UCRL- 96200
PREPRINTRADIATION IMPOSED LIMITS ON SUPERCONDUCTING
MAGNETS: A DATA BASE FOR COPPER STABILIZERSM. W. Guinan
P. A. Hahn
Lawrence Livermore National LaboratoryC. E. Klabunde
R. R. Coltman
Oak Ridge National LaboratoryThis paper was prepared for submittal to
Third International Conference on
Fusion Reactor Materials
October 4-8, 1987
Karlsruhe, F. R. Germany

October 28, 1987

Lawrence
Livermore
National
Laboratory

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

BEST COPY AVAILABLE

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

RADIATION IMPOSED LIMITS ON SUPERCONDUCTING MAGNETS:
A DATA BASE FOR COPPER STABILIZERS

M. W. Guinan¹, P. A. Hahn¹, C. E. Klabunde² and R. R. Coltman, Jr.²

¹Lawrence Livermore National Laboratory
Chemistry and Materials Science Department
Livermore, CA 94550 U.S.A.

²Oak Ridge National Laboratory
Solid State Division
Oak Ridge, TN 37830 U.S.A.

ABSTRACT

Two of eight differently prepared copper stabilizer samples, previously irradiated in the RTNS-II at LLNL, the IPNS-1 at ANL, and the BSR at ORNL, have been irradiated to a fluence of 1.33×10^{22} n/m² at RTNS-II. During the course of the irradiation the samples were periodically removed (without warming) for measurements of the transverse magnetoresistance and returned for continued irradiation. This experiment extends the range of neutron-irradiation-induced resistivity by a factor of five over the previous experiments. A simple model is developed which reproduces the magnetoresistance results of all the experiments to an accuracy of 2.5%.

Work supported by the Reactor Technologies Branch, Office of Fusion Energy, U.S. Department of Energy by the Lawrence Livermore National Laboratory under contract number W-7405-ENG-48.

1. INTRODUCTION

In order to predict the response of the stabilizer material in a large superconducting magnet to cyclic irradiation and annealing, the magnet designer requires four pieces of information:

1. A detailed knowledge of the neutron spectrum in the magnet.
2. The rate of resistivity increase at zero field in the spectrum.
3. The dependence of annealing at room temperature on initial purity, cold-work, and neutron spectrum and fluence.
4. The resistivity at field, arising from variations in the zero field resistivity due to initial purity, irradiation, cold-work and annealing.

The importance of item one cannot be over-emphasized. It is potentially the largest source of error in predicting magnet response.

Given the adequacy of item one, the initial damage rate can be predicted from damage energy calculations and a knowledge of the damage efficiency as a function of recoil energy^{1,2}. At present, an accuracy of 10% or better is achievable. Tables of initial damage rates as a function of neutron energy are available.¹

The annealing response is sensitive, i.e. up to a factor of two, to initial purity, cold-work, neutron spectrum and fluence.^{3,5} We will present only a rough correlation of the available data. Further work is required at higher fluences.

The main concern of this report will be directed at correlating all the data available from a previous RTNS-II experiment,^{1,3} experiments at the ANL/IPNS-I and BSR Facilities by Klabunde and Coltman^{3,4} and the present higher dose results.

2. SAMPLES

The details of sample preparation are given in previous reports.^{3,4} Two samples, one of 99.999% recrystallized copper from ASARCO with an oxidizing anneal, and a second of OFHC copper from Intermagnetics General Corp., unoxidized, with 7.5% cold-work, were utilized in the latest experiment.⁶ The first, designated ASARCO-0, had a residual resistivity ratio ($R_{293}/R_{4.2}$) of 1780. The second, designated OFHCUN-1, had a ratio of 166. Both had 10 mm gauge lengths.

3. EXPERIMENTAL RESULTS

The ASARCO-0 and OFHCUN-1 samples were irradiated in five segments to final fluences of 1.20×10^{22} n/cm² and 1.33×10^{22} n/m², respectively. At the end of each segment, the magnetoresistance was measured at 2, 4, 6, 8, 10 and 12T. During three of the transfer operations to the 12T magnet, unintentioned temperature excursions to 40-50K were experienced. Complete details of the experiment are given in Ref. 6.

