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THE EFFECTS OF IRRADIATION ON THE COPPER NORMAL METAL OF A COMPOSITE SUPERCONDUCTOR[†]

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ABSTRACT

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This report presents a new body of magnetoresistance data for copper that were obtained in neutron irradiation experiments at 4K and 330K. These data are combined with previously obtained results on initial damage rates and saturation effects to yield a projection of total resistivity vs neutron dose (expressed in displacements per atom) for copper in service at < 10 K in a magnetic field.

I. INTRODUCTION

Copper is a candidate material for the stabilizer in composite superconductors to be used in the toroidal and poloidal magnets of the Tokamak type of fusion reactor.¹ The anticipated application entails neutron damage to magnet components ranging up to approximately 10^{-2} displacements per atom per year² (dpa/yr) at component temperatures of < 10K and magnetic fields up to 110 kOe.¹ Therefore, it is of interest to have predictive information on the combined effects of these environmental factors on the electrical resistivity of copper. This paper combines experimental information that we now have on radiation damage and magnetoresistivity to yield a construction of resistivity vs magnetic field and dpa appropriate to the application. Some implications for specific positions in the magnet are discussed.

There are three components in the construction.

- 1) A systematization of magnetoresistance data based on Kohler's rule.
- 2) An "initial damage rate" that gives the rate of change of resistivity with respect to an appropriate function of the damaging radiation (dpa) at 4K with no previous damage in the sample.
- 3) A "saturation law" that implies an upper limit to the damage resistivity.

Data in the first component constitutes the main new experimental contribution of this paper. Data in the second were obtained by the present authors for fission neutrons and will be used here and reported in more detail elsewhere.³ Data in the third are taken from the literature. This construction approach is necessary because test facilities that have very high magnetic fields in cryogenic, high-flux environments do not exist; consequently, the available data span only limited ranges of the pertinent variables. We now discuss in more detail the three components mentioned above.

Magnetoresistivity data are often analyzed in terms of Kohler's rule, which states that

$$\frac{\rho(H) - \rho(0)}{\rho(0)} = f\left(\frac{H}{\rho(0)}\right)$$

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$$f\left(\frac{H}{\rho(0)}\right) \quad \left[\frac{H}{\rho(0)}\right]^n$$

with n ranging downward from 2. The obvious test of the rule is to see how well curves of the function vs the argument agree for cases where H and $\rho(0)$ are varied independently. Before the advent of radiation damage it was inconvenient to vary $\rho(0)$ except by varying the temperature. Radiation enables us to vary $\rho(0)$ by introducing defect damage in tests that are pertinent to the proposed application.

The concepts of initial damage rate and saturation behavior are logically introduced together even though we use separate sources for the associated data. It is generally found that for pure metals the residual resistivity increases linearly with concentration, c , of point defects such as vacancies, interstitials, and impurities, at least for small concentrations. Thus, for Frenkel defects (vacancy-interstitial pairs)

$$* \Delta\rho_D = \rho_F c \quad (1)$$

where ρ_F is the resistivity contribution per unit concentration of pairs and $\Delta\rho_D$ is the change in residual resistivity at zero field. Now suppose that Frenkel pairs are introduced into the material at random locations at a constant rate.

If the interstitial constituent of a pair happens to be created within a certain volume, V_0 (spontaneous recombination volume), surrounding an existing vacancy constituent, the vacancy and interstitial recombine; a symmetrical statement applies to a new vacancy vis-a-vis an existing interstitial. The result is that the net rate of addition of new pairs with respect to fluence, Φ , is

$$\frac{dc}{d\phi} = \left(\frac{dc}{d\phi} \right)_0 (1 - 2 V_0 c) \quad (2)$$

where $(dc/d\phi)_0$ is the constant rate of introduction mentioned above. This is the saturation concept due to spontaneous recombination in its simplest form.

The initial defect production rate, $(dc/d\phi)_0$ for a flux of neutrons in a material depends on the neutron energy spectrum, the interactions of the neutrons with the target atoms and their associated energy-dependent cross sections, the non-damage-producing energy loss mechanisms for the knock-ons in the solid, and the energy required to displace an atom. These factors are brought together using the best current theories in any of several computer codes that calculate dpa per unit of fluence. Each code first calculates the spectrum averaged product of cross section and damage-producing energy (damage-energy cross section $\langle\sigma T\rangle$); dpa is then calculated from the "modified Kinchin-Pease relation"

