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Compressive Properties of $\langle 110 \rangle$ Cu Micro-Pillars after High-Dose Self-Ion Irradiation

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Single-crystal Cu micro-pillars were self-ion irradiated up to 190 displacements per atom, a level commensurate with damage expected after long exposure in a reactor environment. Compression experiments performed along the $\langle 110 \rangle$ to 10% strain were compared against un-irradiated Cu. Two specimen configurations were explored: large 10 μm tall and small 4 μm tall pillars. Compared to un-irradiated Cu, the small irradiated pillars exhibited a flow stress increase of more than 500 MPa and were able to attain peak stresses approaching 1 GPa. These results are discussed in the context of an end of range effect, a damage gradient effect, and size effects.

Keywords: Micro-Pillar Compression, Copper, Radiation Damage, Mechanical Properties

‘In service’ radiation damage experienced by space and nuclear power components can be safely and feasibly simulated in a laboratory environment through proton, neutron, or ion irradiation. These accelerated aging experiments are critical for predicting material performance as it can take decades to reach high levels of radiation damage under operational neutron fluences. For ion irradiation, the damage zone typically extends only a few microns into the material; however, miniaturized mechanical testing methods can probe small volumes enabling site-specific property measurement to elucidate the radiation damage effects.[1]

In terms of damage, the energetic species impinging upon the target material cause displacement damage by knocking atoms out from their regular lattice position. Depending on the size of the resulting cascade, the stacking fault energy of the system, and the temperature of the irradiation event, the resulting damage often relaxes into a multitude of defect structures, including stacking fault tetrahedra (SFT) and dislocations loops [2–6] that serve as obstacles to dislocation motion. Hardening from irradiation, as well as decreases in ductility are well known in metals.[7] Cu in particular is interesting as it has been characterized on a variety of length scales in the un-irradiated state,[8–12] as well as at damage levels under ~ 50 dpa.[13–16] This paper reports micro-compression

experiments of $\langle 110 \rangle$ oriented single-crystal Cu that has been self-ion irradiated with damage up to 190 dpa. Zinkle and Busby [17] and Zinkle and Was [18] outline that materials employed in future Gen IV energy systems will need to tolerate damage upwards of 200 dpa at high temperatures. This study’s purpose is to extend the prior work on ion irradiation of Cu by providing new data in the regime of relatively high radiation damage commensurate with that expected in Gen IV reactors.

This study was performed on a $\langle 110 \rangle$ oriented single-crystal boule of Cu. The crystal was self-ion irradiated parallel to a $\langle 110 \rangle$ direction with Cu^{+5} at 30 MeV with an ~ 200 nA broad beam to a dose of 3.86×10^{16} ions/ cm^2 at an average rate of 8.32×10^{12} ions/ cm^2 s. The irradiation was performed at room temperature with no active heating or cooling of the Cu. The temperature was not monitored during the irradiation. Using the detailed calculations with full damage cascades in Stopping Range of Ions in Matter (SRIM) simulations,[19] the radiation damage profile was estimated. Damage increased in a nonlinear fashion from about 10 dpa at the surface to a peak damage of approximately 190 dpa at a depth of 4 μm and then subsided with significantly less radiation effects estimated beyond 5 μm . An electron transparent foil of the irradiated Cu was prepared with the focused ion beam (FIB) and inspected with a Phillips CM30 trans-

