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Incipient plasticity in tungsten during nanoindentation: dependence on surface roughness, probe radius and crystal orientation

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Abstract

The influence of crystallographic orientation, contact size and surface roughness effects on incipient plasticity in tungsten were investigated by nanoindentation with indenters with a range of end radius (150, 350, 720 and 2800 nm) in single crystal samples with the (100) and (111) orientations. Results for the single crystals were compared to those for a reference polycrystalline tungsten sample tested under the same conditions. Surface roughness measurements showed that the R_a surface roughness was around 2, 4, and 6 nm for the (100), (111) and polycrystalline samples respectively. A strong size effect was observed, with the stress for incipient plasticity increasing as the indenter radius decreased. The maximum shear stress approached the theoretical shear strength when W(100) was indented using the tip with the smallest radius. The higher roughness and greater dislocation density on the W(111) and polycrystalline samples contributed to yield occurring at lower stresses.

Keywords: tungsten; anisotropy; nanoindentation; incipient plasticity

Abbreviations:

<i>BCC</i>	Body centred cubic
<i>FCC</i>	Face centred cubic
<i>ISE</i>	Indentation size effect
<i>NPL</i>	National Physical Laboratory
<i>RMS</i>	Root mean square

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34	Nomenclature:	
35	a	Contact radius
36	Al	Aluminium
37	E or E_s	Elastic modulus of the material
38	E_r	Reduced elastic modulus
39	h^*	Characteristic length
40	h_c	Contact depth
41	h_{max}	Depth under maximum load at pop-in
42	h_r	Residual indentation depth
43	H_0	Macroscopic hardness
44	L	Applied load
45	G	Shear modulus
46	Mo	Molybdenum
47	P_m	Mean contact pressure
48	p_0	Maximum contact pressure
49	R	End radius of the indenter
50	R_a	Average surface roughness
51	Ta	Tantalum
52	W	Tungsten
53	τ_{max}	Maximum shear stress
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58 1. Introduction

59 Tungsten (W) is a technologically important BCC metal with potential applications in the next
60 generation of nuclear reactors, being a favoured choice for plasma-facing components in fusion
61 reactors [1-3]. ISO:14577 specifies that W being very close to elastically isotropic (Zener anisotropy
62 ratio is 1.01) while having a high elastic modulus makes it an important reference material for
63 indirectly calibrating nanomechanical test instruments due to its high sensitivity to the instrument
64 frame stiffness. It has the highest melting point of all the metals and its high temperature
65 nanomechanical behaviour is beginning to be explored [4]. However, as yet relatively little attention
66 has been given to the influence of crystallographic orientation, loading rate and surface roughness,
67 and how these might influence size effects in incipient plasticity and hardness at the nano-/ and
68 micro/-scale [5].

69 During nanoindentation, both BCC and FCC metals can show displacement bursts that are
70 known as “pop-ins” [6-8]. Typically in BCC metals such as W, Cr, Mo and Ta, a single yield event
71 is observed while for close packed metals multiple pop-in (“staircase”) behaviour is more common

72 [9]. It is generally accepted that with a sharp indenter, incipient plasticity at the pop-in event occurs
73 due to homologous dislocation nucleation and the shear stress required can approach the theoretical
74 strength [10]. The presence of thick thermally grown oxide layers can modify the stress
75 distributions under the indenter so that a pop-in may be associated with oxide fracture [6]. However,
76 as the native oxide on tungsten is much thinner, of the order of ~ 0.7 nm thick at room temperature
77 [11], oxide fracture is not thought to contribute to the observed behaviour [12]. Shim et al. [13]
78 noted that the increase in strength as the size of the contact decreases can be considered to be a
79 different type of indentation size effect to that commonly seen in hardness, since the latter depends
80 on the yielding and work-hardening behaviour of the material and the former on the stress to initiate
81 dislocation plasticity. In a fusion reactor, tungsten is subjected to intense bombardment from alpha
82 particles and hydrogen ions which can cause indentation size effects [14]. Being able to deconvolute
83 the origins of the different indentation size effects (ISEs) on the observed behaviour is essential
84 since they will all contribute to the behaviour at a similar scale (e.g. within ~ 100 nm of the surface).

85 Yao et al. [1] reported a dependence on crystallographic orientation on electrochemically
86 polished, vacuum annealed (12h at 950 °C) and D-implanted single crystal tungsten with the critical
87 load for pop-in with a $R = 675$ nm indenter being much larger on (100) and (110) surfaces than on
88 the (111) orientation. Contrarily, they found no orientation dependence for hardness.
89 Stelmashenko et al. [15] reported Vickers hardness measurements showing higher hardness and
90 higher pile-up around the indentations for W(100). Pethica's group noted that after mechanical
91 polishing a number of dislocation systems are active at low load in W(100) and a clear single pop-in
92 was not observed [16]. They also reported that the hardness of mechanically polished W samples
93 determined at depths higher than the pop-in event was higher than that of electropolished samples.

94 Most studies on incipient plasticity of pure metals have used indenters of one or at most two
95 radii, making the effect of tip radius difficult to establish accurately. There have been two recent
96 reports using a wide range of tip radius. Shim et al. [13] studied the influence of indenter radius (R
97 = 0.58 to 209 microns) on pop-in occurring in the FCC metal Ni(100) and reported that the critical

98 loads and maximum shear stresses under the indenter increased as the radius decreased. Wu et al.
 99 [17] investigated the onset of plasticity in the BCC metal chromium using indenters with tip radius
 100 ranging from 60-759 nm and also found that the stress required for incipient plasticity increased
 101 with a reduction in tip radius.

102 There has been recent interest in the influence of the surface state on the load required for
 103 pop-in [8, 9, 12, 14]. Although it is generally accepted that pop-in events require highly polished
 104 surfaces, it is a common practice in the literature for either the surface roughness to not be quoted or
 105 for only an approximate measure of R_a to be provided. On Al(001), Shibutani et al. [8] observed that
 106 the critical load scaled inversely with surface roughness. A reduction in R_a from ~2.5 nm to under
 107 0.5 nm resulted in the critical load increasing by a factor of 3. Bahr et al. [6] reported that, as
 108 opposed to electropolished surfaces, mechanically polished W single crystals did not show pop-ins.
 109 Biener et al [9] reported that on Ta(001) there was no difference between electropolished or
 110 mechanically polished surfaces provided a high-quality surface finish was obtained. They found a
 111 tight distribution in the critical load for pop-in for a Ta(001) surface with the RMS roughness well
 112 below 1 nm. Introducing surface roughness on Ta by low-energy Ar^+ ion bombardment suppressed
 113 the linear elastic regime and the pop-in behaviour.

