TRUE STRESS-STRAIN BEHAVIOR OF AS-IRRADIATED AND POST-IRRADIATION ANNEALED PURE COPPER - D. J. Edwards (Pacific Northwest National Laboratory)*, B. N. Singh and Palle Toft (Risø National Laboratory)

OBJECTIVE

Post-irradiation annealing has been shown to partially restore the work hardening ability of irradiated pure copper, but not to the levels of the original unirradiated material. True stress-strain behavior for the as-irradiated and post-irradiation annealed samples are compared to that of pure copper to further study the work hardening behavior.

SUMMARY

Post-irradiation annealing experiments were conducted on neutron-irradiated pure copper (from 0.01 to 0.3 dpa at 100°C) to explore the possibility of mitigating the effects of radiation hardening on the strength and ductility. The post-irradiation annealing (PIA) condition of 300°C for 50 hours yielded mixed results and did not completely remove the effects of irradiation. However, there were changes brought about in the microstructure that led to the removal of the yield point phenomenon and restored some of the unirradiated ductility and work hardening. The true stress-strain curves have been calculated for both the as-irradiated and the PI annealed Cu and compared to that of the unirradiated pure copper.

The true stresses at which the as-irradiated samples begin to neck (based on the engineering stress-strain curves) are similar for the 0.1 to 0.3 dpa samples and are similar to that of the unirradiated copper. On the other hand, the as-irradiated 0.01 dpa condition and all of the post-annealed conditions begin necking at lower stresses, but at similar strains. The work hardening rate in the as-irradiated copper (0.1 to 0.3 dpa), when compared on Kocks-Mecking plots, shows that after a few percent strain, the macroscopic work hardening behavior mimics that of the unirradiated pure copper in the latter stages of work hardening. The PI annealing does little to restore the elongation compared to the as-irradiated state, yet lowers the yield strength, removes the yield point phenomenon and causes the material to begin necking at a lower stress.

Further work is needed to understand the dislocation interactions (annihilation and storage of dislocations during plastic deformation, sweeping of the defects introduced during irradiation) that produce a situation where the PIA samples neck at similar strains but lower stresses than that of the as-irradiated. Compression testing is offered as a method to extend the mechanical properties testing to minimize the geometrical instabilities inherent in the tensile testing and explore the full range of work hardening up to failure.

PROGRESS AND STATUS

Introduction

Over the past 7 years a series of studies [1-7] have been conducted exploring the effects of neutron irradiation at different temperatures on pure copper and various copper alloys, most notably the precipitation strengthened CuCrZr and CuNiBe alloys and the ODS alloy GlidCop Cu-Al_2O_3. The irradiation experiments on pure copper included a study directed at using post-irradiation annealing to remove the effects of radiation hardening at 100°C and restore

* Pacific Northwest National Laboratory (PNNL) is operated for the U.S. Department of Energy by Battelle Memorial Institute under contract DE-AC06-76RLO-1830.

both ductility and work hardening to the copper. The results of this post-irradiation annealing experiment were recently reported elsewhere [7], and will only be summarized here for background.

Pure copper, when irradiated at 100°C, hardened such that the yield strength increased by a factor of 6-10 compared to unirradiated pure copper, with a saturation in the change in yield stress occurring at ~0.1 dpa. At doses of 0.01 dpa and higher, a sharply defined yield point or yield point phenomenon with a yield plateau appeared while the uniform strain to necking and total elongation decreased sharply. This radiation hardening and loss of ductility appear to be related to the fine-scale defects produced during irradiation in the form of small dislocations loops and small stacking fault tetrahedra (SFT). Raft-like agglomerates of loops also formed, yielding a microstructure that appeared to have a dislocation network at lower magnifications. Characterization of the deformed microstructure revealed that deformation in the as-irradiated copper samples occurred predominately by a localized deformation mode called dislocation channeling. This mode of deformation involved a finite volume of material (narrow bands) being swept clear of irradiation-produced defects by moving dislocations, with little deformation in the areas between the channels. The as-irradiated materials exhibited good reduction in area and failed in a ductile manner according to the fractography.