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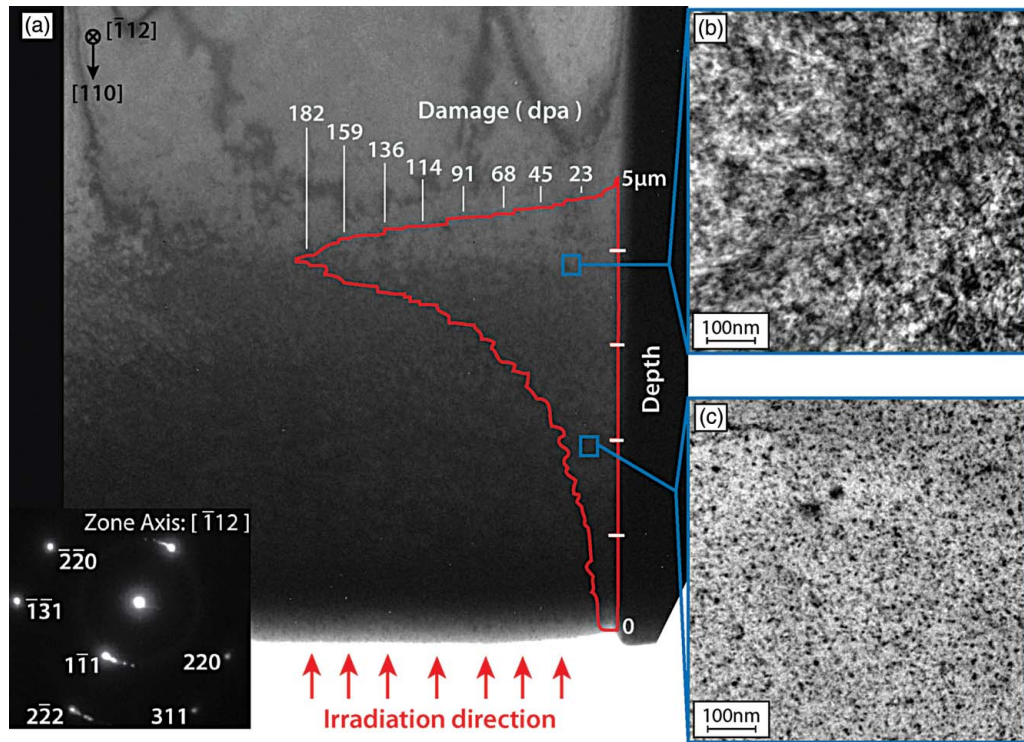


Figure 1. (a) Bright field TEM image of irradiated single-crystal Cu with overlay of the SRIM damage profile. The diffraction pattern was taken 90° from the $[110]$ irradiation direction giving a $[-112]$ zone axis. (b) Irradiated Cu near the peak damage site and (c) at $\sim 2 \mu\text{m}$ in from the free surface.

mission electron microscope (TEM) operating at 300 kV in standard bright field mode. Figure 1(a) shows the irradiated Cu with an overlay of the damage profile. As predicted, radiation defects, evident by the dark contrast, are highly clustered at the peak damage site (Figure 1(b)) and become less dense towards the surface (Figure 1(c)). As Cu has a low-stacking fault energy, the majority of the defects are expected to be SFTs with a small amount of dislocation loops present as well.[13]

Following the approach of Uchic and Dimiduk [20] un-tapered micro-compression pillars with a nominal 1:2 diameter to height aspect ratio were machined using the FIB. All milling was performed with an accelerating voltage of 30 kV for the Ga^+ ion beam. While 30 keV Ga^+ ions have been shown to penetrate 50 nm into Cu,[21] this damage is considered insignificant: rudimentary SRIM estimates suggest that 30 keV Ga^+ will create ~ 788 vacancies per ion strike and have an end of range of 10–15 nm, whereas 30 MeV Cu^{+5} is estimated to generate $\sim 2,505$ vacancies per ion strike and have an end of range greater than $4 \mu\text{m}$. Un-irradiated pillars were also manufactured on the same Cu boule, millimeters away from the well-identified local irradiation zone for direct side-by-side comparison of the irradiated and un-irradiated states. Two pillar geometries were machined for this study; a ‘large’ configuration with pillars $5 \mu\text{m}$ in diameter and $10 \mu\text{m}$ tall and a ‘small’ configuration having a $2 \mu\text{m}$ diameter and $4 \mu\text{m}$ height. Using a Hysitron Performech TriboIndenter operated in feedback displacement control,

the pillars were compressed 10% with a $25 \mu\text{m}$ diameter flat ended cone indenter along the irradiation direction (Figure 2(a)) at a strain rate of 0.025 s^{-1} . The compressive engineering stress–strain curves for both pillar sizes are shown in Figure 2(b) and 2(c). Stress and strain were calculated from the raw load and displacement data using initial diameter and height measurements from electron images of the as-machined pillars. All data were adjusted for elastic displacement of the base using the Sneddon [22] correction.

