Micropillar compression of single crystal tungsten carbide, Part 1: temperature and orientation dependence of deformation behaviour

Helen Jones¹, Vivian Tong*, Rajaprakash Ramachandramoorthy²#, Ken Mingard¹, Johann Michler², Mark Gee¹

¹ National Physical Laboratory, Hampton Road, Teddington, Middlesex TW11 0LW, UK
² Empa, Feuerwerkerstrasse 39, 3603 Thun, Switzerland
* Corresponding author: vivian.tong@npl.co.uk
# Current affiliation: Max-Plank-Institut für Eisenforschung GmbH, Max-Planck-Strasse 1, 40237 Düsseldorf, Germany

Abstract

Tungsten carbide cobalt hardmetals are commonly used as cutting tools subject to high operation temperature and pressures, where the mechanical performance of the tungsten carbide phase affects the wear and lifetime of the material. In this study, the mechanical behaviour of the isolated tungsten carbide (WC) phase was investigated using single crystal micropillar compression. Micropillars 1-5 µm in diameter, in two crystal orientations, were fabricated using focused ion beam (FIB) machining and subsequently compressed between room temperature and 600 °C. The activated plastic deformation mechanisms were strongly anisotropic and weakly temperature dependent. The flow stresses of basal-oriented pillars were about three times higher than the prismatic pillars, and pillars of both orientations soften slightly with increasing temperature. The basal pillars tended to deform by either unstable cracking or unstable yield, whereas the prismatic pillars deformed by slip-mediated cracking. However, the active deformation mechanisms were also sensitive to pillar size and shape. Slip trace analysis of the deformed pillars showed that {1010} prismatic planes were the dominant slip plane in WC. Basal slip was also identified as a secondary slip system, activated at high temperatures.

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1 Introduction

The high compressive strength and wear resistance of hardmetals components is derived from the hexagonal WC phase, which forms the principal component of the hardmetal when sintered, with typically 3-15% of a ductile binder phase such as Co or Ni [1]. These hardmetal components are therefore commonly used for machining and drilling operations, in which they experience high loads and high temperatures of up to 1000 °C [2,3]. Understanding the high temperature properties of the WC phase is therefore important for modelling the material behaviour and predicting properties of new material grades.

Measuring the high temperature properties of any relatively brittle material such as WC can be challenging [4,5], and for WC there is the additional difficulty of anisotropic properties resulting from its hexagonal crystal structure. This means that measuring the change in properties, and ideally changes in deformation mechanisms too, as temperature increases, requires multiple identically oriented test samples. One approach applied to hardmetals has been to use nanoindentation at room temperature [6–14] and at high temperature [15]. This high temperature study produced good statistical data on the variation of hardness with temperature and orientation, and it was possible to give limited information on slip systems by using imaging of deformation around and below indents [15].

In this study, an alternative approach to indentation was used, by testing micropillars milled from a single crystal with a focused ion beam [16–22]. Compression of micropillars yields mechanical property data and enables observation of slip band formation at the pillar surfaces [21]. Analysis of the slip band orientation relative to the pillar crystal orientation enables the identification of the specific slip systems operating in WC to facilitate plastic deformation. As the next section will show, a wide range of studies of the slip systems in WC has generated a wide range of conclusions about which systems operate, so further data on this subject would be useful, especially with its extension to high temperatures. Many previous studies have relied on transmission electron microscopy (TEM) analysis of deformed samples [23–26], where identification of dislocations contributing to deformation can be difficult. This is particularly true if carried out on samples deformed at high temperatures, which also includes effects of thermal stresses caused by cooling. Analysis of slip steps observed at the surface of pillars minimises such issues, although it cannot be used to identify individual dislocation types.

However, micropillar compression is not straightforward. Even at room temperature, there can be issues of reproducibility arising from the variation in precise pillar dimensions, including, for example, edge rounding and side wall taper [27], and possible damage from the ion beam [28–31] used to produce the pillar. Care must be taken to minimise the possible effect of ion beam damage on nucleation and slip of dislocations from the sample surface. Testing pillars of a range of sizes can help in the identification of artefacts produced by the ion beam, and high temperature testing of micropillars can help with annealing of damage to the pillar. In this work, attention was paid to temperature control and especially in maintaining the sample and compression indenter tip at near identical temperatures to minimise thermal drift.

In this work, high temperature micropillar testing up to 600°C was employed on micropillars, milled in two WC single crystal samples, with orientations close to two principal
orientations: perpendicular to the basal \((0001)\) and to the prismatic \((10-10)\) planes. This builds on prior work [27] exploring the effects of micropillar size, shape and crystallographic orientation on the mechanical response and fracture behaviour of single-grain and polycrystalline WC-Co micropillars, but with the additional benefit that many similar micropillars can be fabricated in a single crystal. Measurement of the stress-strain curves and detailed post-test analysis of the slip bands made it possible to identify the slip planes that enabled deformation in the two orientations at room temperature and the operation of additional slip systems when the temperature increased above 300°C.

1.1 Slip systems and dislocation types reported in literature

The operative slip systems in WC control the deformation properties and mechanical strength of WC and WC-Co hardmetal composites, and is critical for interpreting slip trace data. Although an extremely wide range of slip directions and slip planes have been reported in published literature, many slip systems are reported in only once or twice, and only a few slip systems are consistently observed.

Most studies have focussed on room temperature deformation properties, and only a few studies are available on high temperature deformation of WC [15,32–36]. All of these have used indentation to study the degree of mechanical softening at elevated temperatures. However, indentation techniques necessarily produce a non-uniform plastic deformation field, which makes quantitative analysis of the mechanical data difficult. Furthermore, the deforming volume in indentation is constrained by the surrounding material, which may affect the crack propagation behaviour.

High temperature mechanical data of WC is important to understand and predict the in-service performance and lifetimes of hardmetal components, because their operating temperatures range between room temperature and 1000 °C. High temperature uniaxial micropillar compression has been chosen in this study to complement existing indentation work, as uniaxial compression can produce a nominally near-uniform deformation field, and deformation is less constrained compared to the indentation geometry.