Post-irradiation annealing served to remove the upper-lower yield point and restore some measure of the uniform strain and work hardening, but not to the levels of the unirradiated copper. In fact, despite the change in the strength and work hardening, the strains at which necking and failure occurred were only slightly higher than that of the as-irradiated copper. The microstructural characterization revealed that annealing did not remove the defects produced by irradiation, instead it led to a ripening of the SFT defect microstructure and produced large prismatic loops thought to be interstitial in nature. Accompanying the prismatic loops was a dislocation network that presumably evolved from the agglomerated loops formed during irradiation. Annealing also led to the migration of grain and twin boundaries that effectively swept entire regions free of defects, producing soft regions with a low density of dislocations. Deformation in the PIA materials occurred by a mixture of ill-defined channeling and dislocation interactions throughout the bulk of the material. The failure modes were always ductile in nature, often exhibiting good reduction in area.

The following sections detail an analysis aimed at further investigating the work hardening and the overall macroscopic tensile response before and after post-irradiation annealing and comparing it to the unirradiated behavior.

Experimental

The material used in the present investigation was a thin (0.3 mm) sheet of oxygen-free high conductivity (OFHC) copper containing 10, 3, < 1 and < 1 ppm, respectively, of Ag, Si, Fe and Mg. The oxygen content of this copper was found to be 34 appm.

Tensile samples of OFHC copper were irradiated in the DR-3 reactor at Risø National Laboratory (see reference 1 for tensile geometry). Prior to irradiation, the OHFC copper samples were given a solution annealing treatment of 550°C for 2 hour in a vacuum of 10^{-6} torr. The resulting grain size and dislocation density were about 30 μm and ~10^{12} m^{-2}, respectively. The tensile specimens were irradiated at 100°C to different dose levels in the range 0.01 to 0.3 dpa (NRT). All specimens were irradiated with a displacement damage rate of ~5 x 10^{-8} dpa (NRT)/s. A subset of the irradiated specimens were given a post-irradiation annealing treatment of 300°C for 50 hours under vacuum (<10^{-5} torr). Unirradiated, as-irradiated and post-irradiation annealed specimens were tensile tested in an

Instron machine at a strain rate of 1.2 x 10^{-3} s^{-1}. Tensile tests were carried out at 100°C in vacuum (< 10^{-4} torr). The test temperature of 100°C was reached within 30 min.

Results

True Stress-Strain Curves

For the analysis of the work hardening behavior, the engineering stress-strain curves were converted to true stress-strain using the assumption of constancy of volume and homogeneity of strain along the gage length of the specimen up the point of necking [8]. The true stress σ and true strain ε are therefore calculated according to the following equations:

$$\sigma = s(e + 1) \quad \text{eq. 1}$$
$$\varepsilon = \ln(e + 1) \quad \text{eq. 2}$$

where **s** is the engineering stress and **e** the engineering strain [8]. These two equations are only valid to the point of necking, after which the actual dimensions of the specimens need to be measured to determine the true stress and strain. In this report, the true stress-strain curves are calculated only to the ultimate stress in the engineering stress-strain curves since the actual reduction in area was not measured once the material began necking. The true stress-strain curves presented in this report reflect the plastic strain only, that is, the machine compliance and Young's modulus have been removed from the curves.

The engineering and true stress-strain curves for the as-irradiated samples are provided in Figures 1 and 2. Both types of curves demonstrate that the as-irradiated samples exhibit a sharp yield point at 0.01 dpa which further evolves into an upper and lower yield point followed by a small yield plateau at 0.1 dpa and higher. In the engineering stress-strain curves the as-irradiated samples harden to much higher levels that the unirradiated material, that is, the yield and ultimate stresses are much higher. However, the true-stress strain curves give a somewhat different perspective in that the ultimate stress of the unirradiated copper matches that of the as-irradiated samples at 0.1 dpa and higher. The macroscopic strain at which the as-irradiated samples begin to neck is considerably lower than that of the unirradiated material. The 0.01 dpa sample necks at a much lower stress, but at a similar strain to that of the higher dose samples.