Both irradiated and un-irradiated pillars had intermittent flow with load drops which, under displacement feedback control, have been linked to bursts of dislocation activity [23] and the appearance of glide steps.[9] Experiments, discrete dislocation simulations, and statistical analysis [24–27] suggest that a combination of factors to include specimen size, initial mobile dislocation density, and the distribution of pinning points govern the stochasticity of source operation responsible for intermittent flow. As irradiation has been reported to frustrate dislocation motion,[28] it is reasoned that the subtle load drops for the small irradiated pillars stem from radiation defects obstructing dislocation glide thus preventing ‘bursting’ and easy escape at free surfaces which would presumably cause more substantial load drops.

There was only a slight distinction in the behavior for the large $10 \mu\text{m}$ tall, $5 \mu\text{m}$ diameter irradiated pillars compared to the same size un-irradiated pillars (Figure 2(a)). Consider the flow stress at 2% strain. For

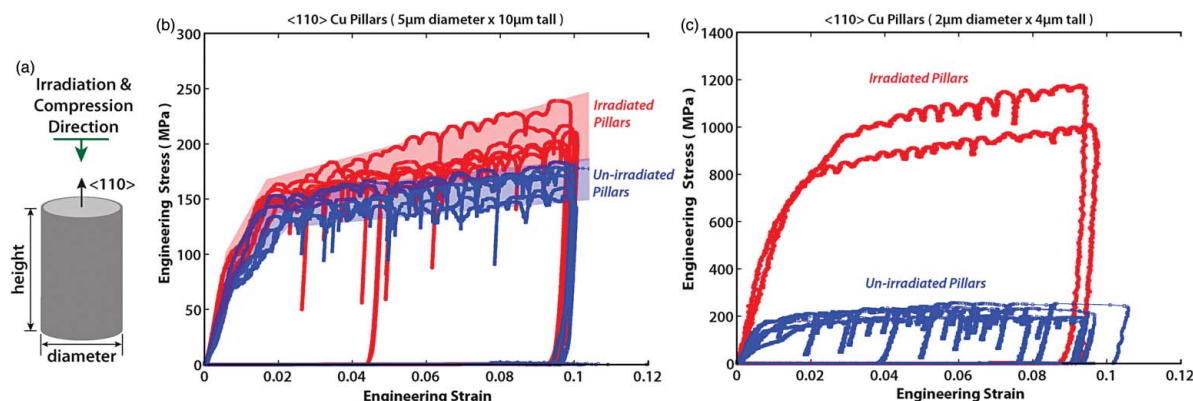


Figure 2. Compression stress-strain curves comparing irradiated and un-irradiated $\langle 110 \rangle$ Cu pillars. (a) Pillar schematic highlighting the crystallographic orientation of the Cu with respect to the irradiation and compression direction. Large pillar data presented in (b) with small pillar data presented in (c). Note the different scales of the y-axis in these two plots.

un-irradiated pillars, the flow stress at 2% strain was 122–153 MPa and the irradiated Cu had a slightly higher flow stress of 144–163 MPa. A small difference was also found to exist for the strain hardening rate, taken as the slope between the true stress—true strain data at 2% and 9% engineering strain. The hardening rate of the irradiated Cu was estimated to be 0.5 GPa, while the un-irradiated Cu was slightly lower at 0.3 GPa.

Unlike the large specimens, the small pillars that were only 4 μm tall with a 2 μm diameter exhibited a more substantial effect of radiation damage as evident in Figure 2(b). The flow stress at 2% strain for the un-irradiated small pillars was 159–211 MPa, while the irradiated pillars had flow levels more than three and a half times higher at 785–793 MPa. In terms of peak strength, the small irradiated pillars approach 1 GPa which is comparable to strengths observed for practically flaw free Cu micro-whiskers tested by Brenner [10,11] but weaker than the $\langle 110 \rangle$ Cu nano-whiskers investigated by Richter et al. [12] that reached theoretical strength levels.

