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Defect Production and Recovery in FCC Metals Irradiated at 4.2 K*

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ABSTRACT

Four fcc metals, Al, Cu, Ni, and Pt, have been irradiated at 4.2 K at the Livermore Rotating Target Neutron Source (RTNS-II) to fluences of 1 - 2×10^{21} n/m^2. The sample resistivities were monitored during the irradiation. Following irradiation, the sample resistivities were monitored after isochronal anneals to 320 K.

The initial resistivity damage rates for all four metals could be predicted to within ±10% from the results of irradiations in a pure fission spectrum by damage energy scaling. The results of isochronal annealing studies in Al and Cu were nearly identical to recovery curves obtained after fission spectra irradiations to comparable damage levels. On the other hand, both Ni and Pt exhibited less recovery than that observed after fission spectra irradiations, due primarily to less recovery in Stage 1.

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1. INTRODUCTION

This work is part of an interlaboratory program to study low temperature defect production in irradiated metals. The primary purpose of this program is to provide a data base from which fusion neutron irradiation effects can be correlated to fission spectra irradiations. In order to extend this data base, the recent results of defect production and recovery experiments on aluminum, nickel, copper and platinum irradiated at the RTNS-II facility [1] are presented in this paper.

2. EXPERIMENTAL PROCEDURE

All of the samples were cut from 25.4 mm wide cold-rolled foil stocks. After shearing, the samples were annealed for one hour in helium at 200°C for Al and Cu, and 800°C for Ni and Pt. Residual resistance ratios, after making size effect corrections, are given in Table 1 along with the sample purities. The annealed ribbons were mounted flat on the end of a commercial Helitran continuous flow cryostat, and niobium dosimetry foils were placed on each side of the gauge section of the samples to measure the neutron fluence.

The resistances of the samples were measured during irradiation with a computerized data acquisition system which incorporated a Vidar Model 520 digital voltmeter and a DEC LSI-11 computer. This system was programmed to measure the resistance of each sample ten times in both current directions and average the results. Further details of this system are described elsewhere [2].

Table 1: Sample Specifications

Sample	Source	RRR[a]	ρ_0(nΩm)	$\Delta\phi t$(n/m^2)
Al	MRC(99.999%)	74	.386	8.20×10^{20}
Ni	MRC(99.99%)	17	4.603	1.81×10^{21}
Cu	A.D. MacKay (99.999%)	172	.098	8.36×10^{20}
Pt	MRC(99.99%)	72	1.592	1.80×10^{21}

a) RRR = $\rho_{298}/\rho_{4.2}$

3. RESULTS

3.1 Irradiation Results

The experimental results are summarized in Table 2. For each sample, the total observed change in resistivity, $\Delta\rho/\Delta\phi t$, is given. Unfortunately, the neutron flux on the samples is very sensitive to small shifts in the deuteron beam position. As a result, discontinuous shifts in the apparent damage rate of up to 20% can occur. Because of this, only values of $\Delta\rho/\Delta\phi t$ can be determined with any precision ($\sim$5%).

We can obtain the initial damage rate, $(d\rho/d\phi t)_{\Delta\rho=0}$, from measured values of the saturation resistivity, $\Delta\rho_\infty$, using the relation [3]

$$(d\rho/d\phi t)_{\Delta\rho=0} = -(\Delta\rho/\Delta\phi t)(\Delta\rho_\infty/\Delta\rho)\ln(1-\Delta\rho/\Delta\rho_\infty) \qquad (1)$$

To determine the initial damage rates, Equation 1 was used together with apparent values of $\Delta\rho_\infty$ for Al and Cu from Coltman, et al. [4] taken over comparable $\Delta\rho$ values. For Pt, $\Delta\rho_\infty$ was taken from Nakagawa [5].