114 This work reports novel findings obtained from nanoindentation experiments performed on
 115 tungsten samples. The objective of this study was to investigate the contribution of size effects to
 116 incipient plasticity in tungsten using a wide range of indenter radius (0.15-2.8 microns). Alongside
 117 this, the influence of crystallographic orientation, loading rate and surface roughness were also
 118 studied on single crystals of tungsten with the (100) and (111) orientation and a reference
 119 polycrystalline tungsten sample. Nanoindentation data at a lower load were supplemented by
 120 measurements to 500 mN to determine the conventional indentation size effect in hardness.

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124 2. Experimental

125 2.1 Materials

126 Two high purity polished tungsten single crystals and a high purity polycrystalline tungsten certified
127 reference sample were tested. The sample with (100) orientation was provided by KRISS (Korea),
128 originally for the VAMAS TWA22 Intercomparison on nanoindentation, being supplied by
129 Goodfellow (USA) and polished by KRISS. The sample with (111) orientation was supplied by
130 Goodfellow (UK) and was of thickness 2 mm and diameter 6 mm, and was polished on one side to
131 better than 1 micron (W 002166). The quoted elastic modulus and Poisson's ratio of the samples
132 were 411 GPa and 0.28 respectively. The polycrystalline certified reference tungsten sample ("JGA-
133 105", Instrumented Indentation Reference Block, DataSure-IIT, NPL, Teddington, UK) was
134 obtained from NPL, based in the UK. Its elastic modulus and Poisson's ratio were determined by
135 NPL in accordance with BS EN 843-2:2006. The certified value of E obtained by NPL was $411.5 \pm$
136 1.9 GPa and the Poisson's ratio was 0.2806 ± 0.0017 . The density of the polycrystalline sample was
137 1.9259 g cm^{-3} . The sample was coarse-grained with an average grain size in the region of $10 \mu\text{m}$.
138 The tungsten samples were tested as-received and no further attempt was made to modify surface
139 roughness or near-surface defect density by further polishing or annealing steps. Surface roughness
140 was measured over a line profile using the Surface Topography option in the Scanning Module of
141 the NanoTest using (i) a spheroconical diamond probe with a nominal end radius of 5 microns (the
142 actual end radius was separately determined as 4 microns) (ii) a well-worn Berkovich indenter with
143 an end radius of $1 \mu\text{m}$. Surface roughness of the single crystal samples was also measured at the 5
144 $\mu\text{m} \times 5 \mu\text{m}$ scale by AFM (NanoSurf Nanite B). Table 1 summarises the surface roughness data.
145 The AFM images revealed the presence of very fine polishing marks on the surface of the (111)
146 oriented W which were absent on the (100) oriented W.

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150 **Table 1. Surface Roughness of the measured samples**

	R_a surface roughness (nm)		
	AFM (5 μm x 5 μm area)	Line scan with $R = 1.0$ μm diamond (over 10 μm length)	Line scan with $R = 4.0$ μm diamond (over 10 μm length)
W(100)	1.4 ± 0.6	2.0 ± 0.3	2.3 ± 0.5
W(111)	4.0 ± 0.7	3.1 ± 0.4	5.5 ± 1.6
Polycrystalline W	Not measured	5.5 ± 1.4	6.9 ± 2.1

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152 *2.2. Nanoindentation*

153 Nanoindentation testing of the tungsten samples was performed with a commercial nanomechanical
 154 test instrument (NanoTest Platform 3, Micro Materials Ltd., Wrexham, UK) which had been
 155 calibrated in accordance with the ISO 14577-4. The polycrystalline W was used to determine the
 156 frame compliance of the instrument which was confirmed by measurements in other reference
 157 metallic samples. The end radii of the diamond indenters were calibrated by fully elastic
 158 nanoindentation measurements into fused silica and sapphire reference samples. Three of the
 159 indenters used were Berkovich indenters of different end radius and one was spheroconical
 160 diamond with a nominal end radius of 5 μm . The fused silica was a nanoindentation intercomparison
 161 reference sample (obtained from KRISS, Korea) with a nominal elastic modulus of 72.5 GPa and
 162 Poisson's ratio of 0.17. Its elastic properties were separately cross-checked against those of a
 163 certified sample (JGC-105, NPL DataSure-IIT reference block) and were found to be consistent to
 164 well within 0.5 %. The sapphire was a single crystal with (001) orientation (an intercomparison
 165 reference sample from the EU "Nanoindent" project supplied by Roditi, UK). The end radii were
 166 150, 350, 720 and 2800 nm.

167 The loading conditions for the four indenters are summarised in Table 2.

168

169 **Table 2. Nanoindentation test conditions**

	Loading rate ($\mu\text{N/s}$)	Peak load (μN)	Hold at peak load (s)	Unloading rate ($\mu\text{N/s}$)
$R = 150$ nm	25, 100	500	3	50
$R = 350$ nm	25, 50, 100, 200	500	3	50
$R = 720$ nm	25, 100	1000	3	333
$R = 2800$ nm	100	3000	3	333

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171 Adjacent indentations were made sufficiently far apart (30 μm) to avoid influence from interaction
 172 of indentations. The mean values of the critical load, depth, mean pressure and maximum shear
 173 stress at pop-in together with their standard deviations for each of the indenters are summarised in
 174 Table 3 (a-d). The mean values shown in Table 3 were derived from 35-50 indents for each
 175 sample/loading rate/indenter combination with the three sub-micron radius indenters and from 20
 176 indents for $R = 2800 \mu\text{m}$.

