BNL-64951

CONF-970201--

RESISTIVITY CHANGES IN SUPERCONDUCTING-CAVITY-GRADE Nb FOLLOWING HIGH-ENERGY PROTON IRRADIATION*

C. Lewis Snead, Jr., A. Hanson, G.A. Greene, A. Ghosh, and C. J. Czajkowski,
Brookhaven National Laboratory
USA

K. C. Dominic Chan
Los Alamos National Laboratory
USA

Thomas E. Ward
Department of Energy
USA

and

H. Safa
C. E. Saclay
France

RECEIVED
DEC 1 8 1997
OSTI

Abstract

Niobium superconducting rf cavities are proposed for use in the proton LINAC accelerators for spallation-neutron applications. Because of accidental beam loss and continual halo losses along the accelerator path, concern for the degradation of the superconducting properties of the cavities with accumulating damage arises. Residual-resistivity-ratio (RRR) specimens of Nb, with a range of initial RRR's were irradiated at room temperature with protons at energies from 200 to 2000 MeV. Four-probe resistance measurements were made at room temperature and at 4.2 K both prior to and after irradiation. Nonlinear increases in resistivity simulate expected behavior in cavity material after extended irradiation, followed by periodic anneals to room temperature. For RRR = 316 material, irradiations to (2 - 3) x 10^{15} p/cm^2 produce degradations up to the 10% level, a change that is deemed operationally acceptable. Without periodic warming to room temperature, the accumulated damage energy would be up to a factor of ten greater, resulting in unacceptable degradations. Likewise, should higher-RRR material be used, for the same damage energy imparted, relatively larger percentage changes in the RRR will result.

Introduction

There are currently several high-energy physics facilities where niobium superconducting rf cavities are being used to accelerate charged particles (1). These cavities are being proposed for use in the Accelerator Production of Tritium (APT). In this application, however, where protons are accelerated both to high energy and high current (1.3 to 1.7 GeV at 100 mA), there exists no operational experience as to the potential degradation of performance of the superconducting Nb due to radiation damage. This damage could occur in the cavities due either to catastrophic beam loss of steering or focus, or by low-current, continued radiation owing to beam halo effects. For a facility such as the proposed APT with a projected service life of 30 years, unreversible degradation of cavities requiring frequent replacement would obviate their use, and result instead in the use of less-efficient room-temperature (normal conducting) rf cavities for the LINAC. Optimal performance depends upon the cavity maintaining a high value of Q, which depends on the matrial's surface conductivity. Changes in the bulk conductivity of the cavity material can adversely impact the performance of the cavity through degradation of the Q, and higher cooling demands and higher risk of quenching becaouse of decreased thermal conductivity. The change of either of these parameters as a function of proton fluence can provide a measure of performance degradation.

In this paper we report the change of the residual resistivity ratio (RRR) for proton irradiations of niobium at room temperature for energies between 200 and 2000 MeV. The RRR is a measure of bulk resistivity. Even though if is not directly related to the cavity surface resistance, it is the property specified for the quality of the niobium because it is related to the chemical, disolved gas, and defect purity of the material. Standard cavity-grade Nb has RRR = 300. Even if the changes in RRR due to radiation damage do not directly couple to changes in Q, they are important since thay do directly relate to changes in the thermal conductivity of the Nb. Degraded thermal conductivity carries deleterious implications on power loss at cavity surfaces resulting in reduced efficiency, and enhanced cavity quenching.

Background

Beam halo losses in the APT superconducting accelerating structure have been estimated at about 0.2 nA/m (2). To estimate the fluence on a given beam-transport element properly requires high-energy transport-code calculations and consideration for greater beam loss at the lower energies. For purposes here we estimate that for a twenty-year operation, the surface of a 5-cm-diameter pipe would see a fluence of about 10^{14} p/cm^2. In actual operation, this fluence will not be the total damaging fluence, because energetic spallation particles including recoiling nuclei and neutrons produce most of the damage.