4. RESULTS AND DISCUSSION

From the analysis of the raw resistance data, resistivities were determined for each sample assuming a thermal resistivity of 17.86-nΩ-m at 36.1°C. All values were then corrected for electrical size effects.⁷ This is the same procedure followed in other experiments.³⁻⁵

Annealing data

In order to compare the annealing data in our previous work^{1,3} with that at IPNS-I⁴ and BSR⁵, we needed a method of data correlation to compare experiments with different resistivity increments per cycle. An obvious choice was to plot the accumulated annealed resistivity ($\Delta\rho_{ANN}$) vs. the accumulated resistivity at 4.2K ($\Delta\rho_{TOT}$). $\Delta\rho_{TOT}$ serves the purpose of an exposure parameter and in fact is proportional to neutron fluence at low

exposures. Furthermore, it allows us to directly compare experiments in different environments. For the purpose of estimates of retained damage, we have plotted $\Delta\rho_{\text{ANN}}/\Delta\rho_{\text{TOT}} \times 100$ (i.e. % of total damage retained) as a function of $\Delta\rho_{\text{TOT}}$, for three cases. The first, Fig. 1, shows the data for sample OFHCUN-1 in three spectra. The RTNS data represent a spectrum much harder than that generally expected at a fusion reactor magnet, the BSR data a much softer spectrum, and the IPNS data one comparable to magnet spectra. As the figure shows, the retained damage varies by a factor of two, with more remaining in the harder spectra.

In the neutron spectra at fusion reactor magnets less than 0.1% of the resistivity changes at 4.2K arise from transmutations. While this is true for the RTNS-II and IPNS irradiations, in the soft BSR spectrum about 3% of the resistivity change is due to Zn transmutation products. Therefore, in order to compare the annealing of defects at the BSR, 3% should be subtracted from each of the points shown in Fig. 1.

Fig. 2 shows the effect of cold-work on retained damage. The RTNS-II irradiation data are for samples OFHCUN-0, 1 and 2 with 0%, 7.5% and 14.3% cold-work respectively. As expected, the difference due to cold-work diminishes as the exposure is increased. This is the result of the buildup of annealed damage and irradiation modification of the microstructure. At very high exposure we expect the samples to approach a single microstructure.

The results for the IPNS-I irradiations are qualitatively similar; since these are at lower damage levels, the differences due to cold-work are larger. At the only overlapping exposure (about $30 \times 10^{-11} \text{ } \Omega\text{-m}$) corresponding to our first cycle, the retained defect resistivity varies from 0.7 to 0.8 of ours with the lower values corresponding to the largest amount of cold-work.

The influence of initial purity is shown in Fig. 3, where results for recrystallized samples ASARCO-0, OFHCUN1-0 and OFHCOX-0 are shown. Comparing these results with those in Fig. 2, we see that an increase in purity corresponding to a reduction of 2×10^{-11} $\Omega\text{-m}$ has about the same effect as increased cold-work corresponding to an increase of 5×10^{-11} $\Omega\text{-m}$. Again the results are qualitatively similar to that observed at IPNS-I. Differences in the retained defect resistivity are almost independent of dose. At 30×10^{-11} $\Omega\text{-m}$ exposure, the IPNS-I retained resistivities were about .80 of those at RTNS-II.

The shape of curves in Figs. 1, 2 and 3 suggest a reciprocal damage rate model⁸, developed for electron irradiation, i.e. $\Delta\rho_{\text{TOT}}/\Delta\rho_{\text{ANN}}$ vs. $\Delta\rho_{\text{ANN}}$ might provide a linear fit to the data. In fact such a fit has been attempted to room temperature irradiated copper in both a reactor⁹ and RTNS-II.¹⁰ All the RTNS-II plots show a slight but real upward curvature, while the IPNS plots show both positive and negative curvatures. The resulting latitude in slope and intercept is sufficient to preclude any simple interpretation in terms of impurity or cold-work effects. The results are close enough to linear, however, that an attempt to develop a similar model in terms suitable for cascade defect production should prove fruitful.

Magnetoresistivity

In our earlier work^{1,3} at RTNS-II we reported that the effects of irradiation, cold-work and annealing on a given material (OFHCUN) all fell on a single Kohler plot, i.e.

$$(\rho(H) - \rho(0)) / \rho(0) = f(H/\rho(0)). \quad (1)$$

This appeared to be true over the limited range of $H/\rho(0)$ for the OFHCUN samples, 0.1 to 2×10^{11} T/ $\Omega\text{-m}$.

When the higher dose data and the results from IPNS-I and BSR became available, it was clear that this was no longer a valid conclusion, particularly at low values of $H/p(o)$.