177 **Table 3(a). Pop-in behaviour with the 150 nm end radius indenter**

	Critical load (μN)	Depth at pop-in (nm)	Mean pressure at pop-in (GPa)	Maximum shear stress at pop-in (GPa)
W(100) at 25 $\mu\text{N/s}$	153 \pm 99	9.2 \pm 3.8	33.7 \pm 6.7	15.4 \pm 3.1
W(100) at 100 $\mu\text{N/s}$	124 \pm 85	7.2 \pm 3.0	32.2 \pm 6.6	14.4 \pm 2.8
W(111) at 25 $\mu\text{N/s}$	36 \pm 13	3.1 \pm 0.9	24.8 \pm 3.4	9.8 \pm 1.1
W(111) at 100 $\mu\text{N/s}$	37 \pm 13	3.4 \pm 0.8	23.2 \pm 2.9	9.9 \pm 3.5

178
 179 **Table 3(b). Pop-in behaviour with the 350 nm end radius indenter**

	Critical load (μN)	Depth at pop-in (nm)	Mean pressure at pop-in (GPa)	Maximum shear stress at pop-in (GPa)
W(100) at 25 $\mu\text{N/s}$	182 \pm 91	7.8 \pm 2.8	20.3 \pm 3.3	9.4 \pm 1.5
W(100) at 100 $\mu\text{N/s}$	173 \pm 102	7.1 \pm 3.0	21.1 \pm 3.6	9.2 \pm 1.7
W(111) at 25 $\mu\text{N/s}$	69 \pm 60	4.3 \pm 2.3	13.3 \pm 2.6	6.7 \pm 1.4
W(111) at 100 $\mu\text{N/s}$	67 \pm 29	4.3 \pm 1.3	16.2 \pm 1.3	6.8 \pm 0.7
Polycrystalline W at 25 $\mu\text{N/s}$	74 \pm 62	4.3 \pm 3.0	14.7 \pm 2.1	6.7 \pm 1.8
Polycrystalline W at 100 $\mu\text{N/s}$	97 \pm 49	5.5 \pm 2.4	18.7 \pm 1.7	7.6 \pm 1.3

180
 181 **Table 3(c). Pop-in behaviour with the 720 nm end radius indenter**

	Critical load (μN)	Depth at pop-in (nm)	Mean pressure at pop-in (GPa)	Maximum shear stress at pop-in (GPa)
W(100) at 25 $\mu\text{N/s}$	226 \pm 91	6.8 \pm 2.0	14.3 \pm 2.8	6.3 \pm 1.1
W(100) at 100 $\mu\text{N/s}$	237 \pm 89	6.5 \pm 1.8	15.8 \pm 2.2	6.4 \pm 0.8
W(111) at 25 $\mu\text{N/s}$	97 \pm 95	4.2 \pm 3.1	8.5 \pm 2.8	4.3 \pm 1.6
W(111) at 100 $\mu\text{N/s}$	100 \pm 51	4.2 \pm 1.5	10.2 \pm 1.6	4.8 \pm 0.8

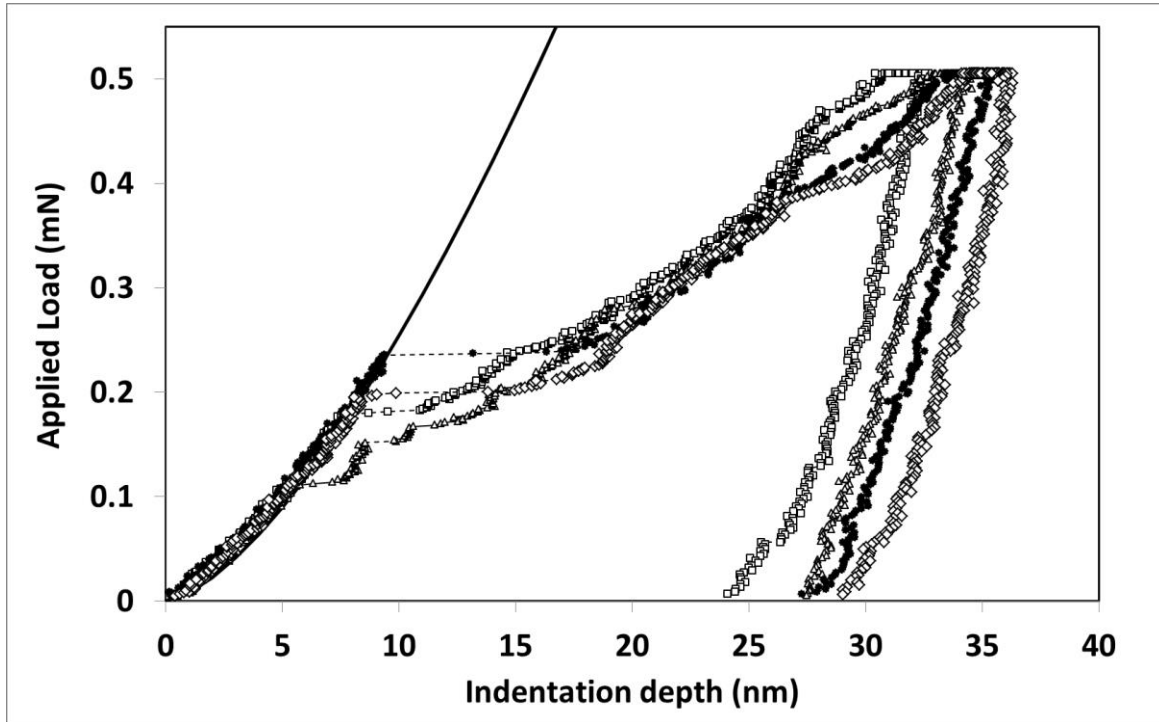
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 184 **Table 3(d). Pop-in behaviour with the 2800 nm end radius indenter**

	Critical load (μN)	Depth at pop-in (nm)	Mean pressure at pop-in (GPa)	Maximum shear stress at pop-in (GPa)
W(100)	380 \pm 134	7.3 \pm 1.7	5.7 \pm 0.8	3.0 \pm 0.4
W(111)	250 \pm 137	5.3 \pm 2.0	5.0 \pm 1.2	2.6 \pm 0.5

185 Measurements were also performed with the indenter with end radius $R = 350$ nm over the load
 186 range 10-500 mN where it has the Berkovich geometry. The loading rate was 10 mN/s, the
 187 unloading rate was 20 mN/s and the hold at peak load was for 30s. Additional tests were run with a
 188 loading time constant equal to 15 s and unloading equal to 2 s. No evidence of rate sensitivity after
 189 the 30 s hold at peak load was found, which is consistent with the small indentation creep strain of
 190 tungsten at room temperature reported in the past [4]. The thermal drift correction was from 40 s in
 191 contact prior to loading and at 90% unloading in all the tests. The reduced indentation modulus (E_r)
 192 is related to the elastic modulus (E_s) of the material according to $\frac{1}{E_r} = \frac{1-\nu_s^2}{E_s} + \frac{1-\nu_i^2}{E_i}$ where E_i is the
 193 elastic modulus of the diamond indenter and ν_s and ν_i are the Poisson's ratios of the sample and
 194 indenter respectively. For tungsten, a reduced indentation modulus of 321 GPa corresponds to an
 195 elastic modulus of 411 GPa. The mean contact pressure up to pop-in can be determined from
 196 Hertzian mechanics as $P_m = L/\pi a^2$ where L is the applied load and the contact radius a is given by
 197 $a = \sqrt{2Rh_c - h_c^2}$ where $h_c = (h_{\max} + h_r)/2$ so that h_c is the contact depth, h_{\max} is the depth under load
 198 at pop-in and h_r is the residual depth which is taken as zero as the contact is fully elastic to the
 199 points considered. The maximum shear stress (τ_{\max}) can also be determined from Hertzian
 200 analysis. At pop-in, $\tau_{\max} = 0.31p_0$ where $p_0 = \sqrt[3]{\frac{6PE_r^2}{\pi^3R^2}}$.