To date there have been but a few experimental attempts to measure cavity degradation due to high-energy proton damage. Halama (3) irradiated a superconducting Nb cavity (4) at 1.4 K operating at about 3 GHz with 175-MeV protons at the Brookhaven LINAC. During the irradiations the cavity was maintained at 4.2 K and the beam had a gaussian shape of a few cm FWHM at the center line of the cavity. The fluence quoted is about 10^{15} p/cm^2 over about 3% of the "effective"surface of the cavity. This irradiation produced significant reductions in the measured Q; (20 - 50)% depending upon the mode selected, the highest change seen for the TM(001) mode. No quantitative estimate of the decrease in normal-state residual resistivity due to the irradiation was made, although the surface resistance defined by the measured Q increased significantly.

Recently Safa et al. (5) irradiated a 1300-Mz cavity at room temperature with 1300-MeV protons at the Laboratoir Nationale Saturne (LNS) accelerator at CEA/Saclay, France. As with the Halama irradiations, a small gaussian beam impinged on a small percentage of the total cavity wall at the equator. The fluence was 5×10^{15} p/cm^2. Measurements of the cavity Q were made before and after the irradiation. No change in Q was observed.

More recently (6) two 3000-MHz Nb single-cell cavities were irradiated at the Los Alamos LANSCE facility with 800-MeV protons to a fluence of 1×10^{16} p/cm^2 over approximately 5% of the effective area of the cavity at the equator. The cavities were maintained at a nominal 2 K during the irradiation and the Q measurements. No changes in the cavity Q were measured. To date the results on cavity degadation owing to high-energy proton irradiation appear to be conflicting.

Mathiessen's Rule states (6) that the resistivity of a simple metal can be expressed as a linear combination of terms as

$$\rho(T) = \rho_{Th} + \rho_0 + \rho_{Def} , \qquad (1)$$

where ρ_{Th} is the thermal contribution, ρ_0 is the residual resistivity due to impurities, etc., in the starting material, and ρ_{Def} is the increase in resistivity due to radiation-induced defects. Other terms such as magnetoresistance and size effects are not relevant here. At low temperatures (4.2 K) the thermal component is nearly zero and the resistivity is given by the residual defect contribution for nonirradiated specimens. At room temperature the thermal contribution dominates. As the purity of a specimen is improved, ρ_0 decreases producing an increasing RRR. Note that for a given irradiation producing a ρ_{Def}, its percentage contribution to the low-temperature resistivity and thus the RRR measured will increase as the impurity contribution decreases. This is an important point to keep in mind when examining results obtained from specimens with different starting RRR's.

We write

$$\Delta(RRR^{-1}) = 1/(RRR)_{irrad.} - 1/(RRR)_{nonirrad.} \qquad (2)$$

which is rewritten as

$$\Delta(RRR^{-1}) = [R(4.2\,K, irr) - R(4.2\,K, non)] / R(RT) . \qquad (3)$$

Since all terms for the several resistances in the numerator contain the same specimen -specific term l / a, we can reduce the resistances to resistivities by dividing by l / a. This leaves

$$\Delta(RRR^{-1}) = [\rho(4.2\,K, irrad.) - \rho(4.2\,K, non)] / \rho(RT). \quad (4)$$

Since, at 4.2 K the resistivity is dominated by the residual resistivities and the thermal contribution can be approximated to zero, the numerator becomes $\rho_0 + \rho_{Def} - \rho_0$. At room temperature the thermal contribution dominates the defect contributions, so the denominator becomes $\rho(RT) = \rho_{Th}$ sothat

$$\Delta(RRR^{-1}) \propto \rho_{Def} . \qquad (5)$$

The quantity $\Delta(RRR^{-1})$ is relevant since it is proportional to the change due to damage in the intrinsic variable, the resistivity. It is also independent of the starting RRR (initial impurity level) of the metal.

Experiment

The intent of this experiment was to establish a measure of the damage to niobium as a function of high-energy proton fluence. The property change to be measured was the residual-resistivity ratio (RRR). The irradiations are performed at room temperature, so the results are limiting in the sense that they closely reflect the defect state that would result from irradiation at low temperature followed by warming to room temperature. It is assumed that in actual application the superconducting rf cavities will be so annealed at intervals. Beam time to perform these RRR irradiations became available as the irradiations could be performed in conjunction with a joint US-French experiment measuring the n/p production for protons on various metal targets over the energy range 400 - 2000 MeV. The high-energy irradiations were carried out at the Saturne Accelerator in Saclay,

DISCLAIMER

DISCLAIMER

France. Two fluences of irradiations were carried out at the 200-MeV Brookhaven Linac Isotope Producer.

