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UCRL-85455
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EX-11-21-64

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Journal of Nuclear Materials
Second Topical Meeting on
Fusion Reactor Materials
Seattle, WA
August 9-12, 1981

August 7, 1981

Lawrence
Livermore
Laboratory

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FUSION-NEUTRON DAMAGE IN SUPERCONDUCTORS AND MAGNET STABILIZERS

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Two NbTi and two Cu wires were irradiated with 14.8 MeV neutrons at 4.2 K to fluences of $6-8 \times 10^{20}$ n/m², using RTNS-II. Electrical resistances of Cu were monitored during irradiation. Magnetoresistances were measured in fields up to 12.4 T before and after irradiation and after isochronal annealing up to 273 K. Critical currents of NbTi were measured after irradiation, in fields up to 10 T. The initial rate of increase of resistivity of the Cu was found to be 2.23×10^{-31} ($\Omega\text{-m})/(\text{n/m}^2)$. This rate could be predicted from fission reactor irradiations using damage energy scaling. The maximum observed change in the NbTi critical current was a decrease of 3% at 4 T. At 6, 8, and 10 T there were no significant changes.

1. INTRODUCTION

The projected cost of a fusion reactor depends on the sensitivity of the superconducting magnet components to radiation damage. The three magnet components of major concern for radiation sensitivity are the superconductor, the stabilizer, and the insulating materials. The effect of radiation on insulating materials was the subject of a recent meeting, where two of us (RVK and MWG) discussed the relative sensitivity of these components [1]. This paper reports on an experiment to measure radiation effects on NbTi, which is a candidate superconductor, and Cu, which is a candidate stabilizer material. Aluminum wires were also included, but are not described here due to space limitations.

In the case of NbTi, the important parameter is the critical current density, J_c , at the operating temperature and magnetic field. For stabilizer materials, the important parameter is the resistivity at the operating temperature and field. Because a fusion reactor magnet will receive repeated low temperature irradiations and (perhaps) room temperature anneals, it is important to determine how the damage will accumulate under these conditions. The intent in the experiment described here was to perform repeated irradiations and anneals on the same samples, and the stabilizer materials were to have purities which bracketed the range of purities of interest. However, the NbTi samples were accidentally destroyed after annealing near room temperature, and one of the stabilizer wires did not have as high a conductivity as was desired. Accordingly, we performed only one irradiation and anneal cycle on these samples. Future experiments will include repeated cycles.

2. EXPERIMENTAL SAMPLES, APPARATUS, AND PROCEDURE

The NbTi samples used in this experiment were taken from the same wire used by R. M. Scanlan [2]. It was multifilamentary wire manufactured by Aircro, Inc., of Nb-45 wt % Ti alloy with a Cu-to-superconductor ratio of about 2:1. The wire was 0.2 mm in diameter and contained 121 filaments 12 μm in diameter. The initial critical current density was 1.9×10^6 A/m² at 4.2 K and 12.4 T. The wire was provided by C. L. Sneed, Jr., and is similar to that used in his experiments at Brookhaven. Two copper wires of 0.12 mm diameter were soldered to each sample 10 mm apart to serve as voltage measurement leads.

The starting material for one Cu sample was commercial OFHC grade (99.95%) rolled to 25 μm thickness. The other, higher purity sample (99.999%) was standardized MFE material, obtained from A. D. MacKay Co., in the form of 25 μm foil. Samples approximately 20 mm long and 0.12 mm wide were cut from the foils using a precision shear. Both samples were only partially annealed (10 minutes at 400°C) to maintain a relatively small grain size and to retain enough of the cold-work to give residual resistance ratios of 200-300.

The samples were cooled as before [2] by a commercially available Helitran cryostat, which had been modified so that six samples could be irradiated simultaneously. The cryostat could then be transferred to the bore of a superconducting solenoid for critical current and magnetoresistance measurements without warming the samples. The cryostat cold finger was made from a Cu block, and had small grooves machined in its face for the wires. The cold finger was first coated with a thin layer of epoxy, and then the samples were cemented in place with General Electric 7031 varnish to insure good thermal contact, taking care not to deform them. Voltage leads were spot-welded to the Cu samples. Specially-designed tapered Cu current leads were helically wound and

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spoxied to the cryostat finger to minimize heat input to the samples.