Post-irradiation annealing alters the tensile behavior as illustrated in Figures 3 and 4. The yield point behavior is clearly removed for all doses and some fraction of the work hardening restored. None of the conditions, however, were completely restored to the unirradiated state for this annealing condition. The true stress-strain curves shown in Figure 4 further demonstrate that annealing leads to little improvement in the strain to necking for those specimens irradiated to 0.1 dpa or higher. The lowest dose specimen did exhibit a significant improvement in the strain to necking. The true stresses at which the PI annealed samples begin to neck are almost identical to the true yield stresses in the as-irradiated samples.

Kocks-Mecking Plots ($\partial\sigma/\partial\varepsilon$ versus σ)

Plotting the work hardening rate $\partial\sigma/\partial\varepsilon$ against the true stress σ allows one to compare the work hardening behavior of the unirradiated pure copper, the as-irradiated and PI annealed conditions. These type of plots also show where the instability criterion $\partial\sigma/\partial\varepsilon = \sigma$ takes affect and causes the tensile specimen to begin necking in tension. The intersection of the individual curves with the straight line represents the stress beyond which the material has

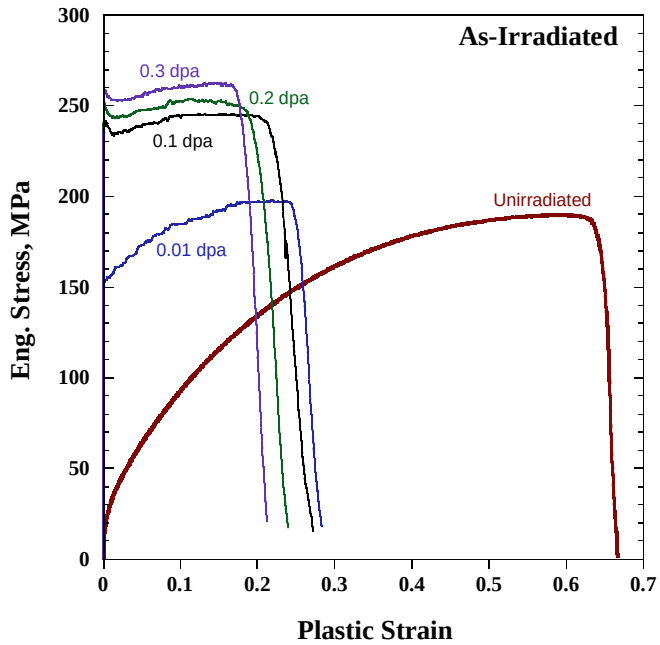

Figure 1. Engineering stress-strain curves for the as-irradiated pure copper and the unirradiated copper.

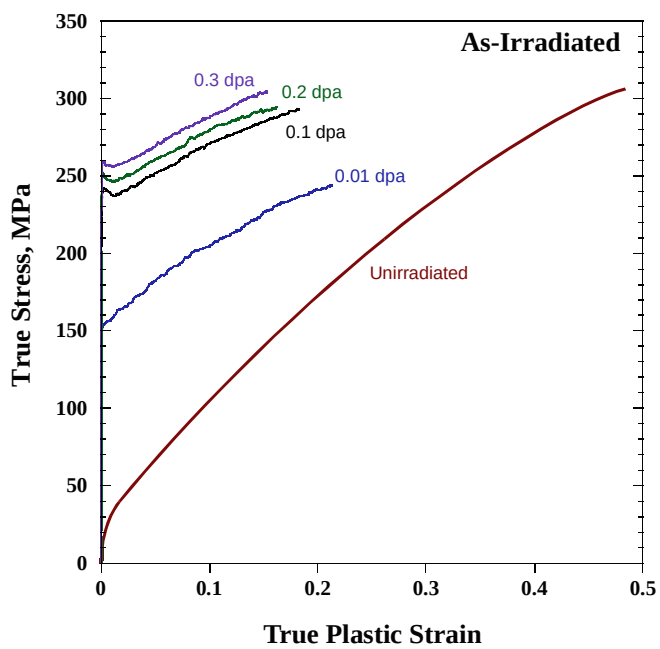

Figure 2. True stress-strain curves derived from the engineering stress-strain curves for the as-irradiated pure copper and the unirradiated copper. The tensile curves demonstrate the as-irradiated samples begin necking at stresses similar to that of the unirradiated copper when irradiated to 0.1 dpa or above.

