14 MeV Neutron Irradiation of Copper Alloys

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14-MeV NEUTRON IRRADIATION OF COPPER ALLOYS

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Copper and copper alloys with 5 atomic percent of either aluminum, nickel or manganese were irradiated at 25°C with 14-MeV neutrons to a fluence of 3 x 10^{21} n/m^2. Resistivity, microhardness and electron microscopy were used to characterize the radiation damage. Using a log-normal distribution to describe the defect cluster size distribution, good agreement was found between resistivity estimates of the cluster density and the observed (TEM) density. At least 11% of the defects created during the irradiation escape correlated recombination.

1. INTRODUCTION

A previous investigation[1] of 14-MeV neutron irradiated copper alloys estimated that about 70% of the defect clusters produced in copper during neutron irradiation were smaller than the normal resolution limit of an electron microscope (~ 1 nm). We have used resistivity methods, which are sensitive to these smaller defect clusters, along with TEM and microhardness measurements in an attempt to characterize the nature of the cluster size distribution. This analysis also allows a comparison to be made of the relative sensitivity of these three experimental tools at low neutron fluences. In addition, using appropriate models, resistivity analysis yields the fraction of defects which escape correlated recombination. This quantity is important for radiation damage modeling.

2. EXPERIMENTAL

Foils of pure (99.99^+ atom %) copper and copper alloyed with five atom percent of either aluminum, nickel or manganese obtained from Hanford Engineering Development Laboratory[1] were cold-rolled to a thickness of 25 μm. Transmission electron microscope (TEM) disks and resistivity foils were cut from these foils, annealed in high-purity argon, and allowed to air cool. The metals were irradiated at room temperature using 14-MeV neutrons from the Rotating Target Neutron Source II (RTNS-II) at Lawrence Livermore National Laboratory. The irradiation consisted of four incremental fluences up to a maximum level of about 3 x 10^{21} n/m^2. Details of the experimental procedure have been previously described.[2]

Changes in the foil resistance were measured with standard resistivity equipment (potentiometer sensitivity = 10 nV). The pure copper sample had an initial residual resistivity ratio (RRR = $\rho_{298}/\rho_{4.2°K}$) of 380.

Vickers microhardness measurements were performed[3] using a Buehler Micromet® microhardness tester at indenter loads of 5g and 10g. A minimum of 60 indentations were made around the periphery of four different TEM disks for each metal at every fluence level. Pre-irradiation materials properties and irradiation conditions of the metals investigated are given in Table 1.

3. EXPERIMENTAL RESULTS

Figures 1 and 2 show the respective irradiation-induced resistivity changes for pure copper and the copper alloys. The data for the pure copper sample scales linearly with the square root of neutron fluence. This fluence dependence is in agreement with previous work for both electron and neutron irradiation at temperatures where the interstitial

TABLE 1
Irradiation Data for Resistivity and TEM Samples

Alloy	$\rho_0(\Omega\text{-m})$	Grain Size (μm)	Control Hardness (kg/mm^2), 10g Load	Maximum Fluence (n/m^2) Resistivity	TEM
Cu	4.48×10^{-11}	13	56.7 ± 4.3	2.9×10^{21}	1.9×10^{21}
Cu-5% Al	3.96×10^{-8}	23	53.8 ± 0.9	2.9×10^{21}	2.1×10^{21}
Cu-5% Mn	1.08×10^{-7}	22	53.4 ± 2.5	2.8×10^{21}	2.0×10^{21}
Cu-5% Ni	5.16×10^{-8}	12	53.4 ± 2.8	2.9×10^{21}	2.2×10^{21}

is mobile.[4] Irradiation of the copper alloys produces an initial decrease in foil resistivity, followed by an increase in resistivity with fluence for the Cu-5% Al and Cu-5% Ni alloys. The resistivity of these alloys became greater than the pre-irradiation value at a fluence of 3×10^{21} n/m^2. The resistivity of the Cu-5% Mn sample remained below its unirradiated value at all fluence levels investigated. The negative resistivity changes observed in the irradiated copper alloys are believed to be due to short-range ordering.[5]