$$D(\text{dpa}) = \frac{0.8\langle\sigma T\rangle}{2 E_d} \phi \quad \text{or thus} \quad \frac{dD}{d\phi} = \frac{0.8\langle\sigma T\rangle}{2 E_d} \quad (3)$$

where E_d is the threshold energy for displacement. The intent of the displacement theory summarized above is to calculate the number of Frenkel pairs that would survive in the absence of losses due to the spontaneous recombination described above. Therefore, $dD/d\phi = (dc/d\phi)_0$. When this relationship is combined with Eqs. 1, 2, and 3, we find that

$$\frac{d \Delta \rho_D}{dD} = \rho_F - 2 V_0 \Delta \rho_D \quad (4)$$

To put experimentally observable quantities into the expression, we note that the initial damage rate, when $\Delta \rho_D = 0$, is

*Notation note: Where explicit designation of magnetic field condition is omitted from a resistivity expression, zero field is implied; i.e., $\rho_x \equiv \rho_x(0)$

$\Delta\rho_D = 0$, is

$$\left[\frac{d\Delta\rho_D}{dD} \right]_0 = \rho_F \quad (4a)$$

and at saturation, when the damage rate becomes zero ($\partial \rho_D / \partial C_\infty = 0$) Eq. 4 yields: $2 V_0 = \rho_F / \Delta\rho_\infty$. (4b)

Putting both of these back into Eq. 4, we find:

$$\frac{d\Delta\rho_D}{dD} = \left[\frac{d\Delta\rho}{dD} \right]_0 \left[1 - \frac{\Delta\rho_D}{\Delta\rho_\infty} \right]. \quad (5)$$

Finally, the integrated form of this differential equation is:

$$\Delta\rho_D = \Delta\rho_\infty \left[1 - \exp \left(- \frac{\left[\frac{d\Delta\rho_D}{dD} \right]_0 D}{\Delta\rho_\infty} \right) \right]. \quad (6)$$

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The Kinchin-Pease approach can be used to calculate initial damage rates by energetic particles other than neutrons. Only for the lowest knock-on energy range (electron irradiation) is the current theory considered to be complete and accurate. At intermediate energies an as-yet-unquantified loss mechanism, which reduces the production efficiency comes into play; it seems to saturate to a constant value for high energies. All fast neutron damage that has been studied is in that saturation region and the efficiency factor is about 30% (for neutron spectra peaked at energies spanning a 15X range). This factor is needed to make ρ_F implied by $(d\rho_D/dD)_0$ measurements agree for neutron and electron damage; but to be consistent with our references, we have not included it in the calculations described below.

The $d\rho_a$ values used here are calculated using "RECOIL" and assume a displacement threshold energy, E_d , of 30 eV. The calculations were made for a pure fission spectrum in which the corresponding resistivity damage rate was measured.³ The result was $[d\rho_D/dD]_0 = 6.49 \times 10^4$ n Ω -cm/dpa. The pure fission spectrum is a well-defined experimentally accessible spectrum to which the theory can properly be applied. The value above is about 25% higher than a value obtained by Kirk and Greenwood⁴ for a reactor spectrum in the VT53 facility of Argonne CP-5 reactor. This percentage discrepancy is consistent for several metals in the same two facilities. Values obtained by Nakagawa et al.,⁵ are about a factor of two higher than these results. This may be due to errors in flux characterization.

Returning to the subject of saturation, we note that there are cases where the ^{linear} decrease in damage rate with accumulated damage is observed to near saturation.⁶ For reactor neutron irradiation, Nakagawa et al.⁵ found that the linear saturation law was not precisely obeyed for copper; they used a cubic fit to get a better description of the data. The corresponding $\Delta\rho_\infty$ was 435 n Ω -cm. However, a large fraction of the curve could be approximated by a linear fit with a $\Delta\rho_\infty$ of 350 n Ω -cm.

For the construction presented below, we chose to use a linear decrement with a $\Delta\rho_\infty$ of 400 n Ω -cm. This approximation is justified considering the overall uncertainties in relating existing data to the application. For example, the higher saturation resistivity, 760 n Ω -cm found for electron irradiations⁷ suggests that the appropriate saturation law depends on the primary knock-on energy and ensuing damage mechanisms.

10 12 We judge that it is still prudent to adopt a saturation law that approximates the existing neutron

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data, although this assumption is not the most conservative one that could be made.

The cubic fit is easily integrated by computer, and we made constructions using it. But because other uncertainties are more overriding than the detailed quality of the fit, we do not believe that use of this refinement is fully warranted at present.