1.1.1 Room temperature deformation

| Name          | Slip dir / Burgers vec | Slip plane     | TEM dislocations | Slip traces |
|---------------|------------------------|----------------|-----------------|------------|
| \(c\) prism 1 | \([0001]\)             | \(\{10\overline{1}0\}\) | \([23,24,37]\)   |            |
| \(c\) prism 2 | \([0001]\)             | \(\{11\overline{2}0\}\) | \([37]\)          |            |
| \(a\) basal  | \(\frac{1}{3}(11\overline{2}0)\) | \((0001)\) | \([37]\)     |            |
| \(a\) prism1 | \(\frac{1}{3}(11\overline{2}0)\) | \(\{10\overline{1}0\}\) | \([24,37]\)   | \([24]\)   |
| \(a\) pyr 1  | \(\frac{1}{3}(11\overline{2}0)\) | \(\{10\overline{1}1\}\) | \([\text{[24]}]\) |            |
| \(a_1 + a_2\) pyr 2 | \(\frac{1}{2}(\overline{1}100)\) | \(\{11\overline{2}2\}\) | \([\text{[24]}]\) |            |
| \(\langle c + a \rangle\) | \(\frac{1}{3}(11\bar{2}3)\) | \(\{10\bar{1}0\}\) | [37–39] | [24,38] |
|---|---|---|---|---|
| Prism 1 | unknown | \(\{10\bar{1}0\}\) | [24,37] | [24,41] |
| Pyr 2 | unknown | \(\{11\bar{2}2\}\) | [24] | [24] |
| Prism 1 | \(\{11\bar{2}0\}\) component | \(\{10\bar{1}0\}\) | [38,42,43] | [38,42,43] |
| Pry 1 | \(\{11\bar{2}0\}\) partial | \(\{10\bar{1}0\}\) | [37,39,40,43] | - |
| \(\langle c + a \rangle\) | \(\frac{1}{6}(20\bar{2}3)\) | \(\{11\bar{2}2\}\) | stacking fault / antiphase boundary | [23,44] | - |
| \(\langle a \rangle\) partial | \(\frac{1}{6}(11\bar{2}0)\) | Sessile stair rod from reaction of \(\langle c + a \rangle\) partials on adjacent \(\{10\bar{1}0\}\) planes | [24,39] | - |
| \(\langle c + a \rangle\) | \(\frac{1}{3}(11\bar{2}3)\) | Screw/unknown | [24,37,40] | - |
| \(\langle c \rangle\) | [0001] | Screw/unknown | [37,40] | - |
| \(\langle a \rangle\) | \(\frac{1}{3}(11\bar{2}0)\) | Screw/Unknown | [37,40,45] | - |

*Table 1: Summary of slip systems and dislocation types reported in WC at RT [23,24,37–44]. References in parentheses show where a slip system is reported but not directly observed from the experiment. Complete dislocation slip systems (i.e. both slip direction and slip plane) reported in more than one paper are shown in red text.*

Table 1 shows reported slip systems in room temperature (RT) deformed WC. Although several slip systems are reported, only three slip systems are reported by more than one study: \(\langle a \rangle\), \(\langle c \rangle\), and \(\langle c + a \rangle\) slip on prismatic planes. In addition, stacking faults bounded by \(\langle c + a \rangle\) partial dislocations are consistently observed in TEM studies of deformed WC.

The reported slip systems in Table 1 can be grouped according to two characterisation techniques: TEM dislocation analysis and slip trace analysis.

In TEM dislocation analysis, activated slip systems are inferred from the population of residual dislocations in a deformed material. The Burgers vector of a dislocation can be identified, either using \((g \cdot b)\) dislocation contrast analysis[44] or by drawing Burgers circuits in HR-TEM[39]; for an edge dislocation, the theoretical slip plane can be identified, but TEM cannot determine whether or not the dislocation was glissile during deformation.
However, the contributions of different dislocation types to plastic deformation cannot be directly measured from the residual defect population.

Slip trace analysis identifies the activated slip plane from the angle of deformation slip traces. The out-of-plane component of the slip direction can also be inferred from the height of surface slip steps, but the character of crystallographic defects which enable slip cannot be determined using slip trace analysis.

Most of the slip trace analysis experiments [24,38,41–43] showed that \{10\bar{1}0\} planes are primary slip planes in WC, and the slip directions can have components along \{1\bar{1}20\}, \{0001\}, or both. If slip is restricted to \{10\bar{1}0\} planes, the ductility of WC is limited because only four degrees of freedom are available with \{10\bar{1}0\} slip, so the von Mises yield criterion for arbitrary deformation is not fulfilled[46].

TEM analysis [23,24,37–40,43–45] show a variety of dislocation types, including perfect dislocations \(\frac{1}{3}\{1\bar{1}20\}, \{0001\}, \text{and } \frac{1}{3}\{1\bar{1}23\}\) Burgers vectors. In addition, \(\frac{1}{3}\{1\bar{1}23\}\) dislocations can dissociate into two identical \(\frac{1}{6}\{1\bar{1}23\}\) partial dislocations bounding a stacking fault in the \{10\bar{1}0\} plane.

A second dissociation reaction proposed was a dissociation into two \(\frac{1}{6}\{20\bar{2}3\}\) \((c + a_1 + a_2)\) partial dislocations[44] and supported by some TEM observations[23] but not others[40]. This reaction leads to antiphase boundaries in ordered alloys, but is most likely energetically unfavourable for WC, because glide of this partial dislocation would require large W atoms to occupy the sites of small C atoms.

Symmetrical variants of \(\frac{1}{3}\{1\bar{1}23\}\) dislocations (either undissociated or as partials with a stacking fault in between) can slip cooperatively on \{10\bar{1}0\} planes under an applied load to produce deformation with \{0001\} and \{1\bar{1}20\} components, or any combination of the two. Dissimilar \(c + a\) partial dislocations in the same \{10\bar{1}0\} plane can react to form glissile perfect dislocations with \(a\) or \(c\) Burgers vectors to allow slip in these directions[43]. However, \(c + a\) partials on different \{10\bar{1}0\} planes will react to form a sessile stair rod dislocation with \(\frac{1}{6}\{1\bar{1}20\}\) Burgers vector[24,39] which inhibits further slip.

### 1.1.2 High-temperature deformation

Few studies are available on high temperature slip in WC[15,32–36].

Activated WC slip systems and dislocation interactions are similar at room temperature (RT) and 1000 °C. TEM of WC crystals indented on the basal plane showed slip on \{1\bar{1}00\} planes and deformed structures containing stacking faults bounded by identical \(\frac{1}{6}\{1\bar{1}23\}\) partial dislocations, with similar defect interactions and sessile configurations for both temperatures[32]. This agrees with electron channelling contrast imaging observations of hardmetal WC grains, which showed dominant \{1\bar{1}00\} slip after nanoindentation on both basal and prismatic planes at temperatures from RT to 600 °C [15].

WC softens considerably with increasing temperature, though the same slip systems are activated[15,32,33]. The general hardness anisotropy of WC is preserved from RT to 900 °C:
basal planes are harder than the prismatic planes in both WC single crystals and hardmetal WC grains [15,33].

However, single crystal WC and cemented/polycrystal WC show different softening rates as temperature increases. The behaviour of hardmetal carbides [15] is also different to single crystals [33]. Single WC crystals of all orientations undergo rapid softening between RT and 500 °C, which levels off with further increase in temperature. Fine-grained hardmetals/polycrystalline WC soften less at lower temperatures, then rapid softening beyond 600 °C [33]. In individual hardmetal WC grains, prismatic-oriented grains show less softening between 100 and 400 °C, then a rapid drop from 500 to 700 °C. Basal-oriented grains have a nearly linear softening rate from 100 to 700 °C [15].

