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21 *In situ* Micro tensile testing of He⁺² ion irradiated and implanted single crystal
22 nickel film
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Abstract

The effect of ion irradiation on the tensile properties of pure Ni single crystals was investigated using an *in situ* micro-mechanical testing device inside a scanning electron microscope. A 12.8 μm -thick Ni film with {001} plane normal was irradiated with 6MeV He^{+2} ions to peak damage of 10 and 19 displacements per atom (dpa). Micro-tensile samples were fabricated from the specimens parallel to the plane of the film using a focused ion beam (FIB) instrument, and tested in tension along [100] direction, up to fracture. The peak strength increased from ~ 230 MPa for the unirradiated material to about 370 MPa and 500 MPa for the 10 dpa and 19 dpa samples respectively, while the ductility decreased with increasing dose. The surface near the peak damage regions fractured in a brittle manner, while the regions with smaller dose underwent significant plastic deformation. Slip bands extended to the peak-damage zone in the sample with a dose of 19 dpa, but did not propagate further. Transmission electron microscopy confirmed the stopping of the slip bands at the peak-damage region, just before the high He concentration region with cavities. By removing the peak damage region and the He bubble region with FIB, it was possible to attain propagation of slip bands through the entire remaining thickness of the sample. This material removal also made it possible to calculate the irradiation hardening in the region with peak hardness – thus enabling the separation of hardening effects in the high and low damage regions.

Introduction

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4 It is well known that radiation damage can cause changes in microstructure and consequent
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6 increase in strength and decrease in ductility, thus reducing the service life of structural parts in
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8 nuclear power reactors [1]. The irradiation of metals with neutrons or ions can cause the
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10 formation of vacancy and interstitial loops, defect clusters and cavities. (Cavities include faceted
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12 regions which do not have matrix atoms and have zero or low gas pressure called “voids”, and
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14 regions which do not have matrix atoms but are fully pressurised with gas atoms such as He to
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16 form a spherical shape, these being called “bubbles”). The effect of such microstructural changes
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18 caused by irradiation on the mechanical properties of structural materials has been the subject of
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20 intense study in the past few decades. Such studies have been integral to the worldwide effort to
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22 produce materials with greater resistance to higher irradiation doses for future designs of nuclear
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24 reactors [2, 3]. Ion beam irradiation has been a method of choice to simulate the effects of
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26 neutron irradiation in a reactor for some time [4, 5], since it enables the attainment of reasonable
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28 doses within hours, instead of years inside a reactor. One of the drawbacks of using this method
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30 is that the damaged region is very shallow (generally $<20\text{ }\mu\text{m}$), and mechanical testing of such
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32 thin layers is extremely difficult. This problem has been solved partially by using
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34 nanoindentation to probe the hardness changes of such thin irradiated materials [6-10]. However,
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36 the results of nanoindentation tests are difficult to interpret due to two reasons: (a) it is a test with
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38 a tri-axial stress state, hence deriving uniaxial tensile properties out of such tests is difficult, and
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40 (b) since the process creates a 3-dimensional plastic zone around it that grows with the depth of
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42 the indent in a manner which is material dependent, the hardness values measured by
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44 nanoindentation are an average of a somewhat uncertain volume of material around the indent
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46 [11, 12]. Micro compression testing [13-15], which has a uniaxial stress state, has been
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48 conducted in the past on ion beam irradiated materials [6, 16, 17]. While yield stress and work
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4 hardening rate are accessible, compression testing comes with its own set of difficulties. Since
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6 the failure modes in compression are considerably different from the necking and subsequent
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8 fracture in tension, detecting a precise point of failure and measuring total compression is
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10 difficult, as the sample does not rupture, but is compressed with accompanying sliding and
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12 barrelling effects [16, 18-20]. Tensile testing of thin regions of irradiated material can obviate
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14 these difficulties, and thus is a preferred method for studying the strengthening effects. However,
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16 until recently, there have been great technical challenges in the manufacturing of sensitive testing
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18 equipment and the fabrication of test samples. Only a limited amount of tensile tests have been
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20 performed in the past, such as those conducted on unirradiated material by Kiener et al. [21],
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22 Bhattacharyya et al. [22] and Wheeler et al. [23], and those conducted on electroplated and
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24 therefore FIB damage free tensile test specimens by Landau et al. [24]. These papers typically
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26 discuss either the sub micrometre scale in the irradiated condition, or the meso-scale dimension
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28 regime (i.e. a range of a few μm to tens of μm) in the un-irradiated condition. The meso-scale
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30 regime has not been evaluated in tension in the irradiated condition, especially on materials for
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32 engineering applications. The technical problems in applying tension at the mesoscale on
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34 irradiated materials have been overcome to a large extent in the past few years, and significant
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36 advances have been made in both the design and manufacture of in-situ micro-mechanical testing
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38 equipment and the fabrication of micro-tensile samples [23]. In this paper, the authors have
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40 attempted to understand the mechanical response of a thin foil of single crystal Ni before and
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42 after irradiation by tensile testing of FIB fabricated samples in which the irradiated region is
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44 parallel to the gauge length. The methods adopted to perform these tests enable the simultaneous
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46 achievement of multiple experimental goals, viz. (a) Real-time visual observation and recording
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48 of the deformation behaviour in the high- and low-dose regions of the sample, (b) real-time
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4 recording of the stress-strain curves and their association with particular events as observed, such
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6 as load drops and strain jumps, (c) calculation of the radiation induced strengthening effects in
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8 different regions of the radiated/ ion implanted material having different doses, and (d) post-test
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10 transmission electron microscopy examination for gaining insight into the slip propagation
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12 process. These methods show, for the first time, the distinctive effect of bubbles or voids
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14 (henceforward collectively called cavities, except where one or the other type is meant) in the
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16 obstruction to slip, resulting in remarkably different deformation behaviour with dosage. These
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18 methods together provide significant insights into the mechanical behaviour of ion irradiated
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20 material which will enable us to extend these results to the behaviour of bulk neutron irradiated
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22 materials in the future.
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31 **Methods**

