_3$ [K$^{-1}$] | $(6.24±0.42)$x$10^{-4}$ | $(8.04±0.38)$x$10^{-4}$ |
| $C_4$ [MPa] | 94±8 | 7.5±0.9 |
| $C_5$ [MPa] | 133±20 | 381±40 |
| $n$ | 0.51±0.07 | 0.56±0.08 |
Table 2. Comparison between this work and previous results on Z&A model parameters for tungsten.

| Parameter | CG [12] | LR [2] | This work |
|-----------|---------|--------|-----------|
| $C_1$ [MPa] | 3000 | 2749 | 4711 |
| $C_2$ [$K^{-1}$] | $2 \times 10^{-3}$ | $2.25 \times 10^{-3}$ | $5.97 \times 10^{-3}$ |
| $C_3$ [$K^{-1}$] | $1 \times 10^{-4}$ | $9.05 \times 10^{-5}$ | $6.24 \times 10^{-3}$ |
| $C_4$ [MPa] | 0 | 49.91 | 94.37 |
| $C_5$ [MPa] | 800 | 194.5 | 133 |
| $n$ | 0.6 | 0.0505 | 0.512 |
Table 3. Comparison between this work and previous results on Z&A model parameters for tantalum.

| Parameter | ZA-HM [14] | CG-HM [12] | CG [12] | CG-AP [15] | This work |
|-----------|------------|------------|---------|------------|-----------|
| $C_1$ [MPa] | 1125       | 1200       | 975     | 1750       | 4166      |
| $C_2$ [K$^{-1}$] | 5.35x10$^{-3}$ | 6.0x10$^{-3}$ | 4.5x10$^{-3}$ | 9.75x10$^{-3}$ | 7.89x10$^{-3}$ |
| $C_3$ [K$^{-1}$] | 3.27x10$^{-4}$ | 3.875x10$^{-4}$ | 2.75x10$^{-4}$ | 6.75x10$^{-4}$ | 8.04x10$^{-4}$ |
| $C_4$ [MPa] | 30         | 25         | 40      | 140        | 7.5       |
| $C_5$ [MPa] | 310        | 310        | 525     | 650        | 381       |
| n         | 0.44       | 0.44       | 0.5     | 0.65       | 0.56      |
Figure 1. The measured and calculated radial velocity of a 0.5 mm diameter tungsten wire at peak temperatures of 920, 1260 and 1450°C.
Figure 2. The yield strength versus peak temperature for tantalum wires of 0.5 and 0.8 mm diameter and for tungsten wires of 0.5 mm diameter [5]. The upper edge of the bands indicates the stress at which the wire started to bend and the lower edge indicates where the wire was not deformed. The characteristic strain rate values are indicated.
Figure 3. Experimental data on tantalum yield strength and the best fit based on Z&A model.
Figure 4. Experimental data on tungsten yield strength and the best fit based on Z&A model.
Figure 5. Tungsten yield strength, calculated using the Z&A constitutive equation, as a function of temperature for the strain rate of $1000 \, s^{-1}$ and strain of 3%. Parameterizations 'CG' [12] and 'LR' [2] are extrapolated to cover the temperature range explored in [5].
Figure 6. Tantalum yield strength, calculated using the Z&A constitutive equation, as a function of temperature for the strain rate of 1000 s$^{-1}$ and strain of 3%. Parameterizations 'CG-HM' [12], 'CG' [12], 'ZA-HM' [14] and 'CG-AP' [15] are extrapolated to cover the temperature range explored in [5].