II. EXPERIMENTAL

Magnetoresistivity data were obtained in two experiments using three copper stocks. One sample was irradiated in the Low Temperature Irradiation Facility (LTIF)¹⁰ of the Bulk Shielding Reactor (BSR). This facility allows irradiations at 4K with thermal neutrons. The damage produced from the n- γ recoils consists of point defects. About 5% of the total $\Delta\rho(0)$ results from attendant transmutations. The facility contained a superconducting magnet capable of 25 kOe with the reactor on. (In situ) resistivity measurements were made vs dose at several fields.

Six other samples, 3 bare and 3 cadmium shielded (to eliminate thermal neutron effects) were irradiated at 60°C in CP-15¹⁰ of the BSR. The neutron spectrum in this facility is typical of core spectra for water-moderated reactors. *At this temperature the damage retained has aggregated into dislocation loops.*

After each increment of radiation, the sample assembly was removed to a shielded dewar containing a superconducting magnet. Measurements were made at seven fields up to 51 kOe. Details of this experiment are reported elsewhere. ¹¹ Some of the information *about this work is* shown in Table I. The neutron doses shown are for general information and do not enter the analysis.

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TABLE I
Summary of Experimental Information

	Sample No.	Stock No.*	Cd Shield	Initial $\rho(0)$ (n Ω -cm)	Final $\rho(0)$ (n Ω -cm)
LTIF Irrad. to 1.8×10^{18} Thermal at 4K	7	2	No	0.780	5.75
CP-15 Irrad. to 4.5×10^{19} (E > 1.0 MeV)	1	3	No	21.610	105.70
Reactor	2	1	No	3.967	94.09
neutrons	3	2	No	0.894	90.15
at 330K	4	3	Yes	20.671	91.05
	5	1	Yes	3.071	71.00
	6	2	Yes	0.844	66.79

*Stock 1 = ASARCO Cu, 99.999%, vacuum annealed at 1000°C
 Stock 2 = Same as above, annealed in 9×10^{-5} torr air
 Stock 3 = Commercial Cu magnet wire, 0.010 dia.

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III. RESULTS

To illustrate typical data, Kohler plots for two individual samples are shown in Fig. 1. Deviations from Kohler's rule are evident within the data on a single sample and from sample to sample. The deviations are outside the range of experimental errors, and there are systematic aspects to them. For example, data from the three runs on *stock 2* (not shown) suggest that some of the deviations depend on the *nature of the electron scattering from different defects* e.g., transmutations, dislocation loops, point defects. Nevertheless, for purposes of the present analysis we have chosen to treat the data as statistical and have plotted all the data for all the samples in Fig. 2. Rather than describe the dispersion by a fit and a standard deviation, however, we have constructed two straight lines that envelop most of the data in the pertinent range of $H/\rho(0)$; i.e., to the right of the vertical tic at $H/\rho(0) = 110 \text{ kOe}/430 \text{ n}\Omega\text{-cm}$. These lines correspond to solutions of the form

$$\rho(H) = 10^\alpha H^\beta \rho(0)^{1-\beta} + \rho(0) \quad (7)$$

where α and β are the intercept and slope, respectively, of one of these lines on the Kohler plot. $\rho(0)$ is

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$$\rho(0) = \rho_o(0) + \Delta\rho_D(0) \quad , \quad (8)$$

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where $\rho_o(0)$ is the preirradiation resistivity, and $\Delta\rho_D(0)$, given by Eq. 6 with the values cited above, is

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$$\Delta\rho_D(0) = (400 \text{ n}\Omega\text{-cm}) \left(1 - e^{-162D} \right) \quad (9)$$

The remaining figures are constructions of $\rho_D(H)$ vs D. They represent simultaneous solutions of Eqs. 7, 8, and 9. The curves are spaced by H as a parameter in the third dimension in Fig. 3. and by $\rho_o(0)$ as a parameter in Fig. 4. For each value of a parameter, a curve is constructed corresponding to each of the two envelope lines in Fig. 2. Thus, the band width between the two curves is a measure of the uncertainty in ρ_D vs D due to dispersion in the magnetoresistance data. To better illustrate the low dose dependency, we have displayed ρ_D vs log D in Fig. 5 for two values of $\rho_o(0)$ at $H = 100 \text{ kOe}$.

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IV. DISCUSSION

Since the neutron dosimetry in the above tests was converted to dpa, we can go directly to the application because Santoro et al.² have calculated the neutron fluxes at several positions in a Tokamak toroidal field coil and in turn converted these to dpa rates. Table II presents resistivity values for a service time of one year at Santoro et al.'s position 4 for two cases, one with a penetration that is being considered for the neutral beam injector, and the other without the penetration. Position 4 is on the plasma side of the winding next to the shield. Resistivity values corresponding to the middle of the magnetoresistance band are given for three representative starting materials.