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33 The tensile specimens tested in this study were manufactured in single crystal Ni foils (obtained
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35 from Princeton Scientific, PA, USA). The thickness of the foils was about 12.8 μm . The foils
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37 were of 99.999% purity or greater. The normal direction of the foils was <001> which is
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39 identical to the ion beam irradiation direction. The tensile axis was roughly parallel to the <100>
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41 direction, within $\sim \pm 2-3^\circ$. The ion beam irradiation of the Ni foils was performed using the 2MV
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43 Tandetron STAR accelerator at the Australian Nuclear Science and Technology Organisation
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45 (ANSTO, Lucas Heights, NSW, Australia). One foil was preserved in the as-received condition,
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47 while two other foils were irradiated with 6 MeV He^{+2} ions to different doses, 2×10^{17} ions/ cm^{-2}
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49 and 3.8×10^{17} ions/ cm^{-2} . The irradiation temperature was approximately $25^\circ\text{C} \pm 5-10^\circ\text{C}$ as
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51 monitored with a thermocouple mounted close to the foil on the aluminium sample clamp. SRIM
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53 calculations of the damage profiles [25] (Figure 1) predict an average displacement damage of
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0.2 dpa and 0.4 dpa through the depth of the two irradiated samples up to about 9.4 microns, with much higher damage peaks (up to 10 dpa and 19 dpa respectively) at the ion stopping point at ~11.85 μm depth. We define the “entry surface” for the ions as the “front surface”, and the other, “exit surface” for the ions as the “back surface” for the sake of consistency. SRIM calculations show that very few, if any, ions actually exit at the back surface, since the sample thickness is greater than the maximum range of the 6 MeV He^{+2} ions used. Since the average thickness of the irradiated foils as measured with an SEM was ~12.8 μm , the peak damage stopping point region was predicted to be located within the sample volume. Thus a highly damaged region was formed about a micron inside the “back surface” of the ions. This situation is schematically depicted in Figure 1, where the region with damage greater than approximately half the peak damage is shown shaded in grey. This region extends from a depth of about 11.6 μm to 12.1 μm . The SRIM calculations show that the peak He concentration occurs at a depth of ~12.0 μm , and is ~5% for the sample with fluence of 2×10^{17} ions / cm^2 , and ~9.5% for that with 3.8×10^{17} ions/ cm^2 . This is plotted in Fig. 1 along with the damage profiles. It is clear that although the peaks of the damage profile are shallower than the He concentration profile by only about 150 nm, the damage profile starts to rise substantially before reaching the peak,