The starting material for the US RRR specimens was superconducting cavity-grade niobium obtained from Wah Chang through Los Alamos National Laboratory. The manufacturer's specification of purity was that the RRR was in excess of 300. A list of maximum levels of impurities was provided. These specifications list the major impurity of Ta at 920 appm, Zr at less than 100appm, and H and N at less than 5 appm. RRR specimens were cut from this plate by Electro-Discharge Machining to sizes of 3.1 x 0.30 x 0.056 cm. In general three specimens were included in each irradiation package. The RRR (and other specimens reported elsewhere) were wrapped in Al foil and sandwiched between circular aluminum discs of 0.025-mm thickness. For the specimens irradiated at LNS the Al discs were cut to size to fit into a pneumatic can (the "doigt de gant") that could be remotely inserted in and retracted from the beam. The doigt de gant is located at the extraction point of the synchrotron before any beam-line transport optics. The irradiation times ranged from 4 h to 19 h, with one package receiving all of the irradiation segments. In some of the irradiations RRR specimens provided by the French were included in the package. These specimens were 2.0 x 0.9 x 40 mm and had higher initial RRR's. These higher-RRR specimens were originally Nb at 300, but were annealed and degassed to provide starting RRR's of 1726.The specimens for the 200-MeV proton irradiations were mounted in zirconium cans and irradiated with a 100-μA beam at the BLIP Facility. These irradiations were carried out for 15 and 200 minutes.

The proton fluences on the various irradiations was determined by foil activation techniques chiefly using the 0.025-mm Al covers on the sample packages and measuring the Na 24 peak. In some cases the French RRR specimens were employed as dosimetry specimens using the several spectral lines. The main lines are the 1836 and 2898 keV gammas from ^{88}Y, but transitions in ^{88}Zr and ^{75}Se were also used.The shape of the beam at the specimen position was elliptical with the long axis vertical and with the beam spot 3.5 x 1.5 sq cm producing a nonuniform gaussian beam spot of 5.25 sq cm. The specimens were aligned vertically with the long axis of the beam. Fluences quoted in this work are derived from the total protons on the foils or RRR specimens as determined by activation divided by the area of the elliptical beam spot. This would connote uniform irradiation, so the reader is reminded that this is an approximation, and that the fluence, and hence the damage, has a gaussian distribution centered on the middle of the specimen. As a check on the dosimetry determinations the time duration of each exposure was logged. Even though the current circulating in the synchrotron was not constant, and the extraction efficiency varied with energy, the plot of the measured fluence versus exposure time as shown in Fig. 1 is rather linear (the straight line is drawn by eye, and not fitted). We estimate from all sources of error and uncertainty an absolute fluence precision of about 20%.

RRR measurements were carried out using standard 4-probe resistance measurements. Specimen holders were constructed to accomodate 3 specimens per dip into liquid helium. Resistance measurements were made at room temperature and the results corrected to 298 K. Low-temperature measurements were carried out at 4.2 K. The specimens at low temperature were mounted transverse to the field of a superconducting magnet to quench the superconductivity of the Nb so that a resistance determination could be made. Measurements were made in fields of 10, 20,

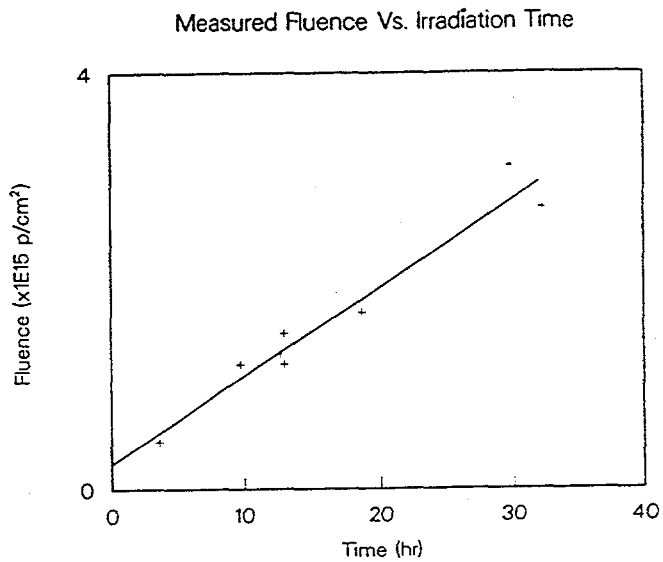

Figure 1: Fluences of the various specimens as determined by foil activation plotted as a function of beam exposure time.