Changes in the Vickers microhardness of the four metals obtained at an indenter loading of 10 grams are shown in Fig. 3 as a function of 14-MeV neutron fluence. Results obtained at an indenter loading of 5 grams showed similar trends.[3] As is evidenced in Fig. 3, the hardness data for all four metals scale linearly with the fourth root of neutron fluence after an incubation fluence. The duration of the incubation period is on the order of 1×10^{20} n/m^2, and is shorter for pure copper as compared to the alloys. The Cu + 5% Mn alloy exhibited significantly larger radiation hardening than the other metals at the fluence levels investigated. All four metals in Fig. 3 have roughly equal slopes in their curve of microhardness vs. fluence.

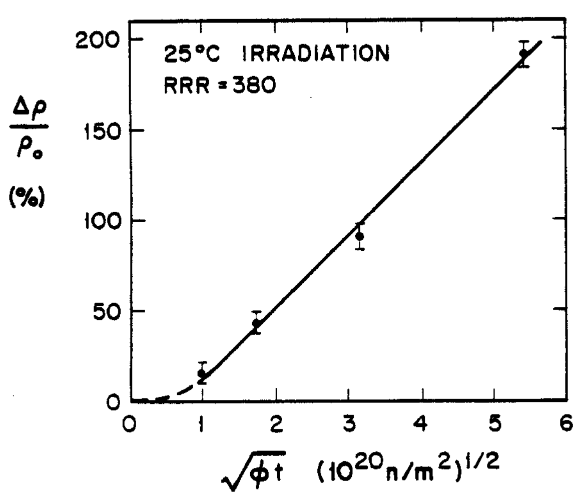

FIGURE 1
Resistance change vs. the square root of 14-MeV neutron fluence for pure copper

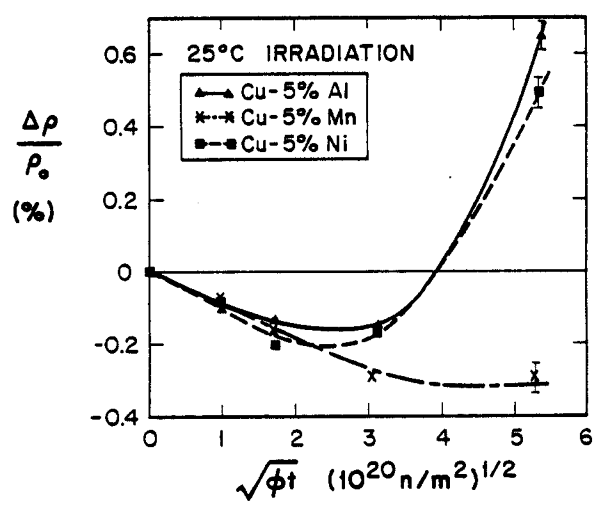

FIGURE 2
Resistance change vs. the square root of 14-MeV neutron fluence for copper alloys

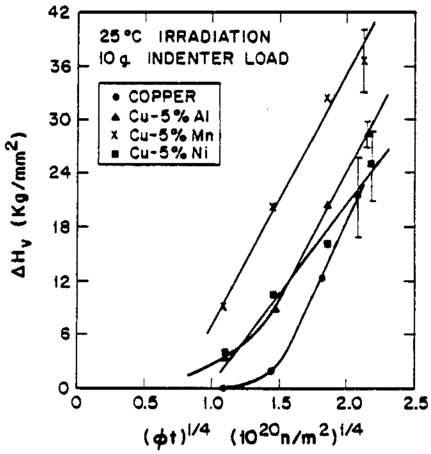

FIGURE 3
Change in Vickers microhardness vs. fourth root of 14-MeV neutron fluence