Tensile specimens of the geometry shown in Figure 2(a) were fabricated in each sample using a Zeiss® Auriga 60TM focused ion beam (FIB). The gauge length was 25 to 30 μm and the cross sectional area was approximately 10 μm (width) x 13.0 $\mu\text{m} \pm 0.2 \mu\text{m}$ (thickness). A grip in the shape of a rectangular hole was milled at the free end of each specimen to provide a structure on which a tensile load could be applied via a hook type gripper (Supplementary Figure 1). Surface imperfections and curtaining, as well as tapering of the specimen, was minimized by performing

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4 a low-current final polishing step at 30 kV, 240 pA. For some specimens, a shallow pattern of
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6 fiducial marks was lightly milled at 30 kV, 50 pA on the top surface to allow post-test high
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8 resolution digital image correlation on the movies.
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14 The *in situ* tensile tests were conducted in a Zeiss® Ultra Plus™ SEM using a micro-test rig
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16 (MTR-3) developed by MicroTesting Solutions® (Hilliard, OH, USA) at the Institute of
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18 Materials Engineering in ANSTO. The samples were subjected to elongation by carefully raising
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20 a flat-faced silicon loading pin into the tensile grip, and applying a tensile load parallel to the
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22 specimen axis. Alignment between the loading direction and the sample axis was adjusted prior
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24 to testing to better than 0.5° in plane and 2° out of plane using a pair of motor-driven screws on
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26 either side of the sample stage. The displacement rate was 20 nm/s, corresponding to a strain
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28 rate of $\sim 8 \times 10^{-4} \text{ s}^{-1}$. High definition SEM images (2048 x 1535 pixels) were acquired at small
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30 strain intervals during the tests. The displacement or strain was held constant during the image
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32 acquisition by pausing the test. The pause at constant strain was accompanied by a small stress
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34 relaxation ranging from 0.1 to 1 MPa. For each image, the load recorded at the start of
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36 acquisition was used to calculate the engineering stress. A Matlab digital image correlation code
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38 was then used to calculate the engineering strain using the displacement of fiducial markers
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40 located on either side of the tensile specimen gauge length [13, 22].
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50 **Results:**

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53 Conducting the tensile tests *in situ* in an SEM allows real-time imaging for a detailed comparison
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55 of the deformation behaviour of the unirradiated versus irradiated samples. The SEM image in
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57 Figure 2(a) shows a tensile test specimen before testing, while Figure 2(b) shows the final post-
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4 tension state of one of the two unirradiated specimens. A movie made from successive SEM
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6 images taken during the *in-situ* tensile test of the unirradiated specimen is given in
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8 Supplementary Video 1. These specimens had an average ultimate tensile strength (UTS) of
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10 ~233 MPa and exhibited ductile behaviour with significant necking enabled by the formation of
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12 alternating slip steps on multiple slip systems (Figure 2b, Supplementary Video 1). The
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14 corresponding engineering stress-strain curve is shown marked as “A” in Figure 5. (Here, each
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16 data point is obtained from the stress and displacement value recorded at points where the
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18 images were taken. The data collection rate for these curves was limited by the image acquisition
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20 rate to some extent, because image acquisition could take up to 22 sec.) There was a clear
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22 deviation from linear elasticity at a strain of ~0.4% and a stress of about 123 MPa. As is evident
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24 from this plot, the specimen underwent significant work hardening after yield, and subsequently
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26 deformed in a rather ductile manner. The maximum stress of 241 MPa is reached at 10.1% strain.
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28 Past the maximum stress, the stress drops, with occasional rise and fall due to initiation and
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30 passage of slip steps, respectively. There was no visible rupture of the surface even up to a strain
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32 of ~56%.