Grain boundary strengthening is an important strengthening mechanism for hardmetals deformed at RT and high temperature. Binderless polycrystalline WC (containing 0.3% Co) and cemented WC/Co composites are harder than all WC single crystals between RT and 900 °C [33], and increasing the WC grain size shifts the softening behaviour towards the single crystal case. This shows that the strength of polycrystalline WC or WC/Co composites between 600 °C and 900 °C is not controlled by WC slip, but instead by the grain boundary strength [33]. This agrees with TEM analysis of hardmetals deformed between 1100 °C and 1400 °C, which showed that plastic strain is accommodated mainly by grain boundary sliding [35]. WC/Co bending experiments at 1000 °C also showed that most plastic flow is accommodated by grain boundary sliding and cobalt flow (‘cobalt drift’) between WC grains [36].

However, in hardmetal creep at 1450 °C, plastic strain is accommodated mainly in WC grains [34], where dislocations with similar Burgers vectors accumulate to form intragranular WC subgrain boundaries. The majority of \( \frac{1}{6} \langle 11\overline{2}3 \rangle \) partial dislocations, which are frequently seen on \{1\overline{1}00\} glide planes in deformed WC [32,39,40,43], are recombined at 1450 °C into \( \frac{1}{3} \langle 11\overline{2}3 \rangle \) perfect dislocations through a dislocation climb process.

2 Experimental method
2.1 Material preparation

The material used in this study was pure WC, in the form of large grains approximately 2 mm in width. These were originally created inside a furnace during sintering, but on the sidewalls of the oven, instead of in the middle where the components were made. The grain growth was uninhibited, leading to very large single crystals. This enabled three arrays of 20 micropillars to be milled onto each single crystal for this experiment.

As the WC crystals were faceted, two crystals with a triangular and rectangular face were chosen, assuming the faces had orientations close to the basal and a prismatic plane respectively. To prepare the crystals, they were embedded in resin with the basal and prismatic planes flat and ground from underneath so that the top and bottom planes were parallel. The top face was then polished using 9 µm, 3 µm and 1 µm diamond pastes with a final stage of 30 nm colloidal silica, to remove scratches and surface damage due to the mechanical polishing. The crystals were removed from the resin and mounted on a stub, compatible with the indenter, using Omegabond600 thermally conductive high temperature
cement. The cement was then covered in a layer of high temperature electrically conductive paste to allow imaging with the SEM and reduce charging.

2.2 Pillar fabrication

Three sizes of pillars were prepared: the target top diameters were 1 µm, 2.5 µm and 5 µm and a target (top) diameter to height ratio of the pillars of 1:2.5. Twenty pillars of each size and in the two grains were fabricated, therefore 120 in total. This ensured there could be at least three pillars tested per temperature, with some spare. For the fabrication, a Carl Zeiss Auriga 60 focussed ion beam scanning electron microscope (FIB-SEM) with FIBICS nanopatterning system was used to mill the pillars. An example of one of the arrays is shown in Figure 1.

A rectangle was milled, long enough to view the side of the pillar at high sample tilt, and then an annulus with an outer diameter of greater than 20 µm, using a 30 kV:4 nA ion beam current. Two more annuli were milled with 30 kV:600 pA and 30 kV:240 pA with smaller diameters to minimise the side taper and gallium ion implantation. The set value of the inner diameter of the annuli were larger than the target diameter of the pillar, as the beam width caused wider milling than the set shape, especially at the top of the pillars. Six 4×5 arrays were milled with the rectangle and large annuli set to run automatically, and the two polishing annuli run manually, to minimise drift errors between the different sized annuli.

The actual geometries of the pillars varied slightly from the target values, where the average top diameters varied by ±0.2 µm, the average side tapers were between 5.9° and 2.3°, decreasing with increasing pillar size, and the diameter to height ratios varied between 1.8 and 2.3 (not dependent on size). The measurement method is shown for each micropillar size in Figure 1. The measurement of the height of the pillar using the SEM images is well-known to be prone to errors, due to the taper at the base of the pillar and milling artefacts caused by multiple passes with different FIB currents. Therefore, a possible uncertainty of up to 10%, especially in the largest pillars where the base was not flat and the bottom of the pillar was less visible, is a source of error when calculating the engineering strain.

The crystal orientations of the polished surfaces were measured using electron backscatter diffraction (EBSD). Unit cells showing the mean surface orientation of the prismatic and basal-oriented crystals are shown later in Figure 5b and Figure 6b respectively. The surface normal vector of the prismatic-oriented crystal was 3° from {10-10}, and basal-oriented crystal was 11° from [0001].
2.3 High temperature in situ setup

An Aleminis in situ high temperature nanoindenter fitted inside a Carl Zeiss DSM962 tungsten filament SEM was used to perform the pillar compression tests. The nanoindenter had a maximum load of 1 N and could operate up to a maximum of 800 °C.

The nanoindenter was tilted to 22° on the stage inside the microscope so that the pillars could be viewed during testing and so that the diamond flat-punch indenter tip could be positioned accurately over each pillar. The imaging conditions were an acceleration voltage of 15 kV, a current of 48 µA and working distance of 27 mm; a higher than usual working distance was used so that the nanoindenter could fit underneath the SEM electron column. The mechanical testing was conducted using a diamond flat-punch with a 10 µm diameter and 60° conical angle at the sides. The maximum displacement target was initially set to half the pillar height, and the test duration was kept constant, resulting in a constant engineering strain rate of 0.008 s⁻¹ for all the tests. In some cases, for example where there was significant softening, the tests were stopped manually to avoid reaching the point of catastrophic destruction. The Aleminis nanoindenter has a PID based feedback loop that allowed all the tests reported in this study to be conducted under true displacement control. Even though all precautions were taken to ensure uniform loading of the pillars, it was likely that there was not a perfect flat-on-flat contact between the indenter flat punch and the top of the pillars. Therefore non-uniform loading is possible in some cases and an initial toe region in the loading curves could be expected.

The high temperature tests were conducted using a heater and thermocouple combination attached to the sample and the diamond tip. The temperature of both the tip and sample was varied from room temperature up to 600°C with measurements taken at five nominal temperatures: 25°C, 150°C, 300°C, 450°C and 600 °C. The temperatures of both the tip and
sample were feedback-controlled using a PID loop, based on the reading from the thermocouples attached. A temperature gradient was present between the actual location of thermocouple and the sample surface, as the thermocouple was not positioned on the sample surface itself. Therefore, in order to avoid thermal drift in the displacement signals, due to potential mismatch of temperatures between the tip and the sample beyond the thermal stabilization period, careful temperature matching calibrations were conducted at each testing temperature for both individual samples. This calibration involved conducting a load-controlled flat punch indentation on bulk sample surface with a drift hold segment. The tip and sample temperatures were then fine-tuned to match, based on achieving a low thermal drift of ~0.03 nm/s during the drift hold segment (more information on the instrumentation available here[47]).