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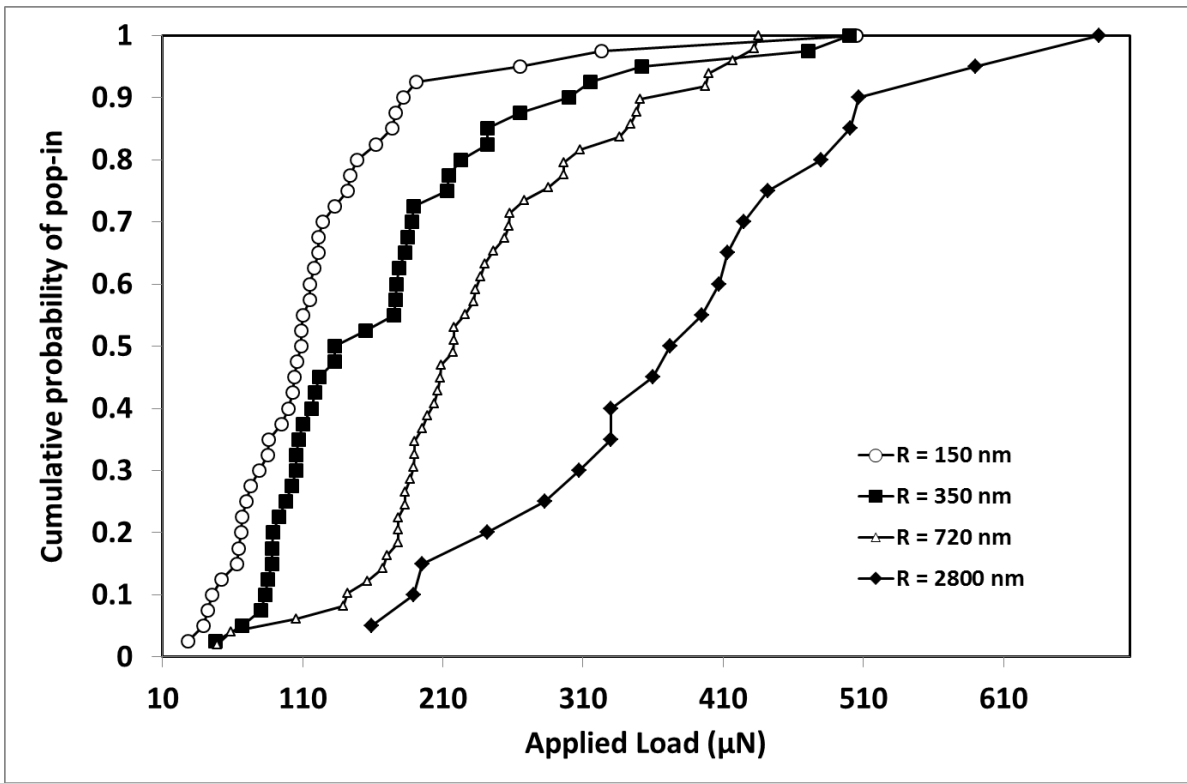
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203 **3. Results**

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Figure 1: Typical indentation behaviour on the W(100) with the $R = 350$ nm tip

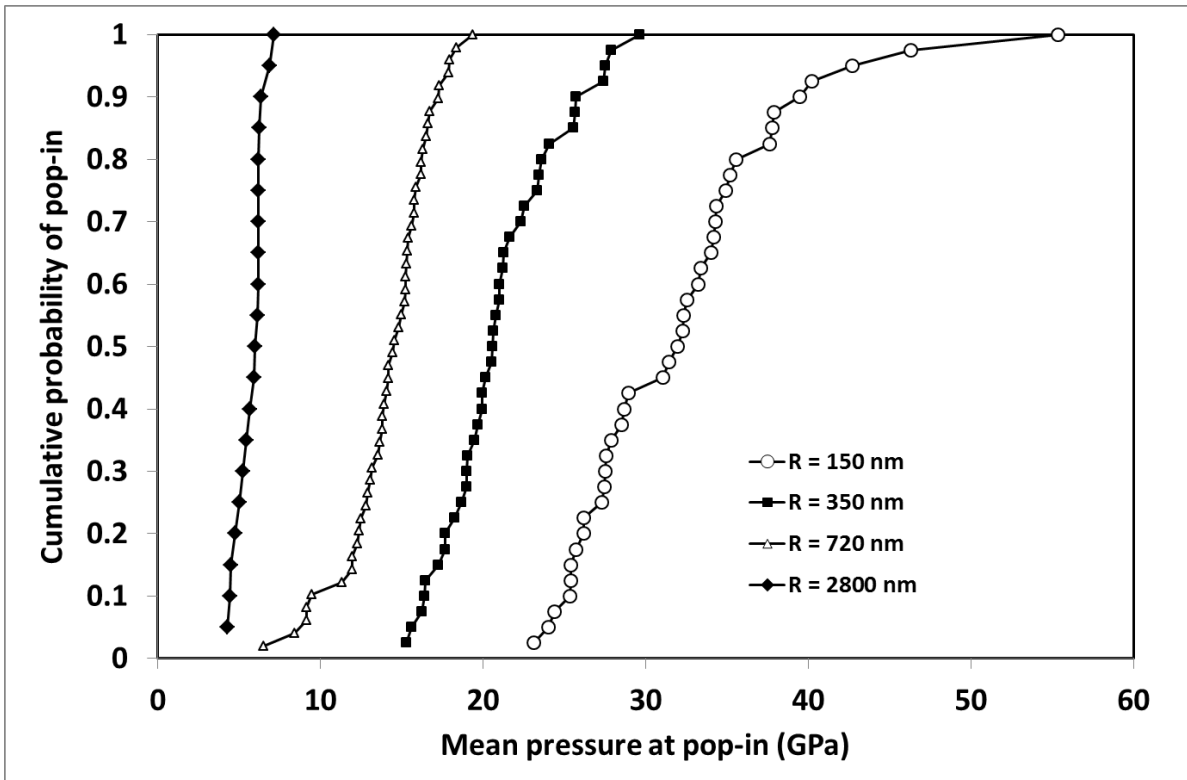
208 Typical indentation behaviour on the W(100) with the $R = 350$ nm probe is shown in figure 1. The
 209 loading behaviour is elastic up until a pop-in occurs. If no pop-in occurred before the peak load was
 210 reached, then the contact was completely elastic and the entire loading curve could be fitted by
 211 Hertzian mechanics (the dotted line in figure 1) using the power-law relationship $P \propto h^{1.5}$ according
 212 to $P = \frac{4}{3} E_r R^{0.5} h^{1.5}$. The pop-in data with the $R = 350$ nm probe at $100 \mu\text{N/s}$ are displayed as
 213 cumulative probability plots in figures 2(a) to 2(d).