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34 The SEM image in Fig. 3(a) shows the final post-test state of a specimen radiated to a total
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36 fluence of 2×10^{17} ions/cm². The movie made from this test is presented in Supplementary Video
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38 2. The sample was irradiated from the top and the peak damage region is near the bottom surface
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40 in this case. The corresponding engineering stress- strain curve is shown marked “B” in Figure
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42 5. After the first major slip event at peak stress, the stress drops with a sudden jump in strain, and
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44 then rises again by more than 20 MPa. This process keeps repeating itself with decreasing stress
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46 jumps each time, until about 26% strain when the sample was unloaded. More significant load
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4 drops were observed following each new slip event in this sample than in the unirradiated case.
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6 Irradiation hardening also resulted in a higher UTS of ~ 371 MPa in these specimens. As can be
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8 seen from Figure 3(a), there is a sign of rupture at the back (or exit) surface, which is close to the
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10 damage peak region. This is confirmed by the image in Figure 3(b), which shows the “exit
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12 surface” of the sample, which has fractured almost along straight lines. There are clear slip lines
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14 going from one surface to the other. The TEM image of a FIB-ed cross-sectional sample taken
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16 from the specimen after the test, shown in Fig. 3(c), clearly demonstrates that the slip lines
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18 indeed reach very close to the “exit surface”. Schematic dose and He content profiles are
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overlayed to indicate regions of interest. The sample exhibits a region of high damage as a darker layer near the exit surface, and a region of high He implantation with a large number of spherical nano-scale cavities, most likely bubbles (~1-3 nm diameter) as seen in the higher magnification image in Supplementary Figure 2. Thus the slip lines penetrate the region of peak damage and He⁺² ion implantation. Although the first rupture at the back surface appears at a strain of about 1.7 %, the sample does not fracture completely even at a strain of ~26%. The cavities appear in a layer which ends at about 15 nm from the exit surface. This does not exactly match the SRIM calculations, which suggest that the stopping range should be ~200-250 nm short of the surface. This discrepancy is possibly due to inaccuracies in the stopping power used in SRIM, and grain orientation effects such as channelling which are neglected in SRIM calculations, and also local thickness variations in the film.

More dramatic changes in deformation behaviour were observed in the case of the highest dose (3.8 x 10¹⁷ ions/cm² irradiated) specimens, which had an UTS of ~500 MPa (curve marked “C” in Figure 5). At that stress, the “exit surface” of the specimen ruptured in a brittle manner, at a

1 strain of about 2.2 %. This is shown in the SEM image in Figure 4(a) and Supplementary Video
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7 3, where the back surface is at the top, as indicated. Due to the fast pace of the process, it was
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9 difficult to capture images at intermediate strains, and only two images were captured after the
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11 first surface rupture event before total sample failure. There was no further stress increase with
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13 strain jumps, unlike in the other two cases. The whole specimen fractured completely (i.e. the
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15 two ends separated from each other) at a strain of about 24%, as calculated from the images
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17 taken from another tensile sample from the same specimen. The image in Figure 4(a) and the
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19 video also show the formation of slip bands which extend from the front surface to a plane which
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21 is about a micron from the back surface. The fracture surface of these specimens, as shown in
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23 Figure 4(b), also displays abrupt intersecting slip steps that terminate at the approximate stopping
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25 point of the ion irradiation in the specimen, marked by a dashed line. A TEM image of a cross-
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27 sectional FIB sample taken from this specimen after testing is shown in Fig. 4(c), which shows a
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29 bright band of cavities, and a darker band to its right (shallower depth), where the high
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31 displacement damage region exists. These regions are indicated with brackets and approximate
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33 overlaid dose and He content profiles. The darker band at the depth of the high damage region
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35 is about 150-300 nm shallower than the bright band of He cavities, in agreement with the SRIM
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37 calculations. In this image, a slip band extends diagonally at an angle of $\sim 43.5^\circ$ from the surface,
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39 which is parallel to $\{100\}$. This suggests that the slip band lies in a (111) plane, since its trace is
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41 close to 45° from the (001) plane on a (010) plane. This is expected because the most common
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43 dislocations in FCC Ni are of the type $\frac{1}{2}[110]$ in the $\{\bar{1}11\}$ planes. The slip band is so densely
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45 packed with dislocations that it is almost impossible to distinguish individual dislocations inside
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47 it. This micrograph reveals clearly the stopping of the slip band just before the bright region with
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49 maximum He concentration, and inside the region with maximum damage (darker band to the
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4 right of the bright band). The slip band already gets lighter in contrast in the dark peak damage
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6 region, indicating that the number of dislocations passing through this region is smaller than that
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8 approaching it. Higher magnification TEM images show the bright region, which had a
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10 maximum of ~9.5% He according to SRIM calculations, had faceted cavities with a maximum
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12 size of ~30 nm (Figure 4(d)). These facets were parallel to at least two sets of planes, {200} and
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14 {220}.
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21 In order to evaluate the effect of the hardened region alone (which includes the stopping peak) on
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23 the mechanical behaviour of the irradiated sample, a tensile specimen of the same dose ($3.8 \times$
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25 $10^{17} / \text{cm}^2$) was prepared, using FIB milling to remove the high-damage region (a thickness of
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27 about 3.7 μm) from the top of the specimen as shown in the post-test image in Figure 5. The aim
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29 was to remove a thickness which would contain the region with the damage peak, however, due
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31 to difficulties in milling at grazing angle, a slightly greater thickness was removed. A tensile test
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33 of this specimen gave an intermediate UTS of ~300 MPa, which was greater than that of the
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35 unirradiated sample, but lower than even the sample with a fluence of $2 \times 10^{17} \text{ ions/cm}^2$, as
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37 evident from the curve marked “D” in Figure 5. Plastic deformation occurred via abrupt, non-
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39 uniform slip steps that penetrated the full depth of the specimen. These slip steps were also
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41 accompanied by significantly larger load drops than those observed in unirradiated and 2×10^{17}
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43 cm^{-2} full-thickness irradiated samples. The first slip step to pass through the sample completely
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45 did so at a strain of ~0.7%, much smaller than the strain of ~5% at which this occurred for the
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47 unirradiated sample. This sample did not show any signs of rupture at the surfaces even up to a
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49 strain of 11%, after which the test was discontinued.
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A summary of results for all tests is presented in Table 1. As is evident from the results of the tensile tests, there is significant hardening of the samples due to irradiation, as expected. Tests on samples with two different amounts of fluence - 2×10^{17} ions/cm² and 3.8×10^{17} ions/cm² showed overall increase in UTS of ~135 MPa and 270 MPa respectively.