3  Results and analysis

3.1  Stress-strain

![Stress-strain curves](image)

**Figure 2:** Loading portion of stress-strain curves of all pillars, grouped by pillar size (rows), loading orientation (columns), and test temperature (plot colours). One line per pillar type is plotted in thicker lines to help visualization, and nominally identical repeats are drawn as thin lines. See text for details of annotation marks.

Engineering stress and strain were calculated from the load and displacement signals through division by the micropillar top cross-sectional area and pillar height respectively.
The stress-strain data was also corrected for the elastic sink-in of the pillar into the substrate using Sneddon's correction[48] and the indenter frame compliance specific to the high temperature version of the nanomechanical tester.

Stress-strain plots of the loading portions for all pillars are shown in Figure 2. The plastic flow stresses were about three times higher for basal than prismatic oriented pillars. Both orientations showed a ‘smaller is stronger’ size effect when comparing flow stresses in the 1 µm and 2.5 µm pillars, but negligible difference between the 2.5 µm and 5 µm pillars.

3.1.1 Basal oriented pillars

The basal oriented pillars could be divided into two groups showing either stochastic or deterministic plastic flow. Deterministic flow is characterised by smooth, reproducible stress-strain curves typically seen in macro-scale mechanical testing of ductile materials. Stochastic flow is characterised by stress drops due to intermittent dislocation activity, and elastic deformation in between, and a wide scatter in flow stresses between nominally similar pillars. Stochastic flow occurs in micropillars because few dislocation sources are available in a small pillar volume, which limits the number of glissile dislocations available for slip. For the micropillars in this study, both stochastic dislocation glide and crack propagation events can lead to stress drops.

Stochastic flow was observed in smaller pillars deformed at lower temperatures: 1 µm pillars deformed at all temperatures, and 2.5 µm pillars deformed at 150 °C and at room temperature. The stress-strain curves contained intermittent stress drops and wide scatter in flow stress between similar pillars. Two characteristic groups of stress drops are visible in these pillars: large stress drops (2-10 GPa) likely to be related to cracking, and smaller stress drops (< 0.5 GPa) likely to be related to dislocation slip. Table 2 shows that the large stress drops are correlated with more severe cracking in the deformed pillars. No significant temperature dependence was observed in basal pillars deformed by stochastic flow.

Deterministic flow was observed in larger pillars deformed at higher temperatures: 5 µm pillars at all temperatures, and most of the 2.5 µm pillars deformed at 300, 450 and 600 °C. The stress-strain curves were repeatable between pillars. An initial peak at around 2 % compressive strain was followed by linear strain softening until 7 % strain, then a nearly flat region after a ‘knee’ in the stress-strain curve. The green and pink arrow annotations in Figure 2e show small horizontal steps in some of the pillars deformed between 300 and 600 °C, just before the main stress peak at 2 % strain. These most likely correspond to dislocation pop-in events, so some stochastic behaviour was still present. Flow stress decreased with increasing temperature in the 5 µm pillars. In the 2.5 µm pillars, flow stresses did not vary much with temperature between 300 °C and 600 °C.

3.1.2 Prismatic oriented pillars

The prismatic oriented pillars deformed by stochastic plastic flow with intermittent stress drops, which could be due to either crack propagation or intermittent dislocation nucleation events. The gradients of the stress drops were steeper at smaller pillar sizes, i.e. a stress drop results in a smaller strain in the 1 µm pillars, and a larger strain in the 5 µm pillars. There was a weak trend of decreasing flow stress with increasing temperature for all pillar sizes, but this was modulated by a wide scatter in the stress-strain behaviour of nominally similar pillars. Many small fluctuating stress drops were observed in all prismatic pillars. As
the pillar size increased, the stress drops increased in both stress amplitude and strain accommodated per drop. This can be seen in the pairs of horizontal and vertical dashed black lines in Figure 2, which mark the start and end positions of a ‘typical’ stress drop. Note that the dashed lines are intended only as a qualitative visual guide, and a wide range of stress drops can be readily observed. The largest stress drops measured in each pillar are shown later in Table 2 and Table 3.

3.2 Load-time

![Load-time graphs for all pillars, grouped by pillar size (rows), loading orientation (columns), and test temperature (plot colours). One line per pillar type is plotted in thicker lines to help visualization.](image)

**Figure 3:** Load-time graphs for all pillars, grouped by pillar size (rows), loading orientation (columns), and test temperature (plot colours). One line per pillar type is plotted in thicker lines to help visualization.

Load-time plots of pillar deformation are shown in Figure 3. All plots showed load drops, regions of high deformation rate, which could be either from dislocation nucleation based pop-in events or unstable deformation modes such as plastic buckling or cracking.

As expected from Figure 2, the applied load increased with pillar size and was higher in basal pillars than prismatic pillars. In these data, 1 µm prismatic pillars (Figure 3b) reached maximum loads of 5 to 10 mN, whereas 5 µm pillars (Figure 3f) reached maximum loads of 50 to 150 mN.

Most of the load drops were immediately followed by a steep load increase. The load drops in prismatic pillars were smaller and tended to fluctuate around an overall increasing load. In basal pillars, the load drops were larger and more widely spaced. Basal pillars with deterministic stress-strain curves all showed a single large load drop, which was not
followed by a corresponding load increase. This load drop corresponded to linear softening in the stress-strain curves between 2 and 7 % compressive strain, in Figure 2c and e.

3.3 Yield

![Graph showing yield stress variation with temperature](image)

**Figure 4:** a) Variation of yield stress with temperature for each pillar orientation and size. Red lines/markers correspond to basal pillars, and blue lines/markers to prismatic pillars. Markers plot yield stress for individual pillars, and lines show the mean value for a group of similar pillars. b) Prismatic oriented pillar data only from a) plotted on a re-scaled vertical axis.

The 'yield point' was defined as the first stress drop in the stress-strain curves, corresponding to the first dislocation pop-in or crack initiation event. Figure 3 plots the yield stresses of individual pillars and trends from the mean yield stress of each pillar type.

The yield strength was anisotropic: basal oriented pillars are between three and six times stronger than the prismatic oriented pillars. Both basal and prismatic pillars exhibited a weak 'smaller is stronger' size effect: 1 µm pillars of both orientations had the highest yield stress, but very similar yield stresses for the 2.5 and 5 µm pillars, with small measured differences likely within the experimental scatter.
The yield strength was weakly dependent on temperature. On average, the pillars yielded at lower stresses at high temperatures, but the temperature dependence was similar in magnitude to the measurement scatter. The 1 μm basal pillars were an exception to this, yield softening from 14 to 12 GPa between RT and 450 °C, but an anomalous increase to 19 GPa at 600 °C. This suggests that the yield mechanism of the 1 μm basal pillars changed between 450 °C and 600 °C.