The sample temperature was monitored by a carbon glass resistor (Lake Shore Cryotronics) from 4-50 K and by a Cu-constantan thermocouple over the range 50-300 K. The carbon glass resistor readings were corrected for field dependence as before. The resistor was mounted in the base of the cold finger, 65 mm back from the samples, in order to minimize radiation effects on it. The temperature of the sample block was maintained at 4.2 K during the irradiation and measurements. The maximum temperature reached during dewar refilling was 6-8 K. A temperature rise of 0.2 K occurred whenever the neutron flux was introduced onto the samples. A small heater was mounted on the cold finger for annealing.

Irradiation was performed using the Rotating Target Neutron Source (RTNS-II). The machine produced an average flux of $5-6 \times 10^{15}$ n/m² - sec on the samples. The neutrons were nearly monoenergetic at 14.8 MeV. The fluences were measured by activation of seven Nb wires which were cemented to the face of the cold finger between each of the six experimental samples. The cross section used for the $^{93}\text{Nb}(n,2n)^{92}\text{Nb}$ reaction was 458 mbarn. The dosimetry wires were mounted in the same plane on each side of the experimental samples. Neutron output was recorded during the experiment using a counter attached to a proton recoil detector. The overall uncertainty in determination of neutron fluences was $\pm 5\%$.

The procedure used was first to cool the samples to 4.2 K and to measure the initial critical currents of the superconductors and the resistances of the Cu samples over the transverse magnetic field range 2.0 - 12.4 T, in the RTNS-II hot work room. The magnetic field was produced by an IGC superconducting solenoid magnet built into a specially-designed dewar so that it has a room-temperature, horizontal bore. The field was measured with a magnetoresistance probe built into the magnet. The critical current was measured by ramping the current up until a 1 μV signal was observed across the 10 mm gauge section, using an Astrodata Model TDA-121 Nanovoltmeter. Then the current was automatically switched off to prevent overheating the samples. The critical currents were reproducible to within 0.1 A.

The resistances of the Cu samples were measured by a computerized data acquisition system which incorporated a Vidar Model 520 Digital Voltmeter and a Digital Equipment Corp. LSI-11 computer. This system was programmed to measure the resistance of each sample ten times in both current directions and average the results. It also measured an internal standard, as well as the carbon resistor, the thermocouple and the neutron count during irradiation. The overall uncertainties in resistivity determination were $\pm 2\%$. The

largest contributor to this uncertainty was determination of the location of the voltage leads on the samples.

After the initial values had been measured, the cryostat was moved to the left target room of the RTNS-II. Electrical and helium gas lines to the control room were connected, and the irradiation was begun. During the irradiation, resistance measurements were made every 12,000 counts, which was equivalent to a fluence of 4×10^{18} n/m². A total beam-on time of 40 hours produced fluences in the range $6-8 \times 10^{20}$ n/m² on the various samples (precise values given below).

A time of 80 hours was permitted to elapse after completion of the irradiation to allow decay of induced radioactivity in the target room. The cryostat was then moved to the hot work room, where critical current and resistance measurements at field were repeated. Finally twenty isochronal anneals of 20 minutes duration were performed between the temperatures 5 K and 300 K. After those at 50 K, 100 K and 273 K the measurements were repeated. Problems were experienced with the cryostat heater at 273 K. Failure to attach the voltage leads properly caused overheating of the superconductor wires after this annealing temperature. The apparatus was then allowed to warm up to room temperature. After 6 weeks, the dosimetry foils were removed and gamma counted.

3. RESULTS AND DISCUSSION

3.1 Niobium-Titanium Composite

The results for both samples are given in Table 1, where the critical current, I_c , at (nominally) 4, 6, 8 and 10 T, and the transition temperature, T_c ($I/2$), are shown before and after irradiation and after annealing. The values for T_c and H_{c2} were taken at the half-resistance point of the transition. At 4 T, critical currents in all cases were greater than the limit of present current leads (27.7 A) and therefore could not be measured.

The only significant changes in the superconducting properties as a result of the irradiation were a decrease in critical current of about 3% at 4 T and a decrease in T_c of about 0.6 K. Within the precision of the measurements no changes were observed in the upper critical field or in the critical current at 6, 8 and 10 T.