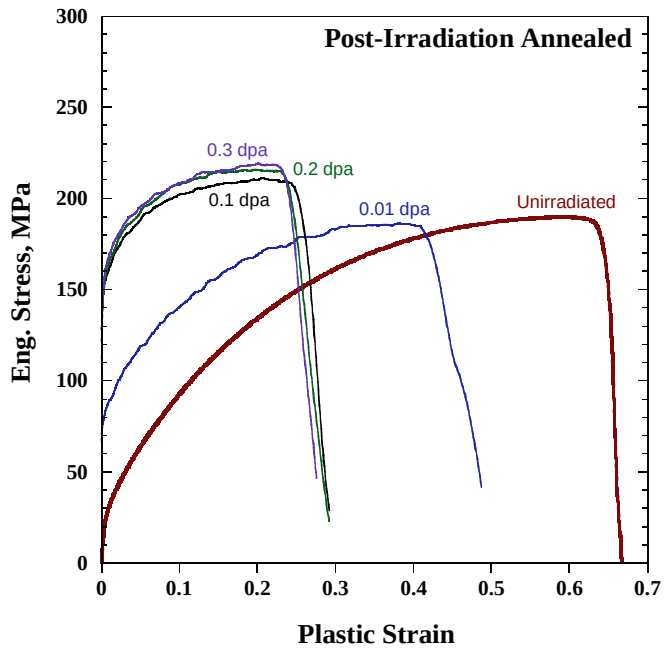

Figure 3. Engineering stress-strain curves for pure copper given a post-irradiation annealing treatment at 300°C for 50 hours.

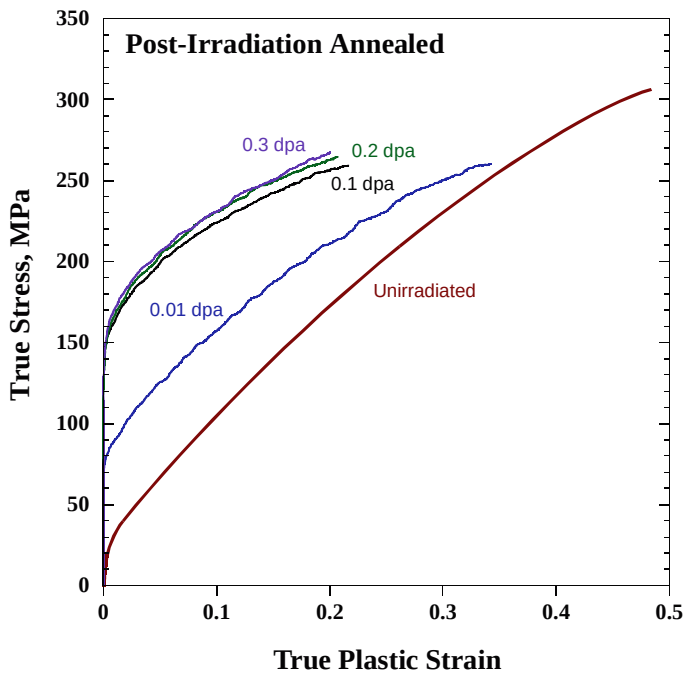

Figure 4. True stress-strain curves after post-irradiation annealing. Annealing lowers the yield stress and removes the yield point phenomenon. Strain to necking is similar to that of the as-irradiated samples, but the stress at which necking occurs is lower.

begun to neck (ultimate stress on the engineering curves). Figures 5 and 6 show these plots for the as-irradiated samples and the PI annealed samples as compared to the work hardening behavior of the unirradiated material.

In the case of the as-irradiated samples (Figure 5), it is clear that significant changes occur between 0.01 dpa and the higher doses. As the dose increases the stress at which necking occurs shifts upward to match that of the unirradiated material and the work hardening rate begins to mimic that of the unirradiated material after a few percent strain. While these curves were generated from a single tensile test result at each condition, and some scatter in the behavior should be expected, the trends are quite clear. The K-M plots show that a few percent strain allows the as-irradiated materials (0.1 dpa and above) to begin behaving macroscopically like unirradiated pure copper in the latter stages of work hardening.