3.4 Strain hardening

Figure 5 shows the change in stress per unit strain for all pillars. We have called this ‘strain hardening’ as the vertical axis plots what would be the work hardening rate in conventional macroscopic tests. In this work, which investigates micropillar deformation dominated by stochastic dislocation activity, the absolute ‘work hardening’ values are not meaningful. This plot is simply a helpful way to visualise the stress-strain data, by removing the large variation in measured stress values caused by stochastic slip and cracking events, so that correlations and trends between the groups of pillars can be explored.

In order to compute the local strain hardening, a Lowess smoothing filter was applied to the data to suppress small high-frequency fluctuations in stress and strain from the electronic
noise of the support hardware. The Lowess filter uses locally weighted linear regression to fit the stress-strain data to a linear spline [49]. A moving window size spanning 1% of the nearest neighbour points was used for these data. Supplementary Figure 1 shows the smoothed stress-strain curves, and the difference between original and smoothed data. The stress differences between the original and smoothed data are small (99.9% of points < 0.2 GPa) and decrease with increasing pillar size.

In Figure 5, negative values indicate strain softening, positive values indicate hardening. Sharp spikes correspond to load drops (negative spikes) and jumps (positive spikes) during stochastic flow. Widely scattered distributions correspond to stochastic slip deformation or unstable crack propagation, whereas a narrow distribution indicates deterministic stress-strain behaviour.

The average hardening of prismatic pillars was near-constant between 100 and 200 GPa, and did not depend on deformation temperature or pillar size. The scatter range was wide for 1 µm pillars, between −400 and 700 GPa, and narrowed as the pillar size increased to 5 µm and deformation behaviour became less stochastic. Load drops (negative regions) were shallower and limited to about −100 GPa in the 2.5 µm and 5 µm pillars.

The scatter range of basal pillars widens with decreasing pillar size as the deformation tends towards stochastic flow. The strain hardening plots in Figure 5 show that all basal pillars of all sizes deform in three distinct regimes: (1) strain hardening (400 to 800 GPa) below 2% strain corresponding to ‘elastic loading’ before the first load drop, (2) strain softening (−50 to −200 GPa) between 2 and 6% strain, and (3) nearly zero hardening (−50 to +100 GPa) beyond 6% strain. The average strain hardening values in these three regimes vary slightly between pillar sizes, but are independent of temperature.

The numerical ‘strain hardening’ values are not physically meaningful in these micropillars, and should not be extrapolated to macroscopic samples. However, plotting the data in this way enabled a common behaviour in the basal pillars, that was not obvious from the stress-strain plots in Figure 2, to be clearly visualised.

### 3.5 Cracking behaviour

After testing, all pillars were imaged at high resolution in the SEM in two sample orientations to characterize the degree of cracking. Representative micrographs for each deformation conditions are shown in Table 2 (basal pillars) and Table 3 (prismatic pillars). Where multiple types of deformation patterning were observed in nominally similar pillars (e.g. 1 µm / 150 °C basal pillars in Table 2), representative SEM micrographs are shown for each type. Larger versions of Table 2 and Table 3 are provided as a supplementary document file.

The stochasticity of stress-strain data (Figure 2), and the largest stress drop measured from the load-time data (Figure 3) are also tabulated for each pillar, since unstable crack propagation could be linked to large stress drops. In basal oriented pillars, the first large stress drop near the yield point was ignored as this was observed in all pillars, even those that did not contain a crack.
Table 2: Relationship between deformation cracking and stochastic stress-strain behaviour in basal oriented micropillars. The unit cells show the crystal orientation on the pillar front wall, and have been vertically rescaled to match the angles at 45° sample tilt used for imaging these pillars. The background colours of the ‘Max stress drop’ cells are formatted to match the stress drop size: 0 GPa = green, 3 GPa = yellow, ≥ 7 GPa = red. A larger version of this table is provided as a supplementary document file.

The basal pillars showed two types of characteristic cracking modes: unstable crack propagation and catastrophic fracture in small pillars deforming at low temperatures, and ductile plastic deformation and stable crack growth in large pillars at high temperatures.

In Section 3.1.1, basal oriented pillars are grouped into pillars with stochastic versus deterministic stress-strain curves. Table 2 shows that stochastic deformation occurred in all pillars with extensive cracking down the pillar length along a \{10\overline{1}0\} plane (red dashed lines), and large stress drops are typically > 5 GPa. In contrast, pillars with deterministic deformation showed minimal cracking, but instead severe pillar bending, resulting in curved slip traces. The cracks which formed in these pillars are in an ‘X’ shape in the tensile part of the bent pillar wall. The crack traces appear to lie along \(c + a\) directions (blue and green dashed lines), although this is uncertain due to the severe lattice bending in these pillars.

The crack plane could be uniquely identified where surface traces are visible on both the top face and side wall, such as the prismatic crack plane marked by red dashed lines in Table 2 (1 µm pillars, 150 °C). The crack plane could be partially identified where only one surface trace is visible, such as the ‘X’-shaped cracks marked by blue and green dashed lines in Table
2 (2.5 µm pillars, 300 °C); in these cases, the crack planes must contain the \(\langle c + a \rangle\) surface trace directions.

| Temp. / °C | Max stress drop / GPa | Stochastic? |
|------------|-----------------------|-------------|
| 25         | 1.9 1                 |             |
|            | 2.1 1                 |             |
|            | 1.3 1                 |             |
| 150        | 0.9 1                 |             |
|            | 1.3 1                 |             |
|            | 0.9 1                 |             |
| 300        | 1.2 1 1 0.8 1 1      |             |
| 450        | 1.6 1 1.4 1 1.2 1    |             |
| 600        | 1.7 1 0.6 1 0.5 1    |             |

Table 3: Relationship between deformation cracking and stochastic stress-strain behaviour in prismatic oriented micropillars. The unit cells show the crystal orientation on the pillar front wall, and have been vertically rescaled to match the 45° sample tilt used for imaging these pillars. The background colours of the ‘Max stress drop’ cells are formatted to match the stress drop size: 0 GPa = green, 3 GPa = yellow, ≥ 7 GPa = red. A larger version of this table is provided as a supplementary document file.

In the prismatic pillars, the severity of cracking increases with pillar size. No temperature dependence was observed in either of the types of cracks that formed or the severity of cracking.

All the 1 µm pillars deformed by planar shear on a single slip plane containing an \(\langle a \rangle\) direction (blue dashed line in unit cell schematic and on images), and some pillars had a small crack lying in the slip plane. In the 5 µm pillars, double slip was activated in most of the pillars, with slip traces along two \(\langle a \rangle\) directions, and cracks propagated along the intersection of the two activated slip planes (orange dashed lines). Some pillars contained several cracks across the pillar width, but none failed catastrophically, which suggests that cracking in the prismatic pillars was a slip-mediated deformation mode. The behaviour of
the 2.5 µm pillars was somewhere in between, showing both single slip and double slip in different pillars.