Unfortunately there have been few other experiments in which NbTi was irradiated with neutrons at low temperature and tested at fields above 5 T. However, Soell et al. [3,4] performed irradiations on NbTi at 5 K with the Munich Research Reactor and tested up to 5 T. The spectrum given for this facility is typical of moderated reactor spectra. Their results would indicate a 3% decrease at a fluence of 1

$\times 10^{22} \text{ n/m}^2$ ($E > 1 \text{ MeV}$). This is fairly consistent with the results of Bircher, Brown, and Scott [5], who found a 2% decrease in J_0 for fields from 2 to 5 T at a fluence of $5.4 \times 10^{21} \text{ n/m}^2$ ($E > 0.1 \text{ MeV}$) in a slightly moderated fission spectrum, and the present results of 3% at roughly $7 \times 10^{20} \text{ n/m}^2$ (14.8 MeV). Note that the damage energy at 14.8 MeV is estimated to be a factor of 4.4 times as great as that of the Bircher, Brown, and Scott spectrum, and 10 times as great as that of the Munich spectrum. Thus the decrease in critical current appears to scale with damage energy [6].

Condition.	$T_c(K)$	$I_c(A)$ at 4.20 K				
		3.98T	6.05T	8.01T	10.07T	
Pre-Irradiation						
Sample (1)	9.42	21.3	13.9	8.6	2.6	
Sample (2)	9.45	21.7	14.1	---	2.7	
Irradiated						
$7.9 \times 10^{20} n/m^2$ (1)	8.76	20.7	14.0	8.7	2.6	
$6.3 \times 10^{20} n/m^2$ (2)	8.91	20.9	14.1	8.7	2.7	
50K anneal						
(1) ---	22.3	14.2	8.7	2.6		
(2) ---	21.6	14.5	8.7	2.7		
100 K anneal						
(1) ---	21.4	14.3	8.7	2.7		
(2) ---	21.4	14.3	8.7	2.7		
273 K anneal						
(1) ---	---	---	---	---		
(2) ---	21.2	14.2	---	---		

Table 1: Niobium-45 wt% Titanium Results

The decrease in transition temperature of 0.6 K is about a factor of four to five larger than observed for this level of damage in fission reactor fast neutron irradiations at 4.2 K, but is comparable to that observed after reactor core irradiations at ambient temperature [7]. Our observed change cannot be the result of radiation damage to the carbon glass resistor, since the fluence at the resistor ($2 \times 10^{19} \text{ n/m}^2$) is less, by more than an order of magnitude, than that required for observable changes in calibration [8]. Unfortunately, no annealing data were obtained for T_0 , and a detailed interpretation will have to await the next experiment.

3.2 Resistivity Damage Rates in Cu

The results of irradiation with 14.8 MeV neutrons on the stabilizer samples are given in Table 2. Listed are the initial resistivities, ρ_0 , final resistivities, ρ_f , maximum fluence, ϕ , and the initial damage rates, $d\rho/d\phi$. The values given have been corrected for electrical size-effects [9], which amounted to about 10% in ρ_0 .

Sample	$\rho_0(\Omega\text{-m})$ ($\times 10^{11}$)	$\rho_f(\Omega\text{-m})$ ($\times 10^{11}$)	$\phi(\text{n/m}^2)$ ($\times 10^{-20}$)	$d\rho/d\phi(\Omega\text{-m}^3)$ ($\times 10^{31}$)
Cu				
99.95%	8.28	24.36	7.25	2.24
99.99%	9.49	27.47	8.35	2.21

Table 2: Resistivity Damage Rates in Cu

In the past, resistivity damage rates for other high energy neutron spectra have been found to scale with the calculated damage energy [12]. Table 3 shows a comparison of the fusion: fission spectrum ratio of measured damage rates to calculated damage energies. The fission spectrum measurement is from Colman et al. [11].

Material & Spectrum	Measured $d\rho/d\phi(\Omega\text{-m}^3)$ ($\times 10^{31}$)	Calc. [10] $\sigma E(b\text{-keV})$	Rel. $d\rho/d\phi$	Re. σDF
Cu				
Fission	.723	85.2	1.00	1.0
RTNS-II	2.22	286.4	3.07	3.36

Table 3: Comparison of Ratio of Measured Resistivity Changes to Calculated Damage Energy Ratio

The agreement is within 10%, which is less than the combined uncertainties in damage energy cross sections, dosimetry and resistivity measurements (approximately 10, 5, and 2% respectively).