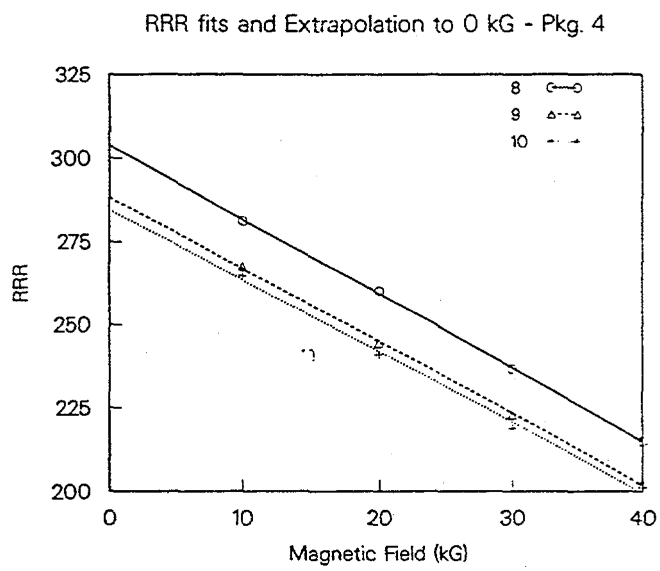

Figure 2: Plot of RRR determinations for three specimens. Sample numbers in the legend correspond to Package # 4.

30, and 40 kG. The low-temperature zero-field value was then determine by a linear extrapolation of the at-field data back to the zero-field ordinate. Fig. 2 depicts the scatter at the ordinate of the extrapolations of one of the determinations. The value for RRR for these runs is given by the average of the intercepts. Note that the slope of these curves defines the magnetoristance for the Nb.

Results and Discussion

The summary of the irradiations of the US specimens at Saturne is given in Table 1. Column 2 gives the calculated average fluence with the equivalent fluence normalized according to the damage-energy cross section (relative to 200 MeV) in parentheses, column 3 the energy of the irradiation, column 4 the measured RRR following the irradiation, column 5 $\Delta(RRR^{-1})$, and column 7 the ΔRRR due to the radiation. The value of RRR for the nonirradiated specimens was determined to be 316, and this value was used in all cases Column 2 gives the calculated average fluence with the equivalent fluence normalized according to the damage-energy cross section (relative to 200 MeV) in parentheses, column 3 the energy of the irradiation, column 4 the measured RRR following the irradiation, column 5 $\Delta(RRR^{-1})$, and column 7 the ΔRRR due to the radiation. The value of RRR for the nonirradiated specimens was determined to be 316, and this value was used in all cases plotted as a linear function of the proton fluence. Indeed, the approach to a saturation value for -ΔRRR is apparent. The scatter in the results at lower fluences is highlighted in Fig. 6 where only the results for fluences up to 4 x 10^{15} are plotted.

Table I Summary of RRR Changes

Pkg. #	Fluence (10^{15} p/cm^2)	Energy (MeV)	RRR	$\Delta(RRR^{-1})$ ($\times 10^{-4}$)	-ΔRRR*
0	5.0 (17.6)	1300	280	4.07	36
1	0.45 (1.74)	2000	301	1.58	15
2	3.1 (11.97)	2000	288	3.08	28
3	8.0 ( )	All	278	4.33	38
4	1.2 (2.0)	400	291	2.72	25
5	3.0 (8.67)	800	290	2.84	26
6	1.7 (4.91)	800	293	2.48	23
7	1.5 (5.52)	1600	288	3.08	28
8	1.2 (4.42)	1600	306	1.03	10
9	1.3 (5.02)	2000	310	0.61	6
A	50. (50)	200	252	8.04	64
B	200 (200)	200	236	10.73	80

* Nonirradiated RRR = 316
Note: Fluences in parens have been scaled by the damage-Saturne is given in Table 1.