4. DATA ANALYSIS

Early resistivity studies of pure metals irradiated at temperatures above 70°K provided support for a theoretical model known as the unsaturable trap model (UTM).[4] More recent developments have concentrated on examination of the reciprocal damage rate (RDR) for studying point defect interactions. Using the analysis of Dworschak et. al.[6], the following result is obtained:

$$\Delta\rho = \rho_f^t \frac{r_t C_t}{r_v} \left\{\sqrt{1 + \frac{2 f\sigma_d \Phi}{r_t C_t / r_v}} - 1\right\} \quad (1)$$

This may be rewritten in the form

$$\frac{\Phi}{\Delta\rho} = \frac{1}{f\sigma_d \rho_f^t} \left\{1 + \frac{\Delta\rho}{2 \rho_f^t C_t r_t / r_v}\right\} \quad (2)$$

where Φ is the neutron fluence, f is the fraction of defects escaping correlated recombination, σ_d is the displacement cross section, ρ_f^t is the specific resistivity of trapped Frenkel defects, r_t and r_v are the capture radii of impurity traps and vacancies, c_t is the concentration of impurities, and $\Delta\rho$ is the radiation-induced resistivity increase. A least squares fit to the present resistivity data yields the result

$$f\,\sigma_d\,\rho_f^t = 8.4 \times 10^{-32}\ \Omega\text{-m}^{-3}\ . \quad (3)$$

The quantity ρ_f^t depends on the amount of clustering which has occurred. To obtain a lower limit for the fraction of defects escaping correlated recombination, ρ_f^t may be taken to be equal to the isolated Frenkel pair specific resistivity in copper,[7] ρ_{FP} = 2.0 μΩ-cm/% F.P. Using a displacement cross-section[1] of σ_d = 3690 barns then gives $f \geq 11\%$.

A more accurate estimation of f requires a determination of the effect of clustering on ρ_F^t. Thompson et al.[8] experimentally investigated the effect of clustering on ρ_{FP} during an electron irradiation at temperatures where only interstitials migrate and cluster. Using his result of ρ_f^t/ρ_{FP} = 0.8 ± 0.1 along with ρ_{FD}^i = 1.4 μΩ-cm/% interstitials and ρ_{FD}^v = 0.6 μΩ-cm/% vacancies,[7] the specific resistivity of trapped Frenkel defects (where both interstitials and vacancies are clustered) can be computed. Assuming an equivalent number of vacancy and interstitial clusters leads to a prediction of ρ_f^t = 1.4 ± 0.25 μΩ-cm/% F.P. With this value, f ≈ 16%.

Brager et al.[1] irradiated identical metals with 14-MeV neutrons up to a maximum fluence of 7.5 x 10^{21} n/m² and examined the foils with TEM. We have found that their reported visible defect cluster size distribution can be fitted very well by a log-normal distribution[9] with d_0 = 2.25 nm:

$$N(d) = \frac{1}{\sqrt{2\pi}\ \ln\sigma} \exp\left[-\frac{(\ln d/d_0)^2}{2\,(\ln\sigma)^2}\right]\ . \quad (4)$$

In anticipation that a large number of small defects might be invisible to TEM methods, we also fit the larger defects of the observed distribution to a log-normal distribution

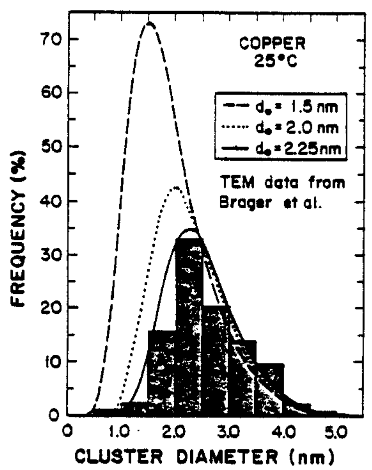

FIGURE 4
Log-normal curve (Eq. 4) fitted to observed TEM cluster data at a 14-MeV neutron fluence of 3×10^{21} n/m^2