3.3 Magnetoresistance of Cu

The results of resistivity measurements to 12.41 T for the copper samples are given in Table 4 before and after irradiation and after annealing at 50 K, 100 K and 273 K.

Corrections for size effects have been made. We note that the OFHC Cu sample initially has a much higher magnetoresistance than the high purity sample, but after irradiation and annealing their behavior is similar. This we believe to be the result of redistribution of impurities as a result of the irradiation.

Resistivity changes as a function of field, H , are usually presented in the form of a Kohler plot [13] in which the fractional change in resistivity at field is plotted as a function of field divided by initial (zero field) resistivity, $\rho(0)$, i.e.

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

Condition	Resistivity (10^{-11} Ω -m)				
	0.00T	2.16T	3.98T	8.01T	12.41T
Pre-Irrad.					
99.95% (1)	8.28	16.47	24.45	44.88	68.52
99.999% (2)	9.49	17.33	24.49	41.25	57.99
Irradiated					
(1)	24.36	30.27	36.87	55.12	76.30
(2)	27.47	33.36	39.60	56.19	74.84
50K anneal					
(1)	19.50	25.84	32.98	51.54	72.40
(2)	22.54	28.55	35.20	52.13	70.07
100K anneal					
(1)	18.37	24.78	32.20	50.34	70.29
(2)	21.20	27.31	34.23	51.08	69.53
273K anneal					
(1)	12.88	20.23	27.72	45.39	64.22
(2)	14.35	21.72	28.57	45.53	63.41
302K anneal					
(1)	12.37	---	---	---	---
(2)	14.04	---	---	---	---

Table 4: Magnetoresistivity of Cold-Worked and Partially-Annealed Cu

Fickett [14], in measurements of annealed Cu up to 10 T, found deviations of less than 5% from a single Kohler plot when $\rho(0)$ was varied by changing both purity and temperature. More recently, Williams et al. [15] found substantially different Kohler plots for irradiated samples in which the nature of the defects varied from point defects to loops.

We are aware of only three studies in which Kohler plots have been determined for samples irradiated at 4-5 K with neutrons. Williams et al. irradiated 99.999% Asarco Cu, internally oxidized and fully annealed, with thermal neutrons at 4 K, and we irradiated cold-worked and partially annealed samples of both 99.95% Phelps-Dodge oxygen-free Cu and 99.999% A. D. MacKay Cu. The initial and final values of $\rho(0)$ and maximum fields used are given in Table 5.

Sample	Maximum Field (T)	Resistivity ($\text{H}\alpha 0$) ($\text{n}\Omega$ -cm)	
		Initial	Final
Asarco[15]	2.5	0.78	5.75
MacKay	12.4	10.68	28.58
Phelps-Dodge	12.4	9.49	25.47

Table 5: Available Data for Neutron-Irradiated Cu

Kohler plots for all three samples deviated substantially from that of Fickett and, to a much lesser degree, from each other, as shown in Fig. 1.

Figure 1: Kohler Plots for Cu: The Solid Lines Represent the Limits Observed by Williams et al. [15], with the Lower Corresponding to the 4.2 K Thermal Neutron Irradiation. The Dashed Line is from Fickett [14] and the Symbols Show the Range of Data Given in Table 4.

Assuming all the samples had started with a $\rho(0)$ of 8.0 $\text{n}\Omega$ -cm (which we believe to be representative of magnet material), we can determine the expected resistivities at 8T and 12T from the individual Kohler plots. These vary by as much as 15%, since the Kohler plots were not identical. The criterion to be applied to the stabilizer in design of a fusion reactor magnet is the maximum value of the resistivity at field at the end of its life. To compare the three samples, we have taken the average resistivities at 8T and 12T, and increased them by 25% to arrive at limiting values of 52 $\text{n}\Omega$ -cm at 8T and 75 $\text{n}\Omega$ -cm at 12T. (We have assumed that 25% is the design factor.) Again, from the individual Kohler plots we can determine the increase in $\rho(0)$ above 8.0 $\text{n}\Omega$ -cm at $H = 0$ required to reach these limiting values at field. These allowed increments in resistivity are given in Table 6 for each of the three samples.