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TABLE II
Projected Copper Resistivities at Position 4
in a Tokamak

Starting Material $\rho_o(0)$ n Ω -cm	Total Resistivities ρ_D (110 kOe) n Ω -cm		
	Unirrad.	1 yr, no penetration 6.4×10^{-4} dpa	1 yr with penetration 1.3×10^{-2} dpa
3	62	89	392
10	62	95	399
30	78	114	419

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This table and the figures above illustrate certain features. At low doses much of the resistivity is due to magnetoresistance and is relatively insensitive to baseline resistivity. At high doses most of the resistivity is due to radiation damage and is insensitive to both the baseline property and magnetoresistance.

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When point defects accumulated by irradiation at low temperature are allowed to move about freely, as they do at higher temperature, the majority will disappear by vacancy-interstitial annihilation. But a certain fraction may agglomerate into clusters of same-type defects which may collapse into stable dislocation loops. In copper annealed at room temperature about 15-20% of the low-temperature damage resistivity will remain in this fashion. The percent residue of stable damage is rather independent of dose for virgin doses.

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If the service life of a Tokamak superconducting magnet involves repeated cycles of irradiation to high damage concentrations followed by anneal at room temperature, then two as-yet-untested conditions arise:
(1) The initial damage rate and saturation behavior of the residue-containing Cu is unknown. It is not expected to be quite the same as unirradiated Cu.
(2) The build-up of the residue from cycle to cycle is unknown; does it saturate?

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We note that another candidate material for the stabilizer, aluminum, probably has higher resistivities over most of the dose range than copper during irradiation at <10K. This is because both the initial damage rate and the saturation value are about a factor of two higher than for copper. Also, the magnetoresistance curves lie above the Cu curves in the applicable range. However, aluminum has the property that, upon annealing to 300K, essentially all the radiation-induced resistivity is removed, offering a prospect for a magnet that can be completely restored by annealing to room temperature.

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Additional uncertainties in the above construction are difficult to quantify, but their possible influence can be identified. The neutron spectra in Tokamak windings may be harder in some places than the fission reactor spectra where most test data have been obtained. Present evidence^{5,7} suggests that damage production efficiency values in this higher energy range have an uncertainty of 20%. Other locations, however, are expected to have considerably softer spectra which might lead to higher damage efficiency values and also higher saturation resistivity values. We have also not dealt with the possible influence of gas production on the resistivity changes and the contributions due to transmutations. As is suggested by Fig. 1, the dispersion in magnetoresistance data resides mainly in differences in slope from sample to sample, and these differences are apparently related to the damage structures. The mix of structures spanned in our tests (loops, impurities, etc.) incorporates the structures expected in Tokamak windings, and it includes certain extreme cases, nearly pure loops (Sample 5) and nearly pure Frenkel pairs (Sample 7). These cases correspond closely with the envelope lines (Fig. 2). Therefore, it is reasonable to expect that magnetoresistance effects in the Tokamak winding will not exhibit drastic deviations from the dispersion band suggested by our envelope.

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The constructions presented in the report are not intended to serve as a base from which precise magnet designs can be developed. The methods formulated here, however, can be used at such time as the necessary data become available.

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FIGURE CAPTIONS

Fig. 1. Kohler plots for two samples. ($H: kOe$, $\rho: n\Omega\text{-cm}$)
 + Sample 5 during irradiation in CP-15

X Sample 7, preirradiation, $\rho_o(0) = 0.78 \text{ n}\Omega\text{-cm}$, H varied

◇ Sample 7, during irradiation in LTIF

The diagonal lines are only to facilitate comparison.

Fig. 2. Kohler plot containing all the data for all the samples. α 's and β 's are the intercepts and slopes of the envelope lines. There are 1218 points on the graph.

Fig. 3. ρ_D vs D for $\rho_o(0) = 30 \text{ n}\Omega\text{-cm}$ at several H 's

Fig. 4. $\rho_D(H)$ vs D for $H = 100 \text{ kOe}$ at several $\rho_o(0)$'s

Fig. 5. $\rho_D(H)$ vs D for $H = 100 \text{ kOe}$ at two $\rho_o(0)$'s

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FIG. 1

FIG. 2

$$\rho_0(0) = 30 \text{ n-}\Omega\text{-cm}$$

FIG. 3

H=100 kOe

FIG. 4

FIG. 5