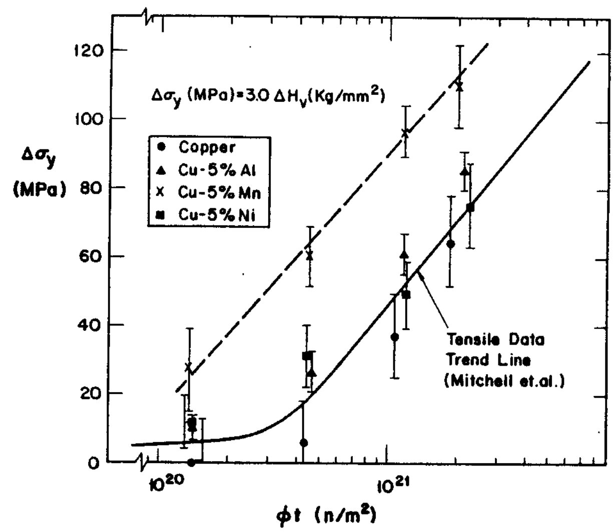

FIGURE 5
Correlated yield strength change vs. 14-MeV fluence

with smaller values of d_0. The fitted distributions are compared to the reported pure copper TEM data of Brager et al.[1] in Fig. 4. Applying the log-normal distribution to the resistivity data gives the result,[2]

$$\Delta\rho = \frac{\pi\rho_F^t N_{c\ell} b}{8} d_0^2 e^2 (\ell n\ \sigma)^2 \qquad (5)$$

where b is the Burgers vector of the defect cluster and $N_{c\ell}$ is the cluster density.

A similar analysis can be applied to tensile data from a room temperature 14-MeV neutron irradiation of copper by Mitchell et al.[10] The hardening due to dislocation loops is given by[11]

$$\Delta\tau = \frac{\mu b}{\beta\bar{\ell}} = \frac{\mu b}{\beta}\sqrt{\sum_j n_j d_j} \qquad (6)$$

where τ = shear stress, μ = shear modulus, and β is a constant. Using the Von Mises criterion to relate yield strength to shear stress, $\Delta\sigma_y = \sqrt{3}\,\Delta\tau$, and once again assuming a log-normal cluster size distribution, Eq. 6 can be solved for the cluster density:

$$N_{c\ell} = \frac{\exp\left[-(\ell n\sigma)^2/2\right]}{3\ d_0} \left(\frac{\beta\ \Delta\sigma_y}{\mu b}\right)^2 . \qquad (7)$$

Vickers microhardness data can be correlated to tensile data results found in the literature, $\Delta\sigma_y$ (MPa) = K ΔH_v (kg/mm^2). A recent correlation[1] found the result K = 3.27 to be valid for irradiated pure copper and also copper alloys. As seen in Fig. 5, we have obtained a reasonable correlation between the pure copper tensile data of Mitchell et al.[10] and the 10g microhardness data for three metals (Cu, Cu-5% Al, Cu-5% Ni) with K = 3.0. The Cu-5% Mn microhardness data do not agree well with the tensile data.

5. DISCUSSION

The general fluence dependence of the radiation-induced change in resistivity and microhardness is in good agreement with theoretical models. Resistivity results for pure copper scale linearly with the square root of 14-MeV neutron fluence, indicating that the cluster density $N_{c\ell} \sim \sqrt{\phi t}$. The microhard-

TABLE 2
Calculated Defect Cluster Densities in Copper at a Fluence of 3×10^{21} n/m^2 Assuming Perfect Dislocation Loops ($b = a_0/\sqrt{2}$)

d_0 (nm)	Fraction of Clusters Which are Visible (Fig. 4)	Predicted Visible Cluster Density (10^{22}/m^3) Resistivity	Microhardness*	Tensile[10]
1.5	0.4	8	7	7
2.0	0.7	9	9	9
2.25	1.0	10.5	12	12

*Extrapolation to $\phi t = 3 \times 10^{21}$ n/m^2 from ΔH_v vs. $(\phi t)^{1/4}$ curve (Fig. 3); assumes $\Delta\sigma_y$ (MPa) = 3.0 ΔH_v (kg/mm^2)