In terms of strain hardening rate, the small 2 μm diameter un-irradiated Cu was similar to its larger counterpart at ~ 0.3 GPa but the estimate for the small irradiated Cu was much higher at 2.8 GPa. The hardening rate of un-irradiated single-crystal pillars is known to increase as sample diameter decreases from microns in size down to hundreds of nanometers.[29,30] The similar hardening behavior between the 4 and 2 μm diameter un-irradiated Cu may stem from the relatively small change in specimen size. As for the small irradiated pillars, the elevated hardening rate would appear to suggest that radiation damage not only suppresses initial dislocation source operation, but also renders source operation more difficult with increasing strain. However, it remains unclear if this hardening rate increase only manifests itself due to the combination of length scale effects and radiation damage. A study on much larger millimeter-scale irradiated polycrystalline tensile bars [15] found yield point increase but little change in hardening behavior compared

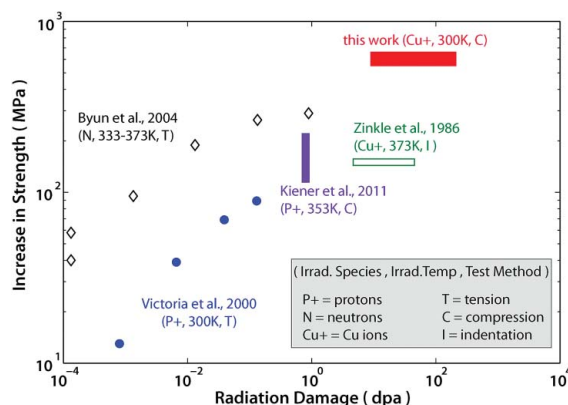


Figure 3. A compiled log-log plot comparing strength increase against various levels of radiation damage for Cu. Note that open symbols denote polycrystalline Cu, while solid markers represent single-crystal Cu.

to un-irradiated material. Additional studies are therefore needed to elucidate the individual impact size effects and radiation damage have on strain hardening.

Several studies have been published on the properties of single and polycrystalline Cu irradiated to damage levels under ~ 50 dpa at temperatures less than 200°C. Through tension,[13,15] compression,[14] and indentation,[16] these investigations all report strength increases for the irradiated material, however none exceed 300 MPa. The small 2 μm diameter irradiated pillars of this study had a larger strength enhancement with the radiation defects providing a 574–634 MPa increase in flow stress at 2% strain. Figure 3 is a log-log plot that assembles estimates of strength enhancement at yield or the 2% flow stress extracted from [13–16]. Comparisons from Figure 3 should not be taken as absolute as each of the aforementioned studies has differences in irradiation conditions (temperature, irradiation species, etc.). Nevertheless, it appears that higher levels of radiation damage generally corresponded to higher amounts of strengthening. This observation was slightly unexpected

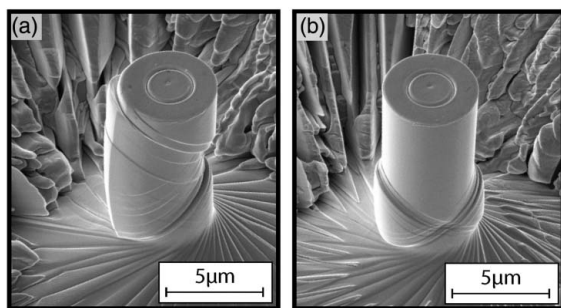


Figure 4. Comparison of large ($5\ \mu\text{m}$ diameter $\times$ $10\ \mu\text{m}$ tall) un-irradiated (a) and irradiated (b) Cu pillars compressed by 10%. Images were captured with the specimen at a 45° tilt.

as several have reported that the density of radiation defects saturates in Cu at ~ 1 dpa [13,31,32]. It is then speculated that irradiation to higher damage levels perhaps evolves the saturated defects into configurations that supply additional strengthening. There is limited experimental and simulation results [33,34] that suggest active dislocations can interact with radiation defects such as full and truncated SFTs in such a manner that a dynamic mechanical response is predicted. Follow up *in situ* TEM irradiation analysis is needed to characterize the radiation defect–defect interactions that form from extensive irradiation and understand how they may afford substantial strengthening.