Since the material in the films had experienced vastly varying extent of damage depending on the depth, these values are averages over **regions** which have very different strengths. For obtaining a better estimate of the relationship between damage dose and strength, the sample with a fluence of 3.8×10^{17} was treated as a bilayer composite – with one layer up to a depth of ~9.4 μm, having an average dose of about 0.4 dpa (marked layer 1 in Figure 1), and the second layer (marked Layer 2 in Figure 1) extending from a depth of 9.4 μm to ~13.1 μm (the thickness of the particular sample from which the **high damage region** was removed) with an average dose of ~4 dpa. Then the overall average strength of the full thickness of the sample, σ_{Av} can be expressed as:

$$\sigma_{Av}(t_1 + t_2)w = \sigma_1 t_1 w + \sigma_2 t_2 w \quad (\text{Eq. 1})$$

where t_1 and t_2 are the thickness of the two layers, w the width of the sample, and σ_1 and σ_2 the strength of the two layers, respectively, at the strain where the peak stress occurs for the full thickness of the sample (i.e. the composite sample). This strain is about 2.2%. Using this approach it is assumed that the E-modulus of the material does not change due to irradiation. Thus, the strength of the layer with the peak damage, σ_2 , can be calculated as:

$$\sigma_2 = (\sigma_{Av}(t_1 + t_2) - \sigma_1 t_1)/t_2 \quad (\text{Eq. 2})$$

Using Eq. 2, and $t_1 = 9.4 \mu\text{m}$, $t_2 = (13.1 - 9.4) = 3.7 \mu\text{m}$, $\sigma_{Av} = 500 \text{ MPa}$, $\sigma_1 = 270 \text{ MPa}$ (since this is the approximate strength of the sample with the top **region** removed at the strain of ~2.2%),

we get a value of ~ 1084 MPa for σ_2 , i.e. for the average strength of the **region** with the peak damage.

When plotting the increase in strength at same strain against average displacement damage for each test along with this estimated value, a linear relationship between the two quantities is found (See Figure 7).

Discussion:

In this paper, an effort has been made to establish the effect of He^{+2} ion irradiation on the tensile properties of single crystal Ni using *in-situ* micro-tensile testing in an SEM. This is a significant step forward from the use of nano-indentation techniques, due to the relative simplicity of the stress-field, and hence greater ease of interpretation of the results in the tensile testing method. In addition the dimension of the specimen geometry covers the mesoscale range (**1-20 μm**) which has not been widely investigated.