Double slip activation increased the average flow strength of the pillars: out of three 5 µm prismatic pillars deformed at room temperature, double slip and cracking occurred in two pillars, and single slip with minimal cracking in one pillar. The corresponding stress-strain curves (Figure 2f, brown lines) showed that the double-slipped pillars had an average plastic flow stress of 5 GPa, whereas the single-slipped pillar had a lower average plastic flow stress, around 3 GPa.

### 3.6 Slip trace analysis

![Figure 6: Slip trace analysis of 5 µm diameter micropillar compressed at 600 °C along [1010].](image)

(a) SEM images of the pillar top face and side walls at different viewing angles, showing three sets of crystallographic slip traces (solid lines) and cracks (dashed lines), marked in red, blue and purple. (b) Unit cell showing crystal orientation from EBSD of the undeformed sample. (c) Closest matching slip traces on the pillar top face: bars show expected slip trace angles from EBSD orientation; lines are experimentally measured from (a). (d) Closest matching slip traces on the pillar side walls: lines show expected slip trace angles from EBSD orientation; scattered points show experimental measurements for red, blue and purple slip traces in (a).

Activated slip planes in the 5 µm pillar arrays were measured for each temperature using slip trace analysis, to infer likely slip systems. Figure 6 and Figure 7 show the slip trace analysis method in example datasets of 5 µm pillars compressed at 600 °C for the [1010] and [0001] orientations respectively.

Pillar top faces were imaged at 0° stage tilt, and pillar side walls were imaged at 45° tilt from eight viewing directions (360° in 45° increments) for slip trace analysis. Pillar slip trace angles were measured in ImageJ[50], applying tilt correction if necessary. The red, blue and purple lines in Figure 6a show slip traces (solid lines) and crack paths (dashed lines) corresponding to the three sets of slip planes that were identified from the images. To
measure a single slip trace angle from a curved side wall surface, tangent lines were drawn from slip traces near the front of the pillar for each viewing direction.

Crystal orientations of the undeformed samples were determined using EBSD. Figure 6b shows the hexagonal unit cell orientation with loading direction going into the page; the pillar top face is near {10\overline{1}0}. Sample orientations during EBSD and slip trace imaging are shown in Figure 6a and Figure 6b as keyhole-shaped schematic diagrams of the pillar and viewing trench.

Expected slip trace angles on the pillar top face and side walls were computed in MTEX for the WC slip systems reported in Table 1. Pillar side walls were approximated as perpendicular to the pillar top face in this calculation, without accounting for the 2-3° taper angle in the 5 µm pillars (Figure 1).

The measured slip traces were matched to expected crystallographic planes in the sample. Since a slip trace is a planar section through the pillar, slip traces on the pillar top face are expected to be straight lines, and the slip trace angles on the pillar side wall are expected to vary sinusoidally as the viewing angle is rotated through 360°.

Figure 6c compares measured slip traces (vertical lines) and the closest matching expected slip traces (bars) on the pillar top face. Figure 6d compares the measured (markers) with closest matching slip traces (sinusoid lines) expected for the pillar side walls. The slip planes can be indexed as (\overline{1}100) (red), (\overline{1}010) (blue), and (01\overline{1}0) (purple) planes, by matching the measured slip traces to the expected slip traces on both the pillar top face (Figure 6c) and side walls (Figure 6d).

A few degrees of difference between the measured and expected slip traces was reasonable, and most likely due to sample misalignments between slip trace imaging and EBSD orientation measurement, in addition to lattice rotations that occurred during deformation, and uncertainty in identifying the front-facing part of the pillar when measuring side wall slip traces. Measured slip traces were taken from the deformed crystal at 45° stage tilt, whereas the expected slip traces were calculated from EBSD of the undeformed crystal at 70° tilt. The slip traces could be confidently indexed because consistent solutions were found from nine measurements (an arbitrarily oriented plane has two degrees of freedom).

Each prismatic slip plane contained four possible slip directions: \langle a \rangle, \langle c \rangle, and two \langle c + a \rangle directions: \langle c \rangle − \langle a \rangle and \langle c \rangle + \langle a \rangle. This can be seen in Figure 6c, where four bars per colour corresponds to four possible slip systems per prism plane. Once the activated slip plane was identified, Schmid factors were used to determine the likely slip systems by finding the slip direction with the best geometric alignment. The legend in Figure 6d shows that the slip system with highest Schmid factor for each activated slip plane was [11\overline{2}0](\overline{1}100) (red slip traces, 0.42), [1\overline{2}10](\overline{1}010) (blue slip traces, 0.38), and [0001](01\overline{1}0) (purple slip traces, 0.25).
Figure 7: Slip trace analysis of 5 µm diameter micropillar compressed at 600 °C along [0001]. (a) SEM images of the pillar top face and side walls at different viewing angles, showing three sets of crystallographic slip traces (solid lines) marked in red, blue and yellow. The dashed purple line shows a theoretical trace expected from the pillar side wall slip traces, but not was not directly observed. (b) Unit cell showing crystal orientation from EBSD of the undeformed sample. (c) Closest matching slip traces on the pillar top face: bars show expected slip trace angles from EBSD orientation; lines are experimentally measured from (a). (d) Closest matching slip traces on the pillar side walls: lines show expected slip trace angles from EBSD orientation; scattered points show experimental measurements for the slip traces in (a).

Figure 7 shows the same slip trace analysis method applied to a [0001] oriented 5 µm pillar compressed at 600 °C. The slip traces were twisted into ‘S’-shapes about the pillar loading axis, and the top of the deformed pillar was rotated away from the base. In contrast, the [1010] pillar in Figure 6 showed planar slip and minimal plastic rotation. Slip traces on the pillar side walls were measured near the top of the pillar to minimise the error contribution from lattice rotation during slip trace analysis (see Appendix A).

Measured slip traces in this pillar corresponded to (0110) (red), (1010) (blue), (1100) (purple), and (0001) (yellow) slip planes. The (0110) (red) and (1010) (blue) planes were most likely the primary slip systems as most of the slip traces were of these planes; the (1100) (purple) and (0001) (yellow) slip planes are likely secondary slip systems. Schmid factor analysis showed that the possible slip directions on the measured planes are all poorly oriented for slip, with Schmid factors ≤ 0.22. The least poorly oriented slip systems were [0001] (0110) (red, Schmid factor 0.21), [0001] (1010) (blue, Schmid factor 0.17), [1123] (1100) (purple, Schmid factor < 0.1), and [1120] (0001) (yellow, Schmid factor 0.22).

3.6.1 Additional slip systems activated at high temperature

One pillar per temperature and orientation was analysed using the method in Section 3.6. Table 4 shows the slip planes activated in each pillar, where additional slip planes activated
at high temperatures are marked in bold font. The basal pillar deformed at RT had cracked during testing, which tilted large portions of the pillar and increased the measurement uncertainty; two sets of secondary slip traces could not be assigned to a crystallographic plane. All sets of primary slip traces were assigned successfully.