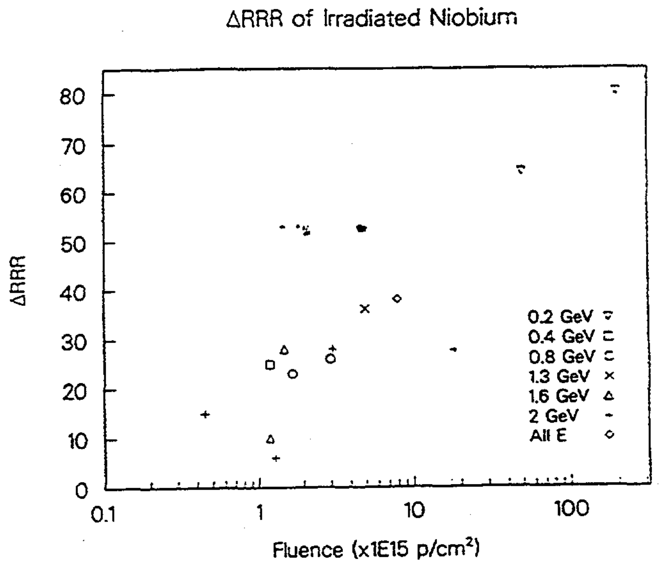

Figure 3: Changes in RRR plotted as the log of the proton fluence.

Nonlinear Damage Production

In Figs. 3, 4, and 5 the nonlinear character of the change in RRR as a function of dose is apparent. That $d\Delta\rho/d\phi$ decreases with increasing dose, so long as the irradiation temperature is above the migration temperature for single interstitials, has long been recognized (7). The rate of change is

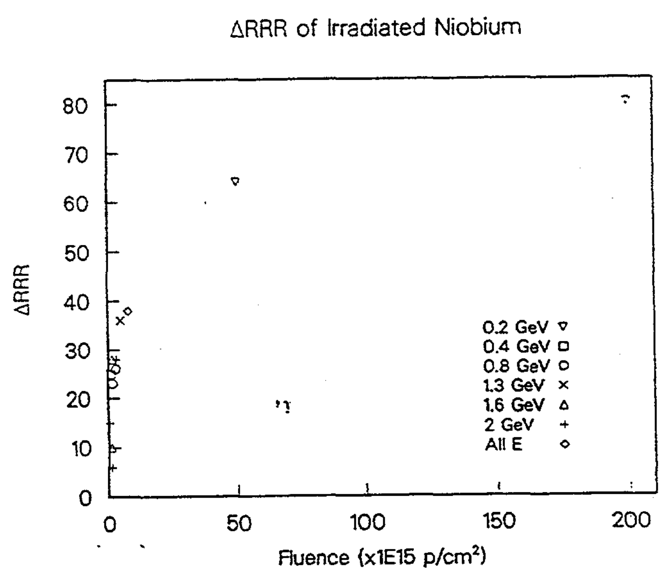

Figure 4: Changes in RRR plotted as a linear function of the proton fluence. The nonlinear response to dose and the apparent approach to a saturation level is apparent.

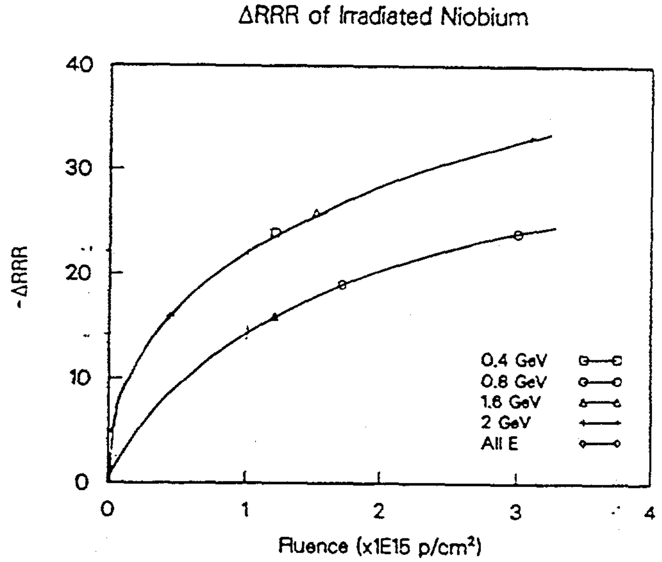

Figure 5: Changes in RRR as a function of lower-fluence proton irradiations. Solid curves have been drawn to bound the spread in the values.