From the results presented above, it is clear that there is significant increase in strength due to ion-irradiation, and that this increase is roughly proportional to the average damage in the material for the Ni single crystal used at RT (See Fig,7, where the average displacement damage varies from 0.4 to 4 dpa). The proportionality constant for the increase in strength ($\Delta\sigma$) for unit increase in average displacement damage is about 238 MPa / dpa. This linearity between $\Delta\sigma$ and damage is observed even if one uses peak damage instead of average damage over a certain **region**. This apparent linearity is, of course, expected to a certain extent, as the strengthening due to dislocations, loops and voids is almost linear in each case for relatively low doses at RT [2]. This is so because although the relationship between $\Delta\sigma$ and dose in dpa is supposed to be of the type

$$\sigma = \sigma_0 + k\sqrt{dose} \quad (\text{Eq. 3})$$

within a limited range of dose values, this equation can yield results close to a linear equation such as

$$\sigma = \sigma_0 + k'(dose) \quad (\text{Eq. 4})$$

as can be confirmed by fitting a line to a curve of Eq. 3 for small dose ranges. As an example, if the dose range is taken to be 0.4-4 dpa, a linear fit will give a value of R^2 equal to 0.982.

It is known that an approximately linear strength increase with dose occurs on most metallic materials up to ~10 dpa [1] and this has been correlated to defect density. In addition, the change in strength with dose as observed here is similar to what has been observed previously at lower dose regimes in polycrystalline 300 series stainless steel [10].

Moreover, no apparent saturation of hardening with increasing dose is observed in this series of experiments. One reason for this may be the absence of sinks for defect annihilation, as this is a single crystal with no grain boundaries.

There is a pronounced decrease in ductility in the **region** with the damage peaks in both irradiated samples, which is evident from the rupture in the “exit surface”. This decrease in ductility is more severe in the case of the sample with a fluence of 3.8×10^{17} ions/cm², as observed from the slip bands in the side face of the sample in Figure 4(a), where these extend up to a **region** about 1μm short of the exit surface. This observation is confirmed by the TEM image in Figure 4(c), where the slip band stops just before the bright **region** containing the He **cavities**. This implies that most dislocations in the slip band were stopped by this **region**, whose average strength was calculated to be ~1083 GPa, leading to dislocation pileup. No slip steps were observed in the He stopping peak region therefore it is assumed that the **region** underwent elastic

elongation until it ruptured under extremely high stresses. This is further confirmed by the stress-strain curve for the high dose sample which shows extremely high stress build-up without significant strain, followed by sudden fracture at the surface with a large drop in load and increase in strain. This also indicates that the **cavities** do not allow dislocations to move further on the slip systems and therefore the field of He **cavities** is a strong dislocation obstacle.

Classical defect free zones are not observed in the He implanted **region** or in the **region** with high displacement damage below it. This implies that materials with high He content stored in partially pressurized **cavities** can fail in a brittle manner but do not experience removal of defects in a localized fashion. For the He implanted **region** this is expected because the dislocations do not cross this region. For the damage peak region, it may be difficult to see the defect free zone especially if it is very narrow and the dislocations are packed in the slip plane with extremely high density. When the **region** with the peak damage was removed, this strong obstacle to dislocation motion no longer existed, and the slip bands could pass across the whole thickness of the remaining low damage **region** without much difficulty, and the sample surface did not undergo rupture (as seen in Fig. 4(d)). In that sample, once some slip lines crossed the whole sample thickness, more slip planes needed to be activated, which explains the repeated (increased) rise and fall of the stress with increasing strain. The sample with the intermediate fluence of 2×10^{17} ions / cm² also showed surface brittleness near the peak damage **region**, but not to the same extent as the one with 3.8×10^{17} ions / cm². This is also made clear by the cross-sectional TEM image from one such sample shown in Figure 3(c), where the slip band reaches much closer to the surface than in Figure 4(c). This indicates that even at a peak damage of ~10 dpa and a He content of about 5%, the barrier to dislocations is not strong enough to stop them from passing across.

It should be pointed out here that the sample with higher dose not only has almost twice the displacement damage due to the ions impacting the atoms of the Ni crystal, it also has about 10-15 times larger **cavities** than the sample with the lower dose, and about twice the number of He ions implanted in it. Thus the pressure inside the features is not enough to render them spherical, since the He concentration is only twice in the sample with the higher fluence. This explains the faceted shape of these features, and thus it is more appropriate to call them voids. Voids can act as strong barriers to dislocation motion, and the void bypassing stress approaches the Orowan stress for impenetrable obstacles under similar distribution of obstacles [26, 27]. Computer modelling by the same authors showed that for constant separation distance between voids, increase in void size and decrease in void distance resulted in increase in strength. The proposed equation for the Orowan stress for a row of obstacles with diameter d and surface to surface distance l is:

$$\tau(Orowan) = \frac{G \cdot b}{A \cdot l} \left[\frac{1}{\ln(d^{-1} + l^{-1})} + B \right]$$

where G = Shear modulus, b = Burgers vector, B is a constant which is about 1.52 for voids [28].