\{10\bar{1}0\} prismatic planes were the dominant slip planes in the basal pillars despite poor geometric alignment, which confirms that prismatic planes are the only dominant primary slip plane in WC between RT and 600 °C, and the few reported TEM observations [24,40] of dislocations on \{10\bar{1}1\} and \{11\bar{2}2\} pyramidal planes (summarised in Table 1) are unlikely to have contributed significantly to plastic deformation.

| Orientation | Room Temp. | 150 °C | 300 °C | 450 °C | 600 °C |
|-------------|------------|--------|--------|--------|--------|
| Prismatic   | [11\bar{2}0](\bar{1}100)\[1\bar{2}\bar{1}0](\bar{1}010) | [11\bar{2}0](\bar{1}100)\[1\bar{2}\bar{1}0](\bar{1}010) | [11\bar{2}0](\bar{1}100)\[1\bar{2}\bar{1}0](\bar{1}010) | [11\bar{2}0](\bar{1}100)\[1\bar{2}\bar{1}0](\bar{1}010) | [11\bar{2}0](\bar{1}100)\[1\bar{2}\bar{1}0](\bar{1}010) |
| Basal       | [0001](0110)\[0001](\bar{1}010) | [0001](0110)\[0001](\bar{1}010) | [0001](0110)\[0001](\bar{1}010) | [0001](0110)\[0001](\bar{1}010) | [0001](0110)\[0001](\bar{1}010) |

*Table 4: Slip planes activated during micropillar compression as a function of test temperature and orientation. The expected slip direction is calculated by maximising Schmid factor in the undeformed pillar orientation.*

4 Discussion

4.1 Effect of pillar shape: unstable cracking versus slip in basal pillars

Over the range of temperatures and pillar sizes tested, the deformation behaviour of basal pillars changes between deterministic and stochastic flow. The stress-strain curves in Figure 2, and stress drop data and SEM images in Table 2 showed that larger pillars deformed at higher temperatures favoured deterministic flow, with small or no load drops, and ductile plastic deformation with stable cracking. Smaller pillars deformed at low temperatures favoured stochastic flow, with large load drops, and unstable crack propagation.

Whether unstable cracking and ductile plastic deformation occurred depended on other factors as well as the nominal pillar size and loading temperature. Pillar-specific parameters such as pillar taper, edge rounding, and indenter tip alignment could also affect the local stress state and cause a switch between the two modes.

For example, in Table 2, basal compression tests at 600 °C / 2.5 µm both show two nominally similar pillars that deformed by different modes. Inspection of their respective stress-strain curves in Figure 2c shows that stochastic flow (thin pink line) correlated with catastrophic cracking, and deterministic flow (thick pink line) correlated with plastic buckling and small X-shaped cracks. SEM images before deformation (Supplementary Figure 3) showed that the pillar that underwent stochastic flow had anomalously rounded top edges from FIB milling, which would have affected both the FIB damage layer thickness and the local stress state.

The effect of pillar shape parameters can also be seen in the proportional loading parts of the stress-strain curves in Figure 2c, which are divided between either stochastic or
deterministic flow, independent of test temperature. All deterministic stress-strain curves in Figure 2c had a higher proportionality constant of about 600 GPa, whereas all stochastic curves had a lower proportionality constant of about 400 GPa. (For comparison, the Young’s moduli of single crystal WC, calculated from elastic constants in Lee and Gilmore[51,52], are 820 GPa and 600 GPa in the basal and prismatic directions respectively.) These observations are elastic, where previous finite element model simulations[27] have showed that increasing the edge rounding in micropillars (1) decreases the measured proportionality constant, (2) amplifies the local compressive stress where the indenter contacts first at the pillar top centre, and (3) produces corresponding radial tensile stresses which work to open cracks on the pillar top face. This explains why, out of the two pillars tested under nominally similar conditions, the pillar with more edge rounding deformed by catastrophic cracking, and the other by ductile plastic deformation and stable crack growth.

Table 2 shows that in basal-oriented pillars, catastrophic cracking is observed in small pillars but suppressed with increasing pillar size, which is the opposite to the typical size effect observed in brittle ceramics, where cracking is suppressed and ductility increases below a threshold size[53,54]. One possible explanation is that the stress state of the pillars changed with pillar size: rounded edges occupied a larger fraction of the cross-section in smaller pillars, and edge-rounding produced high axial compressive and radial tensile stresses in the top centre of the pillars, so that cracks opened outwards. Smaller pillars also had a larger taper angle, which amplifies the axial loading stress in the top part of the pillar where the cross-section area is smaller.

4.2 Origin of load drops and stochastic deformation events

In situ imaging during micromechanical tests was not possible due to limited resolution available at high temperatures in the tungsten filament SEM, so the load drops and other features in the mechanical stress-strain data could not be directly correlated to individual pillar deformation events. Even so, variation in deformation mechanisms could be linked to the mechanical behaviour through representative post-test SEM images and the mechanical data, as summarised in Table 2 and Table 3.

In the basal pillars, the smaller pillars underwent catastrophic fracture, but the larger pillars deformed by plastic yielding. Catastrophically fractured pillars tended to have large stress drops > 5 GPa, whereas plastically yielded pillars had relatively smooth stress-strain curves with stress drops of no more than 0.5 – 1 GPa. The ‘maximum stress drop’ value reported in Table 2 excluded the first large load drop observed in the ductile deformed basal pillars (e.g. Figure 3e), which was seen in pillars that did not crack at all, and unlike all other load drops, was not followed by a steep load increase. This load drop likely corresponds to plastic buckling, where unstable yielding was caused by the pillar softening as it deforms. As such, the deformed pillars were bent, and the slip traces were highly curved (e.g. Figure 7, 5 μm pillars in Table 2).

In the prismatic pillars, the maximum load drop size (Table 3) did not vary strongly with pillar size, but the severity of cracking was much larger for the 5 μm pillars than the 1 μm pillars. The larger pillars were more likely to have double slip activity, presumably due to more dislocation sources available in the larger pillar volume. Double-slipped pillars were also more likely to contain cracks, due to the interactions between the two slip systems. Comparing Figure 2b to Figure 2d showed that the load drops in the 5 μm prismatic pillars
accommodate more compressive strain than the 1 µm prismatic pillars. The load drops in 1 µm pillars were likely related to intermittent dislocation nucleation events, whereas load drops in 5 µm pillars likely corresponded to a combination of dislocation nucleation and crack propagation. Since the crack ran along the intersection of two slip planes, the crack growth was stable, as the stress ahead of the crack tip was quickly dissipated through dislocation slip activity.

In summary, stochasticity in the stress-strain curves of basal pillars were related to unstable cracking, whereas stochasticity in the prismatic pillars were related to dislocation slip and slip-mediated stable crack growth. Plastic buckling was observed in basal pillars as a large load drop near the yield point which was not followed by a stress increase.