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215 Figure 2(a)

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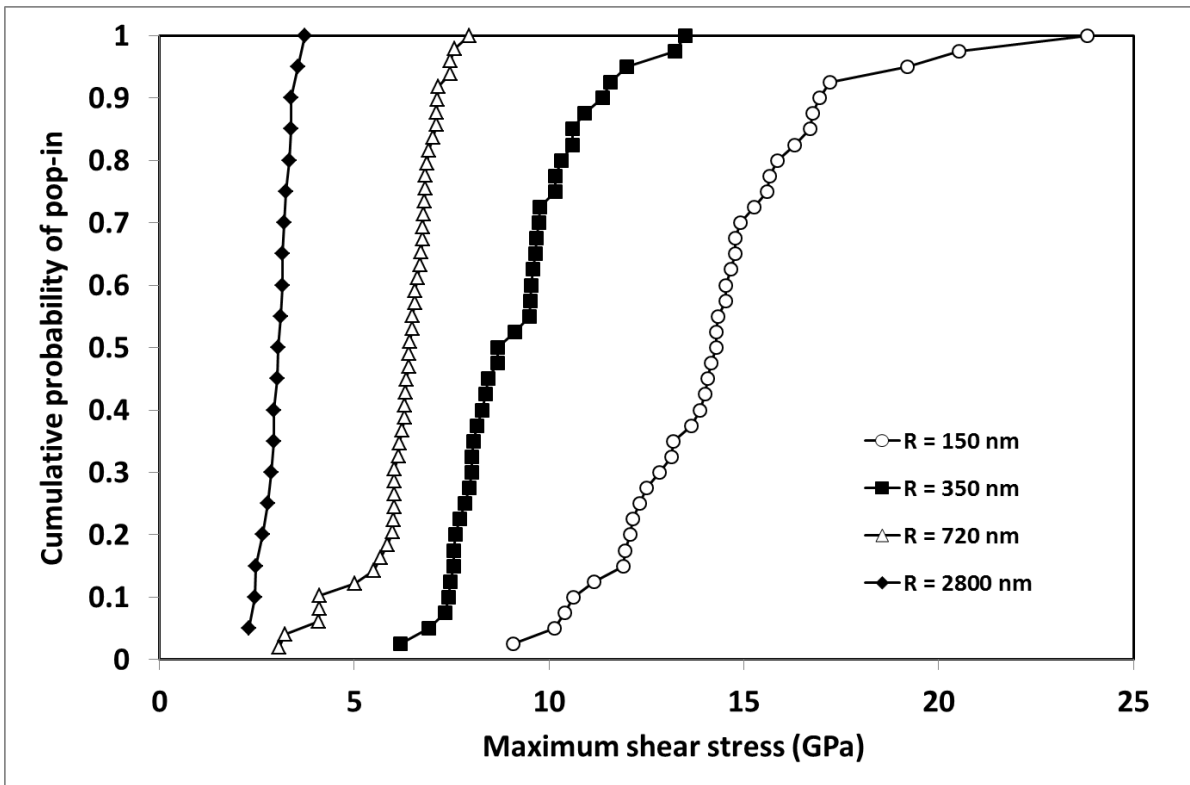


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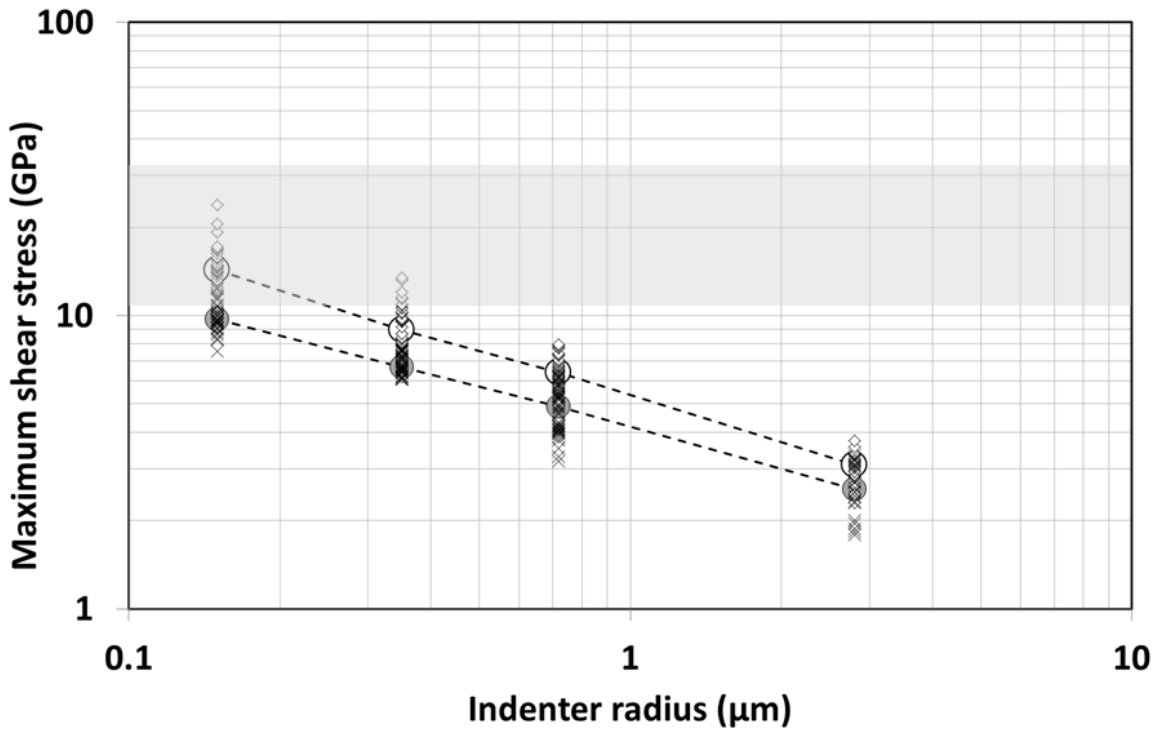
219 Figure 2(b)