There are three key issues that must be given consideration when interpreting these experimental results: (1) an end of range effect, (2) a damage gradient effect, and (3) a pillar size effect. The end of range effect manifests itself in the large pillar data. Since the irradiation damage only extended about $5\ \mu\text{m}$ into the pillar, a $10\ \mu\text{m}$ pillar height for the large pillars means that the top half of the pillar is defect hardened, while the bottom half is nominally undamaged. Figure 4 compares deformed images of the large pillars. Note that the irradiated pillar (Figure 4(b)) had no slip traces in the upper region. It appears then that the experiments on the large irradiated pillars effectively test only the short, nominally un-irradiated bottom half. This is corroborated by the fact that the large irradiated pillars exhibited substantial load drops (Figure 2(b)) suggesting that dislocation glide was not heavily obstructed by radiation defects. Nevertheless some strengthening in Figure 2(b) is noticeable and may stem from the elastic base and irradiation hardened top that confine deformation to a smaller volume and also act as barriers to dislocation motion. This may also explain the slightly higher hardening rate for the irradiated large pillars at $0.5\ \text{GPa}$, while $0.3\ \text{GPa}$ was measured for the un-irradiated Cu. In terms of a radiation effect, the only observation that can be drawn is that the flow stress at 2% strain for the irradiated Cu exceeds $\sim 122\ \text{MPa}$. This end of range effect reiterates the message of Hosemann et al. [35] and Kiener et al. [36] that it is important to

configure experiments to probe a well-known radiation affected zone.

The damage gradient effect refers to the fact that the irradiated Cu pillars have a gradient of defect agglomerations resulting from the radiation damage. For the small $4\ \mu\text{m}$ tall, $2\ \mu\text{m}$ diameter pillars, the experiments are not probing a structure with uniform damage but sampling over a range of damage levels from ~ 10 dpa at the surface to about 190 dpa at the pillar base. As roughly 50% of the pillar is at a damage level of less than 35 dpa, the inhomogeneity is not negligible. It is notable that the flow stress and hardening rate for the small pillars increased substantially raising the prospect that the strength might well exceed $1\ \text{GPa}$ for a Cu single crystal with a mean damage level of 190 dpa. To avoid the damage gradient effect, some have suggested fabricating specimens normal to the irradiation direction.[36] A challenge with such an approach is that the region of relatively uniform damage is limited and may require a reduction in pillar size which could introduce a size effect.

Length scales such as physical specimen size and defect spacing can influence the deformation mechanisms governing strength.[37–39] Issues can arise if the test specimen dimensions are so small that the volume probed is not representative of the irradiated material. This issue is highlighted in the nano-pillar compression experiments by Kiener et al.[14] For pillars with a diameter less than $\sim 400\ \text{nm}$, Kiener et al. [14] found no strength difference for un-irradiated and $0.8\ \text{dpa}$ (100) Cu. It was suggested that the pillar size caused the deformation in both the irradiated and un-irradiated Cu to be limited by same factor; the ability to activate a dislocation source in a limited volume. For pillar diameters above $400\ \text{nm}$, the strength of the $0.8\ \text{dpa}$ pillars remained constant as pillar diameter increased. The un-irradiated pillars on the other hand lost strength as size increased. It was suggested that this trend occurred because the deformation had become dominated by the interaction of dislocations with radiation defects. The small irradiated and un-irradiated pillars of this study do not have comparable strengths suggesting the $2\ \mu\text{m}$ diameter pillars are a representative irradiated volume with the strength enhancement coming from the radiation-induced defects.

In summary, Cu micro-pillars, self-ion irradiated with damage spanning from 10 to 190 dpa, were compressed 10% and compared against un-irradiated Cu. Both large $5\ \mu\text{m}$ diameter $10\ \mu\text{m}$ tall and small $2\ \mu\text{m}$ diameter $4\ \mu\text{m}$ tall pillars were investigated. Since radiation damage primarily penetrated to a depth of $5\ \mu\text{m}$, only the small $2\ \mu\text{m}$ diameter $4\ \mu\text{m}$ tall pillars provided data purely characteristic of a radiation damaged microstructure. This work extends previous observations on radiation damage to higher dpa levels and appears to suggest that strengthening continues to scale with irradiation damage, even at levels commensurate with very long-term reactor damage.

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