Non-linear damage rate behavior is frequently observed in Ni and the following method was used to find the initial damage rate. The platinum and nickel samples were adjacent to each other on the cryostat head. Therefore, periodic changes in the neutron flux were approximately the same in both samples. Platinum has a very large saturation resistivity, and its rate of change with neutron fluence is linear at the low neutron doses used in these experiments. Therefore, we were able to use the platinum sample as an absolute flux monitor to obtain correct values for $\Delta\rho/\Delta\phi t$. We then linearly extrapolated the resistivity damage rate data to $\Delta\rho=0$ to find the value of $(d\rho/d\phi t)_{\Delta\rho=0}$ for Ni. Figure 1 shows the resistivity damage rate data for Ni

Table 2: Irradiation Results

Sample	$\Delta\rho_{tot}$ (10^{-11} Ωm)	$\Delta\rho/\Delta\phi t$ (10^{-31} Ωm^3)	$(d\rho/d\phi t)_{\Delta\rho=0}$ (10^{-31} Ωm^3)
Al	33.57	4.09	4.18
Ni	112.08	6.21	6.39
Cu	19.14	2.29	2.48
Pt	109.44	6.07	6.23

and the assumed values for Pt plotted versus $\Delta\rho$. The apparent saturation resistivity of Ni is 16.4 nΩm. This is in good agreement with the value of 15.6 nΩm obtained by Nakagawa [5] in fission neutron irradiations.

3.2 Annealing Results

Stage 1 recovery in cubic metals is generally attributed to the recombination of close, Frenkel pairs and to the long range migration of interstitials. Because of this, the percent of recovery in Stage 1 is a clue to the spatial separation of the interstitials and vacancies produced during neutron irradiation. Low energy atomic recoils (for example, those produced during electron or thermal neutron irradiation) result in closely spaced Frenkel pairs, and hence should show a large Stage 1 recovery. High energy recoils, which are more likely to form vacancy rich cores with widely separated, surrounding interstitials, should exhibit significantly less Stage 1 recovery.

Figures 2a-d, show the recovery spectra from isochronal post irradiation anneals of the four samples. Table 3 lists the percent recovery after Stage 1. The total radiation induced resistivity is shown in parenthesis. Also included in Table 3 are annealing data from reactor and thermal neutron irradiations.

4. DISCUSSION

4.1 Defect Production

Resistivity damage rates can be written in terms of the displacement cross-section σ_d, as

$$(d\rho/d\phi t)_{\Delta\rho=0} = \rho_f \sigma_d \qquad (2)$$

Table 3: Percent Recovery in Stage 1

Sample	Annealing Temperature(K)	Neutron Source: RTNS-II	Neutron Source: Reactor	Neutron Source: Thermal
Al	50	48(0.34nΩm)	45(.061nΩm)[6], 44(1.19nΩm)[7]	---
Ni	65	23(1.12nΩm)	37(.054nΩm)[8], 33(3.64nΩm)[9]	55(.013nΩm)[10]
Cu	60	32(0.19nΩm)	34(.02nΩm)[6], 31(1.16nΩm)[9]	60(.013nΩm)[11]
Pt	50 [a]	18(1.09nΩm)	43(.95nΩm)[12], 34(2.65nΩm)[9]	67(.014nΩm)[11]

a) Lowest annealing temperature in present experiment

where ρ_d is the resistivity per atomic fraction of Frenkel pairs. The rate at which defects are produced by different neutron sources can be determined once σ_d for the source has been calculated.

We can obtain the displacement cross-section from the calculated spectrum-averaged damage energy cross-section, σ_{de}, by using the modified Kinchin-Pease expression

$$\sigma_d = K\sigma_{de}/2T_a \tag{3}$$

where K is an efficiency factor (determined for a given material and neutron spectrum from experiment) and T_a is an effective displacement energy averaged over crystallographic direction. The values of K from our current fusion neutron irradiations of Al, Ni, Cu and Pt are given in Table 4. Values of ρ_f and T_a are from Klabunde and Coltman [3].