The PI annealed materials (Figure 6) behave differently in that they converge to the same stress point on the instability criterion line, albeit lower than the unirradiated copper and the as-irradiated copper (≥0.1 dpa). The work hardening rate for each condition is different in the early stages of deformation, but eventually each converges to follow a common work hardening rate that changes little with dose. Interestingly enough, the work hardening rate in the 0.01 dpa, as-irradiated condition follows roughly the same behavior as that of the PI annealed samples.

For additional comparison, K-M plots were made of several different copper alloys whose tensile properties have been reported in earlier reports [8,9]. These specimens were flat sheet specimens with a somewhat different gage cross-section than those reported for the irradiated pure copper. Figure 7 shows a comparison of the work hardening rate of these various materials and how they compare to the behavior of the as-irradiated conditions and unirradiated copper shown previously. The solution annealed CuCrZr is a solution

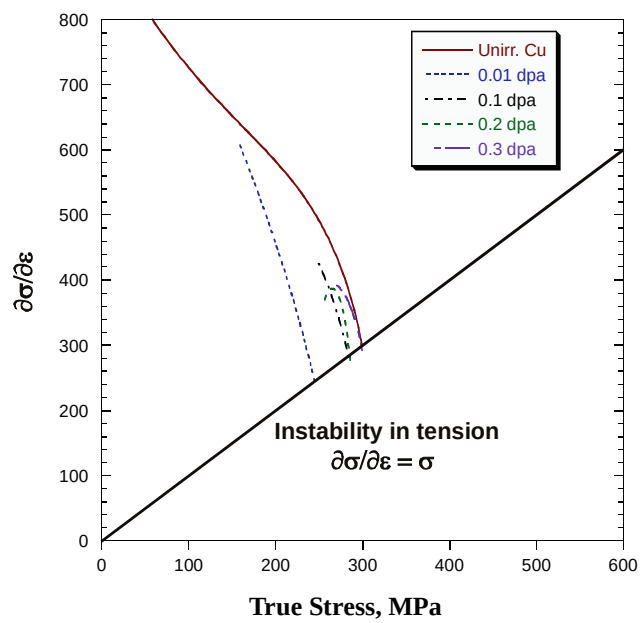

Figure 5. The Kocks-Mecking plots reveal the changes in work hardening with increasing stress in the as-irradiated materials. As the neutron dose increases, the macroscopic work hardening rate begins to match that of the pure copper in the unirradiated state.

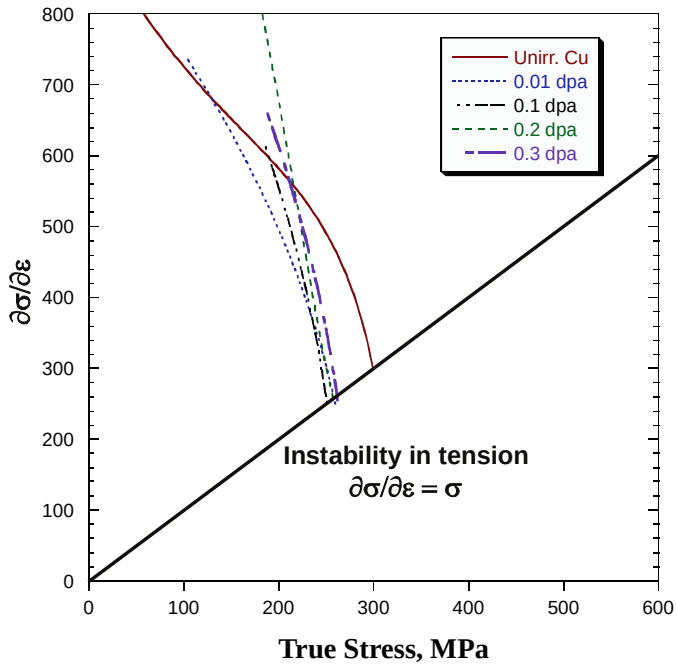

Figure 6. The Kocks-Mecking plots reveal the changes in work hardening with increasing stress after post-irradiation annealing. The work hardening rate is somewhat lower compared to the high dose as-irradiated samples and unirradiated samples, and all samples converge to the same necking stress.