Sample	$\Delta\rho$ ($\text{n}\Omega$ -cm) at $H = 0$ to Reach:	
	52 $\text{n}\Omega$ -cm at 8T	75 $\text{n}\Omega$ -cm at 12T
Asarco	12.4	17.8
MacKay	16.6	25.9
Phelps-Dodge	14.6	21.2
	14.5 ± 2.2	21.6 ± 4.3

Table 6: Allowable Resistivity Increases for Cu at $H = 0$.

We note that the allowed resistivity increments are 48% larger at 8T and 62% larger at 12T than estimates using Fickett's Cu magnetoresistance data. Although our numbers show variations of $\pm 15\%$ at 8T and $\pm 20\%$ at 12T, they are probably more representative of the behavior to be expected in a fusion reactor. In measurements on the conductor to be used in the High Field Test Facility, Scanlan [16] has found a magnetoresistance plot close to that found here in OFHC Cu.

ACKNOWLEDGMENTS

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract No. W-7405-ENG-48.

REFERENCE

- [1] Van Konyenburg, R. A., with Appendix by Gulman, M., Relative Radiation Sensitivity of Insulators, Stabilizers and Superconductors, Lawrence Livermore National Laboratory report UCRL-8543E, a talk presented to the Meeting on Electrical Insulators for Fusion Magnets held at DOE Headquarters, Germantown, MD, (December 2-3, 1980).
- [2] Scanlan, R. M., Low Temperature Irradiations of Niobium-Titanium with 14-MeV Neutrons, Lawrence Livermore National Laboratory report UCRL-79418 (1977).
- [3] Soell, M., van der Klein, C. A. K., Bauer, H., and Vogl, G., The Influence of Low Temperature Neutron Irradiation on Superconducting Magnet Systems for Fusion Reactors, IEEE Trans. Magnetics MAG-11, (1975) 178.
- [4] Soell, M., Influence of Radiation Damage on the Maximum Attainable Magnetic Field for Toroidal Fusion Magnet Systems, J. Nucl. Mater. 72, (1978) 168.
- [5] Personal communication between RVK and Bruce S. Brown, Argonne National Laboratory (1980). Results to be published in Journal of Nuclear Materials.
- [6] Brown, B. S., Radiation Effects in Superconducting Fusion Magnet Materials, J. Nucl. Mater. 97 (1981) 1-14.
- [7] Sekula, S. J., Effect of Irradiation on the Critical Currents of Alloy and Compound Superconductors, J. Nucl. Mater. 72 (1978) 91-113.
- [8] Personal communication between MMG and Ronald M. Scanlan, Lawrence Livermore National Laboratory (1980).
- [9] Dworschak, F., Schuster, H., Mollenberger, H., and Wurm, J., The Influence of the Size Effect on Electrical Resistivity Measurements in Irradiated Metals, Phys. Stat. Sol. 21 (1967) 741-745.
- [10] Personal communication between MMG and Lawrence R. Greenwood, Argonne National Laboratory (1980). Recoil spectra and damage energy cross-sections calculated from ENDF/B-IV using the DISCS code are available as a function of neutron energy on the MFE computer network by calling the global file SPECTER.
- [11] Colman, R. R., Jr., Klabunde, C. E., and Williams, J. M., Rates of Defect Production by Fission Neutrons in Metals at 4.7 K, submitted to J. Nucl. Mater. (1980).
- [12] Kirk, M. A., and Greenwood, L. R., Determination of the Neutron Flux and Energy Spectrum in the Low-Temperature Fast-Neutron Facility in CP-5, Calculations of Primary-Recoil and Damage-Energy Distributions, and Comparisons with Experiment, J. Nucl. Mater. 80, (1979) 159.
- [13] Kohler, M., Zur magnetischen Widerstandsänderung reiner Metalle, Ann. Phys. (Germany) 32 (1938) 211-218.
- [14] Fickett, F. R., Magnetoresistivity of Copper and Aluminum at Cryogenic Temperatures, in Proc. of 4th Intl. Conf. on Magnet Technology, CONF-72C908, Brookhaven National Laboratory, (1972) 539.
- [15] Williams, J. M., Klabunde, C. E., Redman, J. K., Colman, R. R., Jr., and Chaplin, R. L., The Effects of Irradiation on the Copper Normal Metal of a Composite Superconductor, IEEE Trans. Magnetics MAG-15, (1979) 731.
- [16] Personal communication between MMG and Ronald M. Scanlan, Lawrence Livermore National Laboratory (1981).

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