It should be noted that the efficiency factor is a function of the primary recoil energy. Experimentally, K is observed to decrease with increasing recoil energy up to several keVs and then approach a constant value. Energetic electrons, for example, are up to three times more effective on damage energy basis in producing defects in most metals at 4K than energetic neutrons are. This decrease in efficiency is believed to be due to the enhanced defect recombination in the vicinity of the cascade. The leveling off in the efficiency can be explained by the formation of subcascades above a certain recoil energy.

For fusion and pure fission neutron irradiations, the recoil energy spectra is hard enough so that the efficiency factors are approximately the same in a given metal. Therefore, initial resistivity damage rates for both types of irradiations should scale as the damage energy cross-sections

Table 4: Damage Efficiencies

SAMPLE	ρ_f	T_a	RTNS-II σ_{de}	RTNS-II K	BSR(LTIF) σ_{de}	BSR(LTIF) K
Al	3.9	27	178	.33	98.3	.31
Ni	6.0	33	301	.23	82.2	.23
Cu	2.0	29	288	.25	81.3	.26
Pt	10.0	44	198	.28	45.8	.31

(Equations 2 and 3). Table 4 compares values of K for fusion neutrons with those for a pure fission spectrum neutrons [13]. Damage energy cross-sections for both spectra were taken from [3]. Overall, the agreement is well within the uncertainties in both theory and experiment. Damage production in these metals scales with damage energy over the recoil energy range from somewhat less than 10 keV to greater than 200 keV. This is in agreement with earlier work done on several bcc metals [3].

4.2 Annealing Results

For both aluminum and copper the recovery as a function of temperature up to 320 K is nearly identical to that found after fission reactor irradiations to comparable $\Delta\rho$. Numerical comparisons are given in Table 3 for Stage I fractional recovery.

In both Ni and Pt and total recovery at 320 K was lower by 10-20% than that observed after fission irradiations. As can be seen in Table 3, this difference arises almost totally from a reduced recovery in Stage I. In both cases impurities or residual cold-work may be responsible for interstitial trapping or clustering [12], as the resistivity ratios of these samples were much lower than expected. Further studies are needed to eliminate the possibility, in Pt especially, that the difference arises from cascade density effects.

5. CONCLUSIONS

We have irradiated four fcc metals with fusion neutrons at 4.2 K and monitored the resistivity damage rates. Post-irradiation isochronal anneals

were then performed. We observed that defect production scaled with damage energy over the recoil energy range from 10 keV to greater than 200 keV. This was in agreement with earlier work done on several bcc metals.

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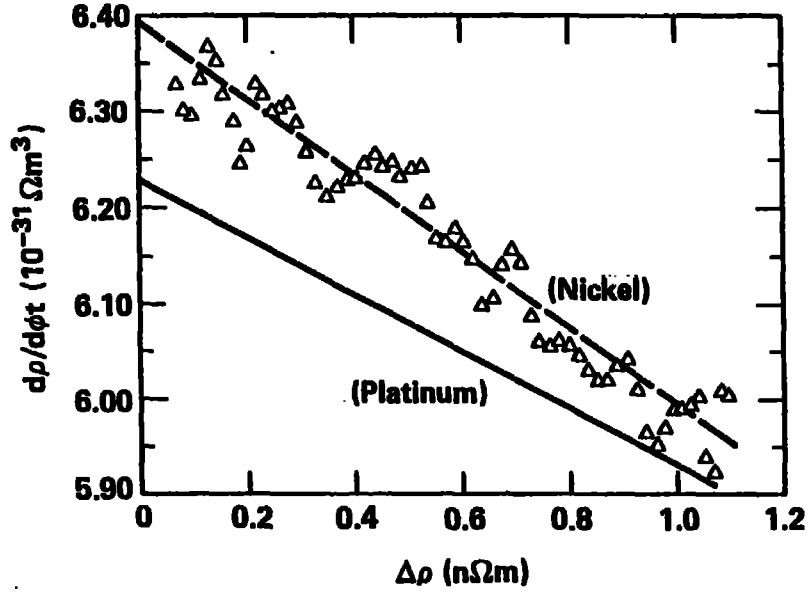
Figure Captions

Figure 1 Resistivity damage rates at 4K for 14 MeV neutron irradiation of Ni and Pt.