TABLE 3
Comparison of Calculated and Observed Defect Cluster Densities in Copper at a Fluence of 3×10^{21} n/m^2 Assuming All Clusters are Visible

Type of Analysis	Method	Copper Type	Visible Cluster Density (10^{22}/m^3) Perfect Loops ($b = a_0/\sqrt{2}$)	Faulted Loops ($b = a_0/\sqrt{3}$)
Calculation	Resistivity	DAFS	10.5	11$^+$
Calculation	Microhardness	DAFS	12	18
Calculation	Tensile[10]	LLNL/Cominco	12	18
Observed	TEM[1]	DAFS	..........13..........	

$^+$Assumed stacking fault specific resistivity $\rho_{SF} = 2.5 \times 10^{-17}$ Ω-m^2 [14].

ness data of all four metals appears to be proportional to the fourth root of neutron fluence, (again indicating that $N_{c\ell} \sim \sqrt{\phi t}$) but there is insufficient high-fluence data to conclusively determine the actual power law dependence. Most previous experiments have found that the yield stress is proportional to the one-third power of the fluence.[12] The roughly equal slopes for all four metals in the curve of microhardness change vs. fourth root of neutron fluence (Fig. 3) may be taken as an indication that there are equivalent damage production rates in these materials following the initial transition period.

Our best estimate of the fraction of defects which escape correlated recombination (16%) agrees well with the value of 15% found in the literature for electron-irradiated copper.[13]

The calculated visible defect cluster densities obtained from resistivity (Eq. 5) and Mitchell's tensile data (Eq. 7) may be compared to the density which was actually observed by TEM methods. The results of this comparison are summarized in Tables 2 and 3. Parameters used in the calculation were ρ_f^t = 1.4 $\mu\Omega$-cm/% F.P. and $\beta = 3.7$[11]. Brager et al. reported a cluster density of 1.3×10^{23}/m^3 at $\phi = 2.7 \times 10^{21}$ n/m^2 for 14-MeV neutron irradiated pure copper. This value is in good agreement with the calculated densities of 1.05×10^{23}/m^3 (resistivity) and 1.2×10^{23}/m^3 (tensile), which were obtained assuming perfect dislocation loops and that all of the defect clusters were visible. If one assumes that a substantial number of small defect clusters are invisible (i.e. a smaller value of d_0 in Eq. 4 and Table 2), then the calculated cluster density becomes much smaller than the observed value. Brager[1] observed significantly higher cluster densities in 14-MeV neutron irradiated copper than other researchers who irradiated to similar fluences.[10,15] One possible explanation for the discrepancy is that Brager carefully investigated defect clusters down to diameters of 1.0 nm. The size limit of the other TEM investigations is uncertain. From this analysis, it appears that it is possible to observe essentially all of the surviving point defect clusters by TEM methods if a very careful study is undertaken, investigating clusters down to diameters of 1.0 nm.

6. CONCLUSIONS

Approximately 16% of the defects created in copper during room temperature 14-MeV neutron irradiation escape correlated recombination events.

Good agreement has been obtained by fitting observed (TEM) defect cluster size distributions to a log-normal distribution. Using this fitted distribution, estimates of defect cluster density obtained by utilizing resistivity and tensile data have been found to agree well with TEM results. Hence, it appears that TEM may be capable of observing essentially all of the defect clusters produced if careful microscopy is performed on clusters down to sizes of ~ 1 nm.

Resistivity, tensile,[10] and microhardness data indicate that, for a room temperature 14-MeV neutron irradiation, the defect cluster density is linear with the square root of fluence up to $\Phi = 7 \times 10^{21}$ n/m^2.

Analysis of the data obtained during 14-MeV neutron irradiation of copper at 25°C is consistent with previous results[8] which indicate that defect clustering causes a substantial reduction (~ 30%) in the Frenkel pair specific resistivity.

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