even larger if the irradiation temperature is also above the temperature of single-vacancy migration. For our case of room-temperature irradiation of Nb, the latter case obtains. Interstitial migration has been established to be near 4.5 K (8) and vacancy migration between 200 and 300 K (9). So, for our irradiations, all Frenkel pair defects created by the irradiation at room temperature are mobile, and thus free to recombine or find other traps. Isochronal annealing of damage induced at 4.2 K (9) resulted in approximately 90% recovery. Thus, the damage we observe as changes in RRR for room-temperature irradiations are about 10% of the change that would be induced by the same proton irradiations at 4.2 K. The nonlinear nature is due to the buildup on radiation-induced defects and microstructure that results in a higher annihilation probability for subsequently produced defects.

Comparison Based upon Damage-Energy Cross Sections

Although there is considerable scatter in the low-fluence RRR changes, the apparent scatter seen in Fig. 4 is partly attributable to the fact that the data are plotted as a function of fluence for all of the proton energies. Estimates of damage when comparing either different impinging particles, or like particles at different energies are best done within the framework of the damage-energy cross sections. Assuming the results for iron closely approximate those for niobium the range of damage-energy cross sections (10) for protons range from 1000 bkeV at 400 MeV to 2300 bkeV at 2000 MeV. Thus, the damage-energy cross section increases by a factor of 2.3 for the increase of proton energies of a factor of 5.

In order to compare the scatter of the low-fluence data with expected differences in damage-energy cross sections, the data were bounded by lines drawn through the uppermost and lowermost points as seen in Fig. 6. That $d\Delta\rho/d\phi$ is a decreasing function of fluence because of defect recovery as the damage is being created was discussed above. It is also true that the values for the final slope of the $d\Delta\rho/d\phi$ curve also depend upon the purity state of the material (11). This implies that the family of curves for irradiations at different energies, having different damage cross sections, would also not track each other since the defect buildup per unit fluence would create the same situation as differing levels of impurities for differing specimens. Because of this expected deviation with fluence, comparison of the upper and lower curves of Fig. 6 would best be done for $d\Delta\rho/d\phi$ values obtained at $\phi = 0$. Extrapolation of the data back to the $\phi = 0$ point, however, is quite uncertain. We therefore make the comparison by taking the ratio of the changes in RRR as measured at the $\phi = 1 \times 10^{15}$ p/cm². point. This gives a ratio of 1.6 for the upper value over the lower value to be compared with the value of 2.3 predicted on a damage-energy cross section evaluation.

The data for $\Delta(RRR^{-1})$ are plotted as a function of relative damage in figures 6 and 7. Using the damage energy vs energy results referenced above of Wechsler et al. [12], the present results are normalized by scaling the damage. Here, relative damage is obtained by multiplying the proton fluence by the ratio of the damage-energy cross section for the energy at that fluence by the damage-energy cross section at 200 MeV. In Fig 6 the relative damage energy is plotted as a linear function of the resistivity change. The nonlinear changes are again evidenced, and the data define a single curve, albeit with considerable scatter at the lower fluences. This scatter is better seen in the plot of Fig. 7 of the log of the relative damage. These data, for

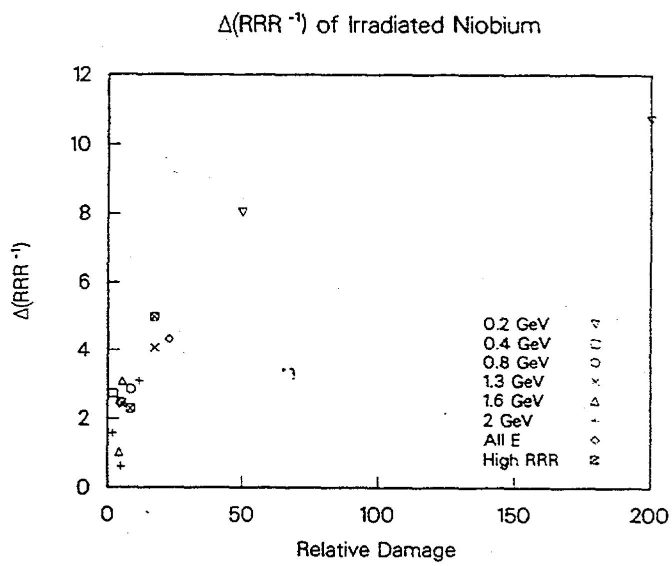

Fig. 6. Plot of $\Delta(RRR^{-1})$ vs the linear change in the relative damage energies. Values are normalized to the damage energy for 200 MeV.