Figure 2 Post-irradiation, isochronal annealing results for a) Al, b) Ni, c) Cu and d) Pt. Annealing results below 50K for Ni and Pt were lost because of warming.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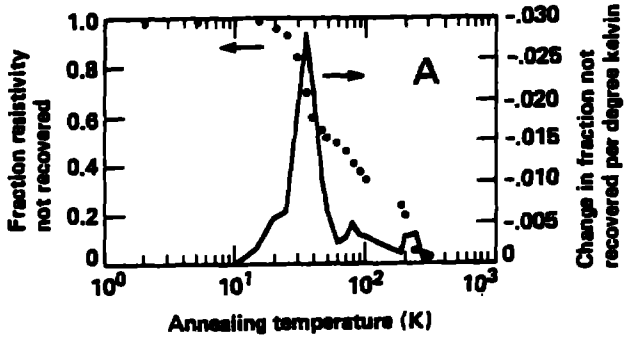
dρ/dφt (10⁻³¹ Ωm³)
Δρ (nΩm)
(Nickel)
(Platinum)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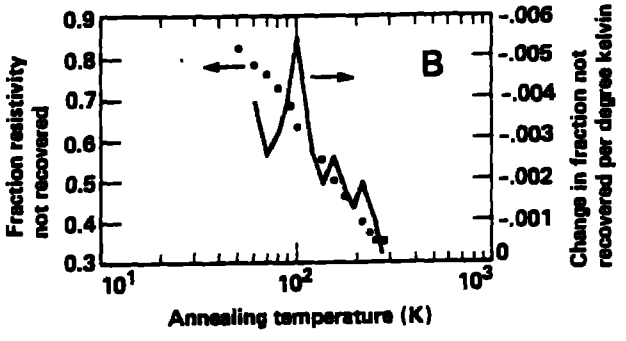
Change in fraction not recovered per degree kelvin
Annealing temperature (K)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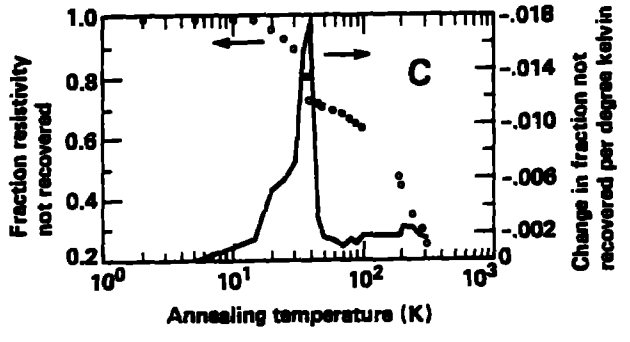
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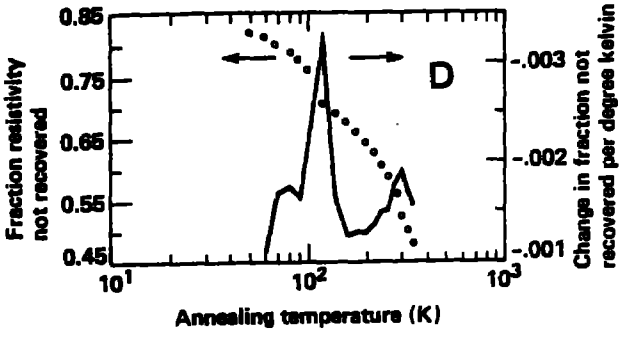
D
Fraction resistivity not recovered
Change in fraction not recovered per degree kelvin
Annealing temperature (K)