strengthened alloy that possesses a higher yield strength then the pure copper, but work hardens to roughly the same levels. Ageing of the CuCrZr shows that the precipitation strengthening changes the overall response in regard to both the yielding, the work hardening rate and stress at which necking occurs. The CuNiBe alloy (AT condition) is a much higher strength copper alloy far removed from the behavior of the other materials. The final alloy is the oxide dispersion strengthened GlidCop Al25, which behaves in much the same manner as the aged CuCrZr. The main conclusion to draw from these comparisons is that irradiation, despite leading to large increases in the yield strength, does not produce obstacles that are close to the effectiveness of the precipitate particles or oxide dispersions.

Discussion

The true stress-strain behavior and the K-M plots appear to indicate that despite a dramatic difference in yield strength and yielding behavior in the as-irradiated copper, the irradiated materials are “conditioned” in the early stages of deformation such that a few percent strain allows the as-irradiated materials to behave similarly to the unirradiated copper. This is based on the macroscopic behavior as measured from the tensile test and assumes that the strain is distributed homogenously over the specified gage length of the sample. These plots demonstrate that the influence of highly localized deformation (dislocation channeling) is not so easy to distinguish from a macroscopic work hardening standpoint. The 0.2 mm/mm plastic strain (20% elongation) present in the as-irradiated samples (up to necking) implies considerable dislocation motion, yet the deformed microstructures reported by Singh et al. [7] did not show any significant dislocation motion in many areas of the sample. Therefore questions arise concerning the reconciliation of the apparently small degree of homogenous dislocation motion and the relatively large strains to necking. The answers likely are found in

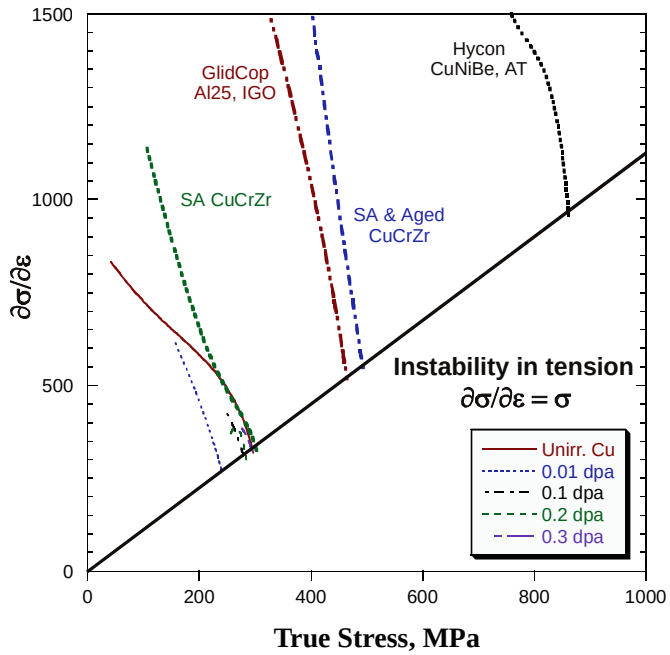

Figure 7. These K-M plots compare the work hardening rates of the as-irradiated pure copper and the unirradiated pure copper to that measured for several different unirradiated copper alloys. Precipitation and oxide dispersion strengthening are clearly more effective in altering the stress-strain response by providing more stronger obstacles that cannot be easily annihilated.

the details of the dislocation storage and annihilation mechanisms that operate once yielding begins and how the clearing of the channels occurs and relates to the dislocation evolution. Since unirradiated copper will form a cellular dislocation network whereas the as-irradiated copper does not, it seems obvious that the mechanisms are different, yet appear to result in the same macroscopic response.