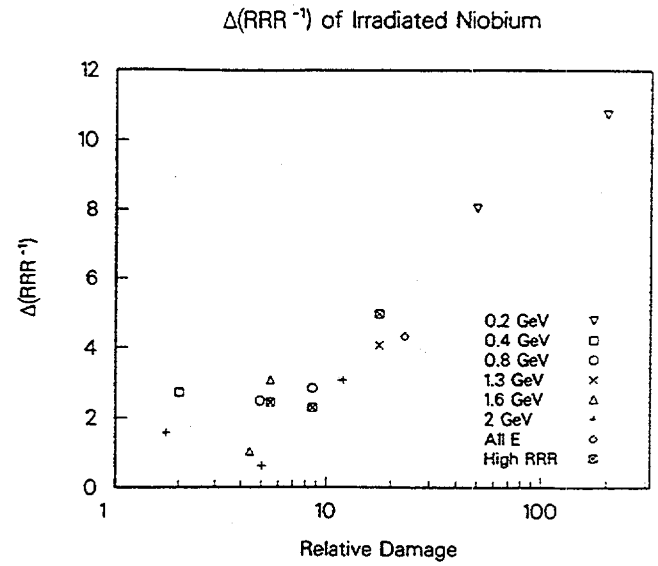

Fig. 7. Same as Fig. 7 but with the relative damage energy plotted on a log scale

both RRR = 316 and RRR = 1716, confirm that the measured $\Delta(RRR^{-1})$ are independent of the RRR of the starting material.

Effects of Differing Starting Resistivity Ratios

The results depicting the RRR results for specimens with different starting RRR values is displayed in Table 2. The results for the three higher energies are from the Saturne irradiations where equal fluences were obtained for both the US specimens and the higher-RRR French specimens. For the 200-MeV irradiations done at BLIP, the RRR's of the French specimens were lower than the US specimens. For the RRR = 175 specimens there is no RRR change measured outside the 10% uncertainty level even for fluences to 2 x 10^{17} p/cm^2, whereas the RRR = 316 evidenced a 25% decrease at that fluence. For the higher-energy irradiations where the two specimen types had RRR's of 316 and 1726, respectively, the percentage changes observed were of the order of four times higher in the higher-RRR specimens, roughly in keeping with the ratio of the RRR's themselves.

One important parameter that characterizes the performance (figure of merit) of a superconducting rf cavity is the Q of the cavity. The higher the Q, the lower the losses, and the more efficient the cavity. This Q value is directly related to the surface resistance R_s by the experssion

$$Q = G / R_s , \qquad (6)$$

where G is a geometrical factor only related to the shape of the cavity The surface resistivity can be divided into two terms. One is the usual BCS resistance which increases exponentially with temperature, and the other is the bulk normal-state resistivity. The BCS part is directly related to the RRR (it increases with RRR), whereas the residual part is mostly independent of the RRR. Therefore, RRR decrease induced by the damage will affect the Q value at higher temperatures (4.2 K, e. g.), but have very little effect on the Q value at lower temperatures (2.0 K, e. g.) As the APT cavities are designed to operate at 2 K, one would expect no significant degradation of of the quality factor even though the RRR might be decreased from 300 to around 200 due to radiation damage over the lifetime of the cavity (assuming no intermediate warming to room temperature). .