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Figure 2(c)

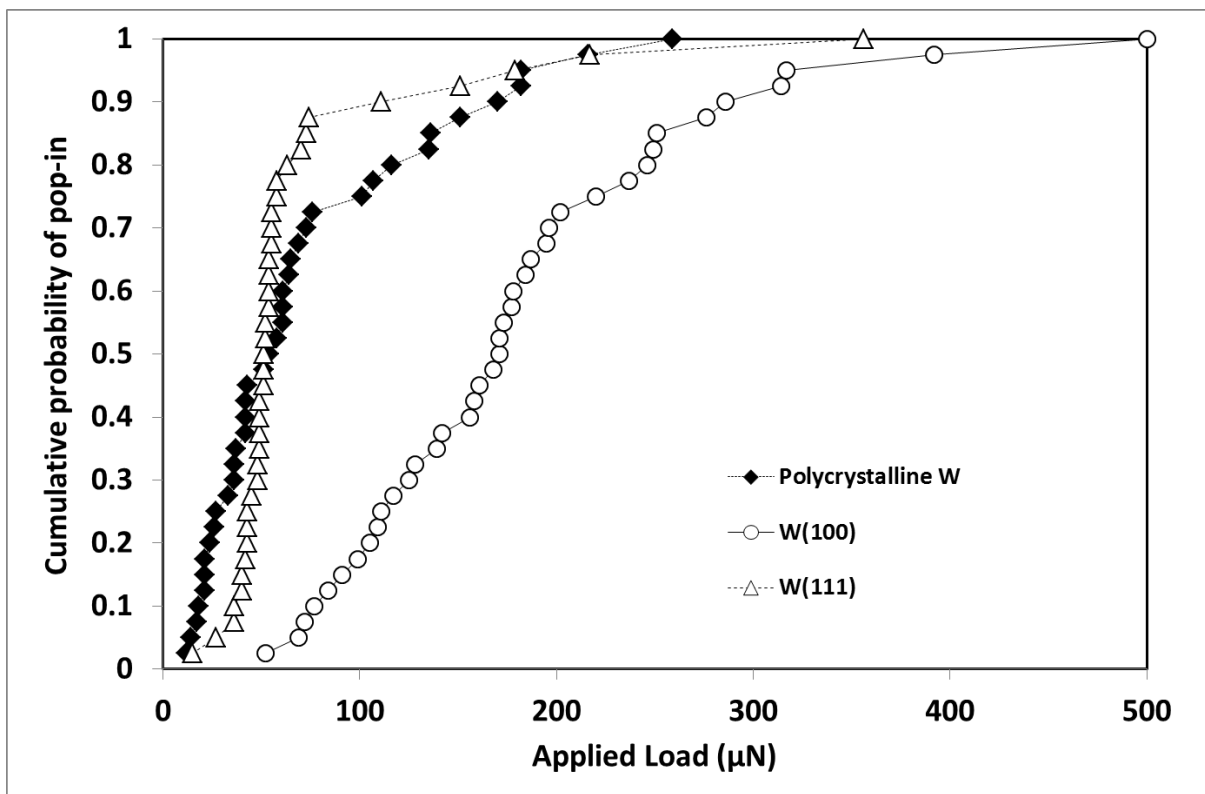


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Figure 2(d)

228 Figure 2: Indenter radius dependence of the pop-in behaviour with loading rate = 100 $\mu\text{N/s}$ (a)
229 critical load (b) mean pressure at pop-in (c) maximum shear stress (d) variation in maximum shear
230 stress with indenter radius for W(100) (diamonds) and W(111) (crosses). The median values at each
231 R are shown by the larger circles. The shaded region covers from $G/5$ to $G/30$ where G is shear
232 modulus.

233 The critical load for pop-in on W(100) varied with the indenter radius as shown in Figure 2(a).
 234 From Figure 2 in conjunction with Table 3, no evidence was found that would suggest the influence
 235 of loading rate on the load required for pop-in on any of the samples studied. The pop-in events
 236 were much less pronounced on the W(111) and polycrystalline tungsten samples, with the yield
 237 event being more commonly associated with a smaller displacement burst followed by further small
 238 periodic events as the load increased. The distribution of cumulative probability of pop-in for
 239 W(111) in tests at 25 $\mu\text{N/s}$ or 100 $\mu\text{N/s}$ was different to that for the other two samples as illustrated
 240 in Figure 3 for tests at 25 $\mu\text{N/s}$.



241
 242
 243 Figure 3: Cumulative probability plots of the critical load on W(100), W(111) and the polycrystalline
 244 W samples with the $R = 350$ nm tip at 25 $\mu\text{N/s}$.
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246 The sample with the (111) orientation showed a tighter distribution. After pop-in, the hardness from
 247 the unloading curve analysis was lower than the mean pressure in the contact area at pop-in.
 248 Analysis of post-yield unloading curves showed the W(111) and polycrystalline tungsten samples to
 249 have consistently higher hardness than the W(100) as summarised in Table 4 (a-d).
 250

251 **Table 4(a).Hardness and elastic modulus from nanoindentation to 500 μ N with the 150 nm**
 252 **end radius indenter**

	H (GPa)	E_r (GPa)	h_c (nm)
W(100) at 25 μ N/s	5.45 ± 0.25	322 ± 36	45.9 ± 1.4
W(100) at 100 μ N/s	5.99 ± 0.35	322 ± 41	43.2 ± 1.6
W(111) at 25 μ N/s	7.84 ± 0.72	345 ± 43	36.3 ± 2.2
W(111) at 100 μ N/s	7.52 ± 0.55	320 ± 36	37.2 ± 1.8

253 **Table 4(b).Hardness and elastic modulus from nanoindentation to 500 μ N with the 350 nm**
 254 **end radius indenter**

	H (GPa)	E_r (GPa)	h_c (nm)
W(100) at 25 μ N/s	6.91 ± 0.34	325 ± 39	31.0 ± 1.2
W(100) at 100 μ N/s	6.91 ± 0.41	321 ± 43	31.1 ± 1.4
W(111) at 25 μ N/s	8.73 ± 1.1	335 ± 47	26.1 ± 2.4
W(111) at 100 μ N/s	8.68 ± 0.86	323 ± 41	26.2 ± 1.9
Polycrystalline W at 25 μ N/s	9.63 ± 1.1	344 ± 45	24.2 ± 2.0
Polycrystalline W at 100 μ N/s	9.43 ± 0.96	338 ± 37	24.5 ± 1.9