The present data set, from all three experiments, cover the range of $H/p(o)$ from 0.015 to 12.5. When all of the data are plotted on a single Kohler plot, a pronounced fan develops in the region below about 0.5×10^{11} T/R-m. At $H/p(o) = 0.015$ the spread amounts to a factor of 4 in $f(H/p(o))$. While this amounts to an uncertainty in $\rho(10T)$ of only 12-15%, after 5-10 anneal cycles the lifetime could be changed by up to 50%. To try and account for the spread, we begin eliminating any data set which produced points in the fan central region. At the end of this process only two branches were left. The remaining data are shown in Fig. 4. The sets of files giving rise to the two branches were distinguishable. One set, the lower, consisted of files for which 95% or more of the resistivity at zero field was the result of point defects or small clusters of point defects, i.e. the files for unirradiated, recrystallized samples and those for which no room temperature anneals had been carried out after irradiation. The remaining set consisted of files for which more than 90% of the resistivity arose either from cold-work and/or annealed radiation damage.

Since our final aim is only to develop an equation which will describe all the magnetoresistance effects arising from any combination of extended and point defects, any form will suffice. It would be better if the limiting behavior of $f(H/p(o))$ followed that observed; proportional to H at high fields and vanishing as H^2 at low fields.

Ziman¹¹ proposed a simple function for the conductivity of a polycrystalline sample with a multiply connected Fermi surface based on behavior expected for a simple model cylindrical Fermi surface averaged over

all crystalline directions. In this case the electrical conductivity is given by

$$\sigma(H) = \sigma(0) (1/\omega_H \tau) \operatorname{atan}(\omega_H \tau) \quad (2)$$

where ω_H is the cyclotron frequency with the field parallel to the cylinder axis and τ is the electron collision time. Since $\omega_H = (e/m^*c)H$ and τ is proportional to $\sigma(0)$, we can rewrite eq. 2 as

$$\sigma(H) = (1/CH) \operatorname{atan}(CH/\rho_0) \quad (3)$$

where C is a function only of the density of states at the Fermi surface. In order to fit the data of Fig. 4, an initial attempt was made to fit the point defect data set to two such terms, reflecting the contribution of two sets of electrons with different Fermi surface orbits to the overall conductivity. This requires three parameters and provided only a poor ($\pm 12\%$) fit to the data.

Three such terms produced an excellent fit to the point defect data sets but require five parameters. The resulting equation for $f(H/\rho(0)) = f(x)$ is

$$f(x) = \frac{1}{(1/CX) \operatorname{atan}(f_1 CX) + (1/DX) \operatorname{atan}(f_2 DX) + (1/EX) \operatorname{atan}((1-f_1-f_2)EX)} - 1. \quad (4)$$

As before C , D , and E depend only on the density of states at the Fermi surface. While f_1 & f_2 are partial contributions to the total conductivity at zero field, and are dependent on the scattering behavior of the electrons

comprising that group. The third term in the denominator insures that the sum of the individual contributions add up to the total conductivity.

The point defect data set is shown in Fig. 5 with the fitted function. The standard deviation of the 180 data points from the fit is $\pm 2.1\%$. Only four points (not shown) had deviations greater than 4%. Values for the constants are given below with their corresponding standard deviations.

$$\begin{aligned} C &= 9.683 \pm .080 \\ D &= 153.5 \pm 11.8 \\ E &= 15.37 \pm .48 \\ f_1 &= .7698 \pm .0091 \\ f_2 &= .2049 \pm .0100 \\ f_3 &= .0263 \end{aligned}$$

While five parameters were required we could find no other 4 parameter fit, using completely arbitrary functions, that did as well. We are now left to fit the data sets in which the resistivity arose entirely from extended defects. Since C, D, and E are material parameters, only f_1 and f_2 can vary. The resulting fit can be seen in Fig. 6. The additional 84 data points of this set are fit with a standard deviation of 2.9%. The value of f_1 decreased and f_2 increased as below, resulting in a net increase in f_3 .

$$\begin{aligned} f_1 &= .6266 \pm .0253 \\ f_2 &= .3436 \pm .0237 \\ f_3 &= .0398 \end{aligned}$$

We note that the resulting fit comes very close to the Kohler function found by Fickett¹² on high purity copper in which $\rho(o)$ was determined by phonon contributions at low $H/\rho(o)$. We also note that within the accuracy of

the fit we could hold the ratio of f_2 to f_3 constant, implying that only the scattering of the f_1 electrons is changed by the introduction of extended defects.

To fit the remaining data, comprising an additional 1000 data points, we only need combine these two fits. Since the f 's are proportional to the partial conductivities, a fit to mixed point defect and extended defect sets would use values for f_1 and f_2 as follows

$$f_1 = f_{iPD}\rho_{PD}/\rho_0 + f_{iED}\rho_{ED}/\rho_0 \quad (6)$$

where ρ_{PD} and ρ_{ED} are the portions of the resistivity at zero field arising from point and extended defects, respectively.