Table 2 Effect of Differing Starting RRR Specimens

Energy (MeV)	$RRR_{ini.}$	Fluence 10^{15} p/cm^2)	-ΔRRR	$RRR_{fin.}$	% Change
200	316	50(50)	64	252	20
200	175	50(50)	12	163	0.74
200	316	200(200)	80	236	25
200	175	200(200)	18	157	3.1
800	316	3.0(8.62)	23	293	7.3
800	1726	3.0(8.62)	488	1238	28
1300	316	5.0(17.6)	36	280	11
1300	1726	5.0(17.6)	797	929	46
1600	316	1.5(5.52)	28	288	8.9
1600	1726	1.5(5.52)	510	1216	30

Conclusions

1. The damage rates as measured by resistivity change for energetic protons in the energy range from 200 MeV to 2 GeV are nonlinear in fluence, decreasing sharply with increasing fluence.

2. In comparing damage of particles of differing energies over this energy range, the values scale well using calculated damage-energy cross sections.

3. The percentages of measured resistivity increase scale roughly with the initial RRR's of the starting materials. Higher-percentage changes for a given damage energy is observed for higher-RRR starting material.

4. The implications of the resistivity-change results on use of niobium for superconducting cavities operating in radiation

environments and operating at 4.2 K and above is subject to several caveats. The first is that the assumption here is that the results we measure for room-temperature irradiation compare well with results that would be obtained for equal damage-energy irradiations at cryogenic temperature, followed by an anneal to room temperature. The assumption is also made that percentage changes in bulk resistivity due to the radiation are as least as large as changes in the surface resistance as defined by measurements of the cavity Q would be from equal damage increments.

For these results, the RRR = 300 material evidences less than 10% reduction for proton fluences in the $(2 - 3) \times 10^{15}$ p/cm^2 range (for all energies). One concludes, then, that this fluence over a twenty-year period of time should produce no unacceptable degradation in the cavity Q, assuming the cavities are warmed to room temperature several times over that time span. Were the cavities to be continuously maintained at cryogenic tempeatures, the equivalent damage would be produced by a fluence of $(2 - 3) \times 10^{14}$ p/cm^2, assuming 90% recovery upon warming.

If cavities are used that have a higher initial RRR, the limiting fluences above will be scaled down by the ratio of our baseline RRR = 316 to the RRR used.

For cavity operation at 2.0 K and below, the radiation damage will produce no degradation of the quality factor.

5. The results and interpretations here are consistent with the recent cavity irradiation results in France and at Los Alamos. They are inconsistent with the much earlier results at Brookhaven where large changes in Q were measured for irradiations at 2.0 K or below.

Acknowledgments

At Brookhaven we wish to acknowledge the technical assistance of E. Sperry T. Roberts, and E. Schwarz. At Saturne we appreciate the assistance of J. Faure. J-M. Lagniel, and P-A. Chamouard . From Saclay (SEA) we wish to thank M. Bolore, E. Jacques, and V. Da Ponte.

* Work performed under the auspices of the U. S. Department of Energy under Contract # DE-AC-02-76CH00016.

References

1. See for instance, Y. Kojima, "Superconducting Accelerator Cavities on a Large Scale", Trans. IEEE 0-7803, (1991), 16-20.

2. B. Rusnak, et al., Proceedings of the 1997 Particle Accelerator Conference, Vancouver, BC, May 1997, (to be published)

3. H. J. Halama, Applied Physics Lett., 19, (1971), 90 - 91.

4. H. J. Halama, IEEE Trans. Nucl. Soc. 18, (1971), 188 - 192.

5. H. Safa, private communication.

6. D. Keil, W. Decker, and J .Diehl, Radiation Effects 27, (1976), 155 - 162.

7. See, for instance, A. Sosin and W. Bauer,Studies in Radiation Effects, Vol. 3, Ed. by G. J. Dienes, Gordon and Breach Pub., (1969) 153 - 323.

8. J. Fuss and H. Schultz, Radiation Effects 40, (1979)., 181 - 186.

9. K. Faber and H. Schultz, Radiation Effects 31, (1977), 157 - 168.

10. Monroe S. Wechsler, C. Lin, and W. F. Sommer, Proceedings of the International Conference on Accelerator-Driven Transmutation Technologies and Applications, E. D. Arthur, A. Rodriguez, and S. O. Schriber, Eds., AIP Conference Proceedings 346, (1995), 466 - 475.

11. See, for instance, J. Fuss, K. Schulze, and H. Schultz, J. Phys. F: Metal Physics, Vol. 8, (1978), 497 - 500.