256 **Table 4(c).Hardness and elastic modulus from nanoindentation to 1000 μ N with the 720 nm**
 257 **end radius indenter**

	H (GPa)	E_r (GPa)	h_c (nm)
W(100) at 25 μ N/s	5.98 ± 0.25	326 ± 24	40.6 ± 1.4
W(100) at 100 μ N/s	6.21 ± 0.27	319 ± 18	41.8 ± 1.4
W(111) at 25 μ N/s	7.28 ± 0.68	324 ± 21	35.9 ± 2.6
W(111) at 100 μ N/s	7.45 ± 0.55	324 ± 21	35.1 ± 2.0

259 **Table 4(d).Hardness and elastic modulus from nanoindentation to 3000 μ N with the 2800 nm**
 260 **end radius indenter**

	H (GPa)	E_r (GPa)	h_c (nm)
W(100)	4.84 ± 0.20	297 ± 24	35.6 ± 1.5
W(111)	5.84 ± 0.67	301 ± 39	29.8 ± 3.2

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 263
 264 The elastic moduli of all three samples were very similar, with the polycrystalline sample being
 265 typically ~ 3 % stiffer. The measurements at higher load confirmed the expected ISE upon hardness
 266 for all three samples but not depth dependence of their elastic properties. There was a linear
 267 relationship between H^2 and $1/h$ over the depth range of the 10-500 mN data so they were analysed
 268 with a Nix-Gao [18] plot using the formula $\frac{H}{H_0} = \sqrt{1 + \frac{h^*}{h}}$ to determine the characteristic length h^*
 269 and macroscopic hardness, H_0 , which are shown in Table 5.

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 273

274 **Table 5. Nix-Gao fitting parameters**

	H_0 (GPa)	h^* (nm)
W(100)	3.87	342
W(111)	4.79	463
Polycrystalline W	5.21	278

275

276 **4. Discussion**

277 Marked crystallographic and contact size effects on the incipient plasticity of tungsten were found
 278 in this study. Surface profilometry measurements and indentations to higher loads were used to
 279 provide relevant information regarding the surface roughness and conventional size effect upon
 280 hardness of the samples. Across the entire load range of the instrument used (0-500 mN), the
 281 unloading curve data showed constant elastic modulus, consistent with the literature value ($E_r = 321$
 282 GPa; $E = 411$ GPa) with the surface roughness increasing the variability at low indentation depth.
 283 Pile-up around the indentation due to cross-slip can increase the contact area leading to an over-
 284 estimate of the elastic modulus [19] with an increase of >10% reported for W(100) [20] and ion-
 285 irradiated polycrystalline tungsten [21]. In the current study, a much smaller effect was found in line
 286 with other recent results on polycrystalline tungsten [21]. The slightly lower modulus obtained from
 287 the measurements with the largest radius may also be an effect of the surface roughness together
 288 with a reduction in pile-up. Walter et al. [22] reported that the modulus of CrN films with $R_a = 2-10$
 289 nm was under-estimated by 5-14 % in simulations with a spherical indenter having an end radius of
 290 50 μ m.

291 All three samples showed a strong ISE upon hardness with the Nix-Gao plot revealing
 292 marked differences in their characteristic length and macroscopic hardness thereby implying a
 293 higher dislocation density at or near the surface of the W(111) sample. The rough surface model of
 294 Kim et al. [23] suggests that the characteristic length can be under-estimated in the standard Nix-
 295 Gao treatment unless roughness is taken into account. For the Ni surfaces, they tested and found
 296 that the under-estimations were around 70 nm for surfaces with $R_a = 3.2$ and 8.7 nm. If similar
 297 behaviour of tungsten samples used in this study is assumed, the characteristic length of the
 298 polycrystalline sample becomes very close to that of W(100), but its difference with that of W(111)

299 is increased.

300 Significantly higher critical loads for pop-in were found for W(100) than for W(111),
301 consistent with previous observations by Yao et al [1]. With an indenter of $R = 675$ nm, Yao et al [1]
302 reported critical loads on electropolished W single-crystals of the order of 7 mN and 2.5 mN for
303 W(100) and W(111) respectively, corresponding to median shear stresses at pop-in of around 21 and
304 14 GPa. On Ta, the increase in pop-in load on (100) compared to (110) and (111) has been ascribed
305 to differences in the stresses. FEA analysis showed that the high hydrostatic pressures in the
306 nanoindentation test aid nucleating defects (e.g. twins, stacking faults) [24], with more recent
307 support shown by MD simulations [25].

308 For W(100), the mean maximum shear stress determined with the $R = 150$ nm indenter was
309 around 15 GPa, with a maximum value of 23.8 GPa. The theoretical shear strength of crystalline
310 metals can be estimated by dividing the shear modulus by 2π , and is generally quoted to be in the
311 range of $G/5$ to $G/15$. As the shear modulus (G) of W is 161 GPa, the theoretical strength will be in
312 the range of 10.7 to 32.2 GPa, and is 25.6 GPa at $G/2\pi$. The limits at $G/5$ and $G/15$ are shown by
313 the shaded region in Figure 2(d). The data from use of the sharper indenters is consistent with the
314 pop-in occurring when the τ_{max} under the indenter approaches the theoretical strength, as has been
315 reported in previous studies on BCC[9], FCC metals [7] and BCC high-entropy alloys [10]. As
316 shown in Figure 2(d), values of τ_{max} were lower for the (111) orientation of W. Hertzian contact
317 mechanics assumes an ideally flat surface which is however not a practical reality. Although all the
318 tungsten samples were highly polished, Table 1 shows there were differences in surface roughness
319 with the W(111) and polycrystalline W being rougher than the W(100). With an increase in surface
320 roughness, the pressure on the surface of the asperities will be higher than that predicted by the
321 Hertzian treatment (which assumes an initially flat surface) so that although the apparent pressure at
322 pop-in is lower for rougher surfaces the real pressure may be significantly higher, as has been
323 shown in MD simulations of thin copper coatings [26].