4.3 Comparison to existing studies

4.3.1 Activated slip systems

The slip trace analysis results in Table 4 confirmed literature reports that slip in WC occurs predominantly on \{10\overline{1}0\} planes (Table 1). The slip traces of the deformed pillars in this study were qualitatively similar to the 2 µm diameter WC micropillars shown by Csanádi [21]. Although, in this study, additional secondary slip planes were also identified to be activated at high temperatures: a third \{10\overline{1}0\} slip plane in the prismatic pillars at 600 °C and the basal pillars at ≥ 300 °C, and \{0001\} slip in the basal pillars at 600 °C.

Slip on only prismatic planes cannot accommodate compressive strain along [0001]. Prismatic slip dominated in both prismatic and basal oriented pillars in this work, because the pillars were oriented a few degrees away from prismatic or basal planes. The experimentally measured pillar loading axes were 3° from \{10\overline{1}0\} for the prismatic pillars, and 11° from \(\langle0001\rangle\) in the basal pillars.

From the present data, we can expect prismatic slip to be the dominant slip plane for loading directions ≥ 11° from [0001], but cannot extrapolate this to loading directions closer than 11° from [0001]. This is consistent with the basal-oriented, RT deformed, WC micropillars of Csanádi et al. [21], which showed no signs of plastic deformation before brittle failure.

Slip on the \(\langle0001\rangle\) plane has not been directly observed in WC literature, although Greenwood and Loretto observed \(\langle a\rangle\) edge dislocations in the basal plane[37]. Density functional theory simulations from Nabarro et al.[55,56] predicted a Peierls stress only twice that of \(\langle a\rangle\) dislocations on \{10\overline{1}0\}, so from their analysis alone, basal slip is expected as a secondary slip system. Out of the reported WC slip directions and dislocation types (Table 1), \(\langle a\rangle\) dislocations have a Burgers vector in the basal plane and therefore can cross-slip onto the basal plane, but \(\langle c + a\rangle\) dislocations cannot. The slip direction of the basal slip trace was not experimentally determined in this work, but the maximum possible Schmid factor is 0.22 for \{11\overline{2}0\}(0001) \(\langle a\rangle\) basal slip.

4.3.2 Temperature dependence

Literature studies[15,33] of high temperature WC indentation report that indentation hardness is higher on basal planes than prismatic planes. The yield anisotropy in this study
(Figure 4) showed that basal planes are stronger than prismatic planes in compression, which is consistent with these reports.

The same studies[15,33] also reported a two to three-fold reduction in hardness between RT and 600 °C. However, in the present study, the micropillar compression yield stresses (Figure 4) were only weakly temperature dependent. Also, the increase in cracking at nanoindentation temperatures ≥ 500 °C reported in De Luca et al.[15] was not observed in the micropillar compression in this present study.

A significant difference between the deformation conditions in nanoindentation and micropillar compression was that the deforming volume is fully constrained in nanoindentation, but laterally unconstrained in micropillar compression. This suggests that increased cracking at high temperatures observed in nanoindentation is caused by increased constraint around a larger plastically deformed volume, and is consistent with the slip-mediated cracking seen in the 5 µm prismatic micropillars at all temperatures (Table 3). Similarly, indentation softening between RT and 600 °C might be specific to the loading geometry and constraint imposed by the surrounding material, as it is not observed to the same extent in micropillar compression where the pillar side walls were unconstrained.

The anomalous increase in yield strength of 1 µm basal pillars as deformation temperature increases from 450 °C to 600 °C is neither seen in literature reports nor reproduced in the larger pillars in this study. The reason for this is unknown from the present data and left as an open question for future investigations.

5 Conclusion

Micropillar compression has been used to measure the anisotropic and temperature-dependent deformation behaviour of tungsten carbide single crystals. The pillars were approximately three times stronger when compressed along near-basal orientations compared to near-prismatic orientations.

On average, small basal pillars at low temperatures deformed by unstable cracking, large basal pillars at high temperatures deformed by unstable plastic deformation, and prismatic pillars deformed by planar slip and slip-mediated stable cracking.

The activated deformation mechanisms were anisotropic and temperature dependent, but were also sensitive to pillar size and shape, notably side wall taper and edge rounding, which cause non-uniaxial pillar loading, stress gradients within each pillar, and variable stress states between pillars.

The dominant slip plane was \{10\bar{1}0\} for temperatures between RT and 600 °C, and orientations with loading direction ≥ 11° from [0001]. This was consistent with results reported in the literature. Only slight softening was observed with increasing temperature to 600 °C, which was different to nanoindentation studies of similar materials and temperatures.

Basal slip was also identified as a secondary slip system active at high temperature. \langle a \rangle basal slip has been previously suggested as a possible slip system[37,55,56], but to the authors’ knowledge, not been experimentally observed.
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7 Author Contributions
HJ prepared the micropillar samples; HJ and RR performed the compression tests under guidance from JM; HJ and VT performed the SEM imaging. VT analysed the mechanical data using MATLAB scripts written by HJ. VT performed the slip trace analysis. KM supervised the work throughout. VT drafted the manuscript with input from KM, HJ, RR and MG. MG and JM inspired the project by initiating a collaboration between Empa and NPL.

8 Appendix – lattice rotations from mechanical constraint during micropillar compression
During a compression test, the pillar is mechanically constrained by the rigid flat-punch indenter face on top of the pillar, and by the rest of the sample at its base. The indenter constrains the pillar top face in the vertical direction, but the in-plane rotational and lateral constraint depends on friction between the pillar and indenter. The coefficient of friction is not known in this experiment.

Supplementary Figure 2 shows a similar basal pillar to the one in Figure 7, which rotated during compression. The pillar has bent so that the top part of the deformed pillar is laterally displaced from the base, but the top face has remained normal to the loading direction. This confirms that top of the pillar was vertically constrained and could not tilt out of plane, but laterally less constrained so that it could slide under the indenter. Since the top of the pillar can slide, but cannot tilt, crystal rotation from slip is likely to be minimum in this region.

EBSD analysis of a basal pillar deformed at 600° is described in part 2 of this paper [57]. The crystal lattice near the top of the deformed pillar is within 5° of the undeformed orientation, which confirms the validity of this model.
9 Supplementary Figures

Supplementary Figure 1: *(a1-f1)* Stress-strain curves with Lowess smoothing filter applied to remove thermal drift before calculating strain hardening in Figure 5. *(a2-f2)* Difference between the smoothed and original stress-strain curves (plotted in Figure 2).

Supplementary Figure 2: Mechanical constraint at the top and middle of a micropillar during compression. The top of the pillar cannot tilt but may slide laterally or rotate in-plane under the flat punch indenter tip. The middle of the pillar is constrained by the material around it so that it cannot slide, except where dislocations are emitted at the pillar surface.
**Supplementary Figure 3:** SEM images of 2.5 µm basal pillars before and after deformation at 600 °C. a) Pillar with less edge rounding deformed by deterministic stress-strain behaviour, unstable yielding and stable crack growth; b) Pillar with more edge rounding deformed by stochastic stress-strain behaviour and unstable axial cracks.

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