The value of A is 2π for edges and $2\pi(1-\nu)$ for screws.

Thus the hardening in the samples occurs due to two types of sources – displacement damage clusters and **cavities**. Therefore the sample with higher dose is indeed expected to have increased strength at and near the peak implantation **region**, since it has much larger voids, in addition to almost double the density of displacement defect clusters.

Apart from the strengthening and embrittlement effects, the other prominent phenomenon evident in the samples is the intermittent rise in stress, followed by a small stress drop and strain jump. As is well known, these small stress rise events occur due to the build-up of stress before

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2
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4 the initiation of a dislocation source. When a source is initiated and becomes active, more and
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6 more dislocations pass through the sample in that plane, and the strain increases rapidly. In the
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8 case of an unirradiated sample (Curve A in Fig. 6), when one set of dislocations from a given
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10 source passes completely through the sample, a new source of dislocations needs to be initiated
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12 and activated, thus requiring another stress rise. But this stress rise is not very high (~ 5 MPa)
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14 because the sample is devoid of irradiation defects, and allows dislocations to pass through easily
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16 once a dislocation sources is nucleated.
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21 In the case of the sample with peak damage of 10 dpa (fluence of $2 \times 10^{17} / \text{cm}^2$, curve B in Fig.
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23 6), the stress rises are higher than in the unirradiated sample (~ 18 -22 MPa max). This is likely
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25 due to the fact that once a set of dislocations passes completely through the sample, a relatively
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27 large amount of stress is required to pass another set of dislocations in a new plane, since there
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29 are a large number of obstacles in the way in the form of damage clusters. Once a few
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31 dislocations pass on a plane, it becomes easier for many more dislocations to pass through on the
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33 same plane, and there is a large strain jump. As all the dislocations formed in quick succession
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35 pass through the sample, the process of source initiation becomes necessary, and the cycle is
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37 repeated.
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43 In the case of the sample with a peak damage of 19 dpa (fluence of $3.8 \times 10^{17} / \text{cm}^2$, curve C in
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45 Fig. 6), these periodic stress rise events are not observed. There is a great increase in stress in the
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47 initial elastic period, and this is followed by rupture and a large stress drop (~ 130 MPa)
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49 accompanied by a strain jump of $\sim 13.8\%$ in one step, and a second stress drop (40 MPa) and a
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51 strain jump of $\sim 6\%$. This happens because as the stress increases at the beginning of the elastic
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53 process, any dislocation source that is activated creates a set of dislocations which can go up to
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55 the void region only, and no further at that stress. As the stress increases further, a new source of
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dislocations is activated, since the stress needed to activate a new set of dislocations would be less than that to push the first set through the barrier of the bubble/ void region. The new set of dislocations again reaches the void region and gets stopped. Thus one set of dislocations after another is initiated with increasing stress, and is barred from going further, causing the process to be repeated, until the stress becomes too high for the region at the top (the peak He concentration region) which had been undergoing mostly elastic deformation up until now, and it ruptures, causing the large drop in stress and strain jump.

When the top region including the peak damage and He concentration regions was removed (Curve D in Fig. 6), the phenomenon of the stress rise and drop and accompanying strain jump was evident again. This is most likely because the removal of the top region which was preventing the propagation of dislocations, allowed any given set of dislocations to completely pass through the sample (as is evident from the slip steps on the top surface in Fig. 5). Thus new sources of dislocations could be activated after one set of dislocations went out of the sample, with a stress increase which was lower than that required for doing the same in the original sample (A) with the top region present. But the stress increase required in this case was greater than in the case of sample B with a peak damage of 10 dpa. This is likely because the overall stress level in Sample D (top region removed) is much lower than in Sample B, but the average damage even in the low damage region in Sample D is greater (almost double) than in similar regions in Sample B.