324 In tests on annealed and electropolished tungsten with spherical diamonds with end radii of

325 1 and 13.5 μm , Pathak et al. [12, 14] observed higher stresses at pop-in with the sharper probe. Data
326 with the blunter probe was more stochastic in nature. Notwithstanding the fact that they tested an
327 electropolished surface, their data was in quite good agreement with the results for W(100) shown
328 in Figure 2 (b). Shim et al [13] provided a qualitative explanation for the radius dependence they
329 found in Ni(100) based on the average dislocation spacing and the stresses required to activate
330 existing dislocations (low stress) or to nucleate new ones in dislocation-free regions (higher stress).
331 Changing the indenter size changed the size of the highly stressed zone (which has been estimated
332 as $\sim 2.4a$ by Pathak et al. [12, 14]) relative to the average dislocation spacing. If the radius of the
333 indenter tip is much smaller than the spacing needed between dislocations for plasticity to occur,
334 then the applied stress needs to be sufficiently large to nucleate a dislocation. With larger tip radii,
335 the size of the indenter is much larger than the spacing between the dislocation and the stress
336 required to move pre-existing dislocations is lower. Wu et al. [17] recently developed a combined
337 statistical model for the radius dependence providing further evidence that incipient plasticity could
338 be triggered either by homogeneous nucleation of dislocations when a sharp indenter is used or by
339 the activation of existing dislocations when indenting with tips with larger end radii. The strength
340 drops more rapidly with increasing R due to the increasing possibility of encountering pre-existing
341 defects. The model does not consider surface roughness and it seems likely that this will also
342 contribute to the observed size effect. Knap and Ortiz used multiscale simulations to investigate tip-
343 radius effects during nanoindentation of Au(001) with 7 and 70 nm indenters [27]. In their
344 simulations, they found that the dislocation activity occurred before any deviation in the force curve
345 was observed. If a similar trend is also found for BCC metals and continues to larger indenter sizes,
346 then the maximum shear stress-radius dependence would be even larger than has been reported in
347 experimental studies to date.

348 In the experiments on tungsten performed in this work, there was no discernible rate
349 dependence over 25-200 $\mu\text{N/s}$ in either the stochastics of the pop-ins or the mean load value.
350 Although stress-based thermally activated dislocation nucleation is expected to result in the onset of

351 plasticity increasing with loading rate [28], the effect is slight in BCC metals compared to FCC
352 metals [9]. Biener et al. [9] reported a very small rate dependence on Ta(001) with RMS roughness
353 well under 1 nm, with the median value of the critical load for pop-in increasing by around 12%
354 over a x100 increase in loading rate from 50 $\mu\text{N/s}$ to 5000 $\mu\text{N/s}$. The absence of rate dependence in
355 this particular study over a much smaller load range appears to be due to a combination of the
356 intrinsic minimal rate sensitivity of tungsten (where creep strain during the 30 s hold period at peak
357 load in the higher load indentation tests is less than 0.015) and the higher surface roughness
358 (presumably local differences in roughness) of the samples.

359 Surface preparation is important as it influences the dislocation density and roughness of the
360 final surface [29]. Pathak et al. [12, 14] noted that rough mechanical polishing generally leaves a
361 disturbed surface layer with higher dislocation content which can be removed by electropolishing.
362 On another BCC metal, Mo(001), Wang et al. [30] reported that the highest pop-in critical load was
363 observed after electropolishing. Smaller loads were found after colloidal silica polishing, and
364 polishing by alumina produced defects sufficient to fully suppress pop-in. In a study on a FCC
365 metal, Al (111) by Minor et al. [7], the loading data was fitted to a plot of a Hertzian elastic
366 response. Although surface roughness was not mentioned, the presence of roughness could be
367 inferred by deviation of the experimental data from the elastic fitting by up to ~ 1 nm. Shibutani et
368 al. [8] studied the influence of surface roughness on the pop-ins observed when indenting Al(001)
369 with a tip of ~ 50 nm end radius, finding much lower critical loads for less highly polished surfaces.
370 In interfacial force microscopy on a passivated gold surface the critical load for pop-in was reported
371 to be 30-45 % lower near a step than in defect-free regions [31]. In a molecular dynamics study of
372 the influence of surface roughness on nanoindentation, it was reported that defects typically initiate
373 at the side of an asperity [26, 32].

374 The pop-in events were much less pronounced for the W(111) and polycrystalline tungsten
375 samples, with the yield event being more commonly associated with a smaller displacement burst
376 followed by further small periodic events as the load increased. In addition to the roughness effect

377 described above, this appears to be partially due to higher dislocation density in these samples
378 causing an increase in the hardness. Studies have also shown that higher pre-existing dislocation
379 density lowers the critical load for pop-in. In high-purity aluminium, a reduction in probability of
380 pop-in was observed when dislocation density increased [33]. In MgO indented with a 9.5 μm tip,
381 Montagne *et al.* [29] noted the contact was elastic up to a load of 300 mN when there were no pre-
382 existing dislocations but reduced nearly to zero for a pre-existing density of $1.2 \times 10^7 \text{ cm}^{-2}$. There
383 are differences in the distributions of cumulative probability of the critical load for pop-in between
384 the samples (Figure 3). The extent of dispersion in the first critical load on the FCC Al has been
385 reported to widen with a reduction in roughness [8]. Figure 3 shows that similar behaviour can be
386 seen in W(111). While the average surface roughness is higher on the polycrystalline sample, there
387 are smoother regions so that when indentations are made into these regions the data can more
388 closely approach that obtained from the W(100), but if measurements are made in rougher regions
389 the corresponding critical load is much lower. Yao *et al.* [1] reported a reduction in critical load for
390 pop-in on W after D-implantation and Biener *et al* [9] reported a complete suppression on Ta(001)
391 after ion energy ion bombardment. In studies such as these, it is not yet clear how much of the
392 reduction in pop-in is due to surface roughening and how much is due to higher pre-existing
393 dislocations in the near-surface layers of the tungsten. While they are to some extent interlinked,
394 further work on ion-irradiated samples may help to more fully deconvolute these effects.

395

396 **5. Conclusions**

397 The results being reported in this work confirm the statistical nature of incipient plasticity in the
398 nanoindentation response of tungsten over a wide range of conditions. Indenter radius (and
399 therefore contact size), surface roughness and crystallographic orientation were varied during the
400 experiments. The conclusions can be summarised as follows:

- 401 1. A strong size effect was observed, with the stress for incipient plasticity increasing as the
402 indenter radius was decreased. The maximum shear stress approached the theoretical shear

403 strength when W(100) was indented with the tip with smallest radius, whereas the (111)
404 orientation showed pop-ins at lower stress levels, which has been attributed to surface
405 roughness and greater dislocation density on the W(111) sample

- 406 2. Surface preparation plays an important role in the statistical nature of pop-in during loading
407 in nanoindentation tests. While they are to some extent interlinked, it was not clear whether
408 the roughening of the surface itself or the defect generation in the near surface layers caused
409 by it has the greater effect in reducing the load at which pop-in occurs.

410

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416

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