The PI annealed samples and the lowest dose as-irradiated samples raise additional questions since these materials exhibit a decidedly different tensile response. The cause for the onset of necking at lower strains and somewhat lower stresses is not yet known, but bears further investigation. The microstructural characterization of the deformed PI annealed samples (≥0.1 dpa) indicated that deformation occurred via a mixture of channeling and deformation in-between the channels. The 0.01 dpa PI annealed sample exhibited a deformation substructure similar to that of the unirradiated copper, that is, it contained a strongly developed cellular dislocation structure. The removal of the yield point phenomenon was attributed to a decrease in the degree of dislocation decoration by small loops and any impurities in the system. This allowed a more homogeneous production of dislocations, but the presence of channeling even in the PI annealed samples indicates that the dislocation sources still remain pinned to some degree.

The data shown here illustrate that considerable uncertainty exists concerning the exact mechanisms that occur during deformation after yielding. It seems that the tensile test introduces a further complication since the onset of necking disguises any further work hardening that may occur. The data tends to suggest that in a macroscopic sense work softening does not occur in this materials when irradiated at 100°C, but different tests are

needed to further study this. As compression testing does not lead to necking or tensile instability, this test method may provide more information on the work hardening behavior to much higher strains.

One final point to consider is that at a lower irradiation temperature of 47°C pure copper was observed to begin necking after yielding with no measurable uniform strain [1]. It is interesting to note that the maximum stress in this case was around 320 MPa, similar to the stresses found to cause necking in the tensile samples irradiated at 100°C that are reported in this study. This suggests that at lower temperatures, radiation hardening increases the yield strength to the point that upon yielding, the tensile specimens have already reached the instability criteria at small strains and will therefore neck.

FUTURE WORK

The preliminary analysis shown here of the true stress-strain behavior of the as-irradiated copper and PI annealed copper indicates that further analysis of the mechanism of channeling and its effects on the macroscopic strain response to the material bear further investigation. Attempts will be made to relate microstructural observations to the tensile behavior to explain the conditions that lead to necking at lower stresses in the post-irradiation materials for strains similar to that observed in the as-irradiated materials.

ACKNOWLEDGEMENTS

D. J. Edwards would like to thank J. D. Embury and R. G. Hoagland for their helpful discussions regarding the work hardening analysis. The present work was partly funded by the European Fusion Technology Programme. The authors wish to thank B. F. Olsen for the tensile testing in the original experiment. D.J. Edwards' work was also partly supported by the U.S. Department of Energy under contract DE-AC06-76RLO 1830 with the Battelle Memorial Institute at the Pacific Northwest National Laboratory.

REFERENCES

[1] B. N. Singh, A. Horsewell, P. Toft, and D. J. Edwards, J. of Nucl. Mater., 224, (1995), p. 131.

[2] B. N. Singh, D. J. Edwards and P. Toft, J. Nucl. Mater. 238 (1996) 244.

[3] B. N. Singh, D. J. Edwards, M. Eldrup and P. Toft, Journal of Nucl. Matls. 249 (1997), p. 1.

[4] B. N. Singh, D. J. Edwards, M. Eldrup and P. Toft, Effect of Bonding and Bakeout Thermal Cycles on the Properties of Copper Alloys Irradiated at 350°C, Risø-R-971 (EN), Risø National Laboratory, Roskilde, Denmark, September (1997).

[5] D. J. Edwards, B. N. Singh, P. Toft, and M. Eldrup, J. Nucl. Mater. 258-263 (1998) 978.

[6] B. N. Singh, D. J. Edwards, M. Eldrup and P. Toft, Journal of Nucl. Matls. 295 (2001) pp. 1-15.

[7] B. N. Singh, D. J. Edwards and P. Toft, accepted for publication in the Journal of Nuclear Materials, 2001.

[8] G. E. Dieter, Mechanical Metallurgy, 3rd Edition, McGraw-Hill, 1986, pp. 284-290.

[9] D. J. Edwards, Fusion Materials Semiannual Progress Report for the Period ending December 31, 1997, DOE/ER-0313/23, (Oak Ridge National Laboratory), p. 213.

[10] D. J. Edwards, Fusion Materials Semiannual Progress Report for the Period ending December 31, 1998, DOE/ER-0313/25, (Oak Ridge National Laboratory), p. 185.