Conclusions

- (a) In situ tensile testing of micro-tensile samples of pure Ni single crystal foils was conducted inside the SEM, before and after irradiation with energetic He^{+2} ions. The results showed

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4 increase in fracture stress roughly proportional to damage dose, as expected. The strain at
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6 which rupture occurred also decreased with irradiation.
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- 9 (b) The unirradiated material showed great ductility, and did not undergo fracture even at ~56%
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11 engineering strain. The peak strength was ~ 230 MPa.
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- 14 (c) The samples with a peak dose of 10 dpa showed an increase in strength to about 370 MPa,
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16 and showed first rupture at a strain of about 1.7%. They subsequently showed strain jumps
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18 with stress drops, followed alternately by stress rise due to activation of new channels of
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20 dislocation slip.
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- 23 (d) The material irradiated to a peak damage of 19 dpa ruptured at a stress of ~505 MPa, at
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25 about 2.2% strain and rapidly went to complete fracture without stress jumps, at about
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27 22.5% strain. These samples did not show any stress increases after the first rupture event,
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29 most likely because the stress increase needed to activate slip on a new plane would be
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31 higher than the stress required to continue slip on the planes already active.
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- 34 (e) Removal of the top region from the sample with a peak dose of 19 dpa confirmed that it was
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36 the peak damage region which was preventing the slip steps from passing through the whole
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38 sample. Thus although low doses (up to ~ 10 dpa) cause strengthening, only very high doses
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40 (> 10 dpa) and high He concentration (~9.5 At. %) can prevent the passage of slip bands so
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42 as to cause brittle fracture.
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- 45 (f) The increase in strength was found to be almost linear with respect to the dose, and this was
46
47 in agreement with previous studies. In single crystal nickel, the hardening/dose slope was
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49 found to be 230-240 MPa/ dpa.
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Tables

Fluence (ions/cm ²)	Sample No.	Peak / Max Damage (dpa)	Peak strength (UTS) (MPa)	Strain at peak strength (%)	Strain up to first rupture (%)
0	1	0	241	10.1	-
0	2	0	226	11.3	-
2.0×10^{17}	1	10	384	1.7	1.7
2.0×10^{17}	2	10	358*	1.5	1.7
3.8×10^{17}	1	19	503	2.2	>2.2
3.8×10^{17}	2	19	506	2.0	> 2.0
3.8×10^{17} (Peak damage removed)	3	0.97	300‡	0.7	-

* This is the first stress peak, after which the sample ruptures, but starts to strengthen again, and reaches a maximum stress of about 365 MPa at ~ 6.8% strain.

‡ This is the first stress peak, after which the sample ruptures, but starts to strengthen again, and reaches a maximum stress of about 316 MPa at ~ 11% strain.

Table 1: Summary of tensile test results for all irradiation conditions

Figure Captions

Figure 1: SRIM calculated damage and He concentration profiles for the two irradiated samples, showing peak damage of ~10 dpa and ~19 dpa for fluence of 2×10^{17} ions/cm² and 3.8×10^{17} ions/cm², respectively. The grey region in dashed outlines indicates the highly damaged region.

Figure 2: (a) Tensile sample before test, and (b) unirradiated tensile sample after test, showing ~56% plastic elongation.

Figure 3: (a) SEM image of post-tensile test condition of the sample irradiated with 6 MeV He⁺² ions to a fluence of 2×10^{17} ions/cm², (b) bottom surface of specimen after test, showing slip steps going through the surface and rupture in almost straight lines at the surface, and (c) FIB cross-sectional TEM sample showing the extension of the slip band to a point close to the back surface.

Figure 4: (a) Post-test image of sample with 3.8×10^{17} ions/cm² total fluence, showing almost brittle fracture at the “exit” surface (top surface in this image), and slip lines stopping some distance below the top surface, (b) fracture surface of another similar sample showing slip steps stopping about a micron short of the exit surface (top surface), the marked line showing the limit of propagation of the slip lines, (c) FIB cross-sectional TEM image (~parallel to $\langle \bar{1}01 \rangle$) showing slip line nearly 45° from the edge of the sample, which is $\sim \langle 100 \rangle$, and (d) post-test image of sample radiated to a fluence of 3.8×10^{17} ions/cm², but with the peak damage region removed from the back “exit” surface. The dose in removed region is indicated by the dotted line portion of the damage profile.

Figure 5: Engineering stress-strain curves of one sample from each irradiation condition.

Figure 1

Figure 2(a)

Figure 2(b)

Figure 3(a)

Figure 3(b)

Figure 3(c)

Figure 4(a)

Figure 4(b)

Figure 4(c)

Figure 4(d)

Figure 5

*Graphical Abstract