5. CONCLUSIONS

A simple model has been developed which can predict the magnetoresistance of copper as a function of initial purity, the extent of cold-work and the presence of both irradiation produced point defects and the extended defects produced by annealing. The remaining area of uncertainty in a complete description of the response of the copper stabilizer in superconducting magnets to irradiation-anneal cycles is the dependence of annealing on dose, impurity concentration and cold-work. Design equations are in preparation for inclusion in the Fusion Materials Handbook.¹³

6. ACKNOWLEDGEMENT

The authors wish to thank R. A. Van Konynenburg for his participation in the experiments at RTNS-II.

REFERENCES

1. M. W. Guinan and R. A. Van Konynenburg, J. Nucl. Mater. 122 & 123, 1365-1370 (1984).
2. R. A. Simons, Damage Analysis and Fundamental Studies Quarterly Progress Report for Jul-Sept. 1980, U.S. DOE Report DOE/ER-0046/3, 41 (1980).
3. M. W. Guinan and R. A. Van Konynenburg, Fifth Annual Progress Report on Special Purpose Materials for Magnetically Confined Fusion Reactors, U.S. DOE Report DOE/ER-0113/2, 99-123 (1983).
4. C. E. Klabunde and R. R. Coltman, Jr., Sixth Annual Progress Report on Special Purpose Materials for Magnetically Confined Fusion Reactors, U.S. DOE Report DOE/ER-0113/3, 53-58 (1984).
5. R. R. Coltman, Jr., J. Nucl. Mater. 108 & 109, 559-71 (1982).
6. M. W. Guinan, R. A. Van Konynenburg and J. B. Mitchell, Sixth Annual Progress Report on Special Purpose Materials for Magnetically Confined Fusion Reactors, U.S. DOE Report DOE/ER-0113/3, 30-39 (1984).
7. F. Dworschak, W. Sassin, J. Wick and J. Wurm, "Tables for Correcting the Size Effect of the Electrical Resistivity in Foils and Wires," Jul-575-FN, Feb. 1969, KFA Julich, Germany (1969).
8. R. M. Walker, "Radiation Damage in Solids," Proc. of the Inter. School of Physics (Enrico Fermi) Course XVIII, Ed., D. S. Billington, 594 (1962).
9. R. L. Chaplin and R. R. Coltman, Jr., J. Nucl. Mater. 108 & 109, 175-182 (1982).
10. S. J. Zinkle and G. L. Kulcinski, J. Nucl. Mater. 122 & 123, 449-454 (1984).
11. J. M. Ziman, Electrons and Phonons, Clarendon Press, Oxford, 512-521 (1960).
12. F. R. Fickett, Proc. of the Fourth Intl. Conf. on Magnet Technology, Brookhaven National Laboratory, CCNF-720908, 539-541 (1972).
13. J. W. Davis, "Materials Handbook for Fusion Energy Systems," this conference (1987).

FIGURE CAPTIONS

- Fig. 1 The dependence of the retained resistivity ($\Delta\rho_{\text{ANN}}/\Delta\rho_{\text{TOT}}$) on the accumulated resistivity at $4K(\Delta\rho_{\text{TOT}})$ for three neutron spectra; RTNS-II(Δ), IPNS-I($\diamond$), and BSR($\square$) for sample OFHCUN-1.
- Fig. 2 The dependence of the retained resistivity ($\Delta\rho_{\text{ANN}}/\Delta\rho_{\text{TOT}}$) on the accumulated resistivity at $4K(\Delta\rho_{\text{TOT}})$ for three levels of cold-work; OFHCUN-0(0%)($\square$), OFHCUN-1(7.5%)(Δ) and OFHCUN-2(14.3%)($\diamond$).
- Fig. 3 The dependence of the retained resistivity ($\Delta\rho_{\text{ANN}}/\Delta\rho_{\text{TOT}}$) on the accumulated resistivity at $4K(\Delta\rho_{\text{TOT}})$ for three levels of purity; ASARCO-0(Δ), OFHCOX-0($\diamond$) and OFHCUN-0($\square$).
- Fig. 4 Kohler plots for samples for which the resistivity at zero field is due entirely to either point defects(Δ) or extended defects($\square$).
- Fig. 5 The Kohler plot for samples for which the resistivity at zero field is due entirely to point defects. The solid line is the fit to the data (see text).
- Fig. 6 The Kohler plot for samples for which the resistivity at zero field is due entirely to extended defects. The solid line is the fit to the data (see text). The dashed line is the fit to the data of Fig. 5.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6