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High-Energy-Neutron Damage in Nb_3Sn: Changes in Critical Properties, and Damage-Energy Analysis*

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DISCLAIMER

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ABSTRACT

Filamentary wires of Nb_3Sn have been irradiated with fission-reactor, 14.8-MeV, and d-Be neutrons and the changes in critical properties measured. The changes observed scale reasonably well with the calculated damage energies for the irradiations. A critical dose for operation of these conductors in fusion-magnet applications is determined to be 0.19 eV/atom damage energy or 0.0019 dpa.

INTRODUCTION

Since the superconducting magnets used in magnetic-confinement fusion devices must operate in an energetic-neutron environment, the changes in the superconductor's physical properties to neutron damage have been of some interest. For high-field applications (above the fields where NbTi is a candidate; i.e., above ~7 T) the A15 materials hold the greatest promise at present, with Nb_3Sn the most promising. Considerable work has been performed using low temperature, room temperature, and at reactor-ambient-temperature neutron irradiation[1] with studies of the changes in properties that result and the annealing/recovery behavior investigated.

For helium-temperature irradiations at low fluences (2×10^{18} n/cm^2, E > 0.1 MeV), monotonic reduction of T_C with fluence is observed commensurate with a linear increase in the normal-state resistivity.[2] Annealing at 600 K results in partial recovery of both properties. The behavior of the critical current I_C, however, is to effect an increase for applied fields greater than ~7 T, the relative increase being greater for the higher applied fields. This increase evidenced a maximum at fluences of 0.5×10^{18} n/cm^2 (E > 0.1 MeV) and thereafter a monotonic decrease in the critical current with fluence was seen. Annealing to room temperature produced further decreases in the high-field measurements of the critical current indicating that recovery predominantly of the increases produced by the irradiation was seen. This recovery increased with field in the same way the increase of I_C with fluence did.

At ~ 10^{19} n/cm^2 I_C is less than the preirradiation value at all fields. This is the demarcation fluence between the low-fluence regime where I_C is enhanced at high fields, and the high-fluence regime where I_C is degraded below the initial values for all fields. Figure 1 summarizes this behavior of both I_C and T_C as a function of fluence for single-core Nb_3Sn irradiated at the High Flux Beam Reactor (HFBR). The low-fluence regime of I_C (increasing I_C) and the high-fluence one (decreasing I_C) are clearly evident with I_C (irrad.) < I_{CO} at about ~ 10^{19} n/cm^2 (E > 0.11

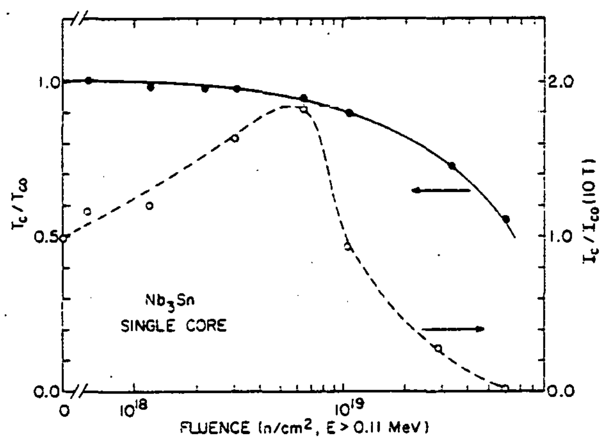

Fig. 1. The reduced critical temperature (midpoint of resistive transition) and the reduced critical current (measured at 10 T) are plotted as a function of neutron fluence (HFBR). The open circles and dashed curve are associated with the right ordinate, the close circle and solid curve with the left ordinate. The curves are drawn as guides for the eye only.

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MeV). The correlation of this behavior with that of the resistivity argues that the I_C increases with fluence are brought about through increases in the upper critical field H_{C2} which is a linear function of the normal-state resistivity.[3] The decreases in T_C are attributed to the decreased order in the damaged Nb_3Sn lattice. Neutron irradiation at reactor ambient temperature demonstrated that T_C degrades with fluence to the degree that at 1.4 x 10^{20} n/cm^2 (E > 0.11 MeV), $T_C \lesssim 3$ K, down from 18 K.[4]

There have been very few irradiations of Nb_3Sn with high-energy neutrons (defined here as from sources that give neutron energies that are greater than those typical of fission reactors). A previous report on lower-fluence 14-MeV-neutron results on some of the same samples[5,6] reported on here showed I_C, T_C, and H_{C2} behavior with fluence similar to the results from the reactor irradiations. By comparing changes in T_C of identical specimens irradiated at the HFBR at Brookhaven and the Rotating Target Neutron Source (RTNS) at Livermore, a damage-energy cross section for 14-MeV neutrons was experimentally determined for Nb_3Sn of E_D: 313^{+30}_{-30} b keV, in reasonable agreement with theoretically predicted results.

It now appears that this earlier agreement was fortuitous. A recent careful determination of the flux and spectrum in the HFBR reveal that earlier estimates (based on calculated flux and spectrum) were substantially in error. The flux above 1 MeV was found to be ~2/3 previous estimates while that below 1 MeV was more than a factor of three larger. This underscores the need for careful dosimetry in making absolute comparisons of neutron damage from different sources.

The physical properties associated with changes in superconducting behavior in A15 components (normal-state resistivity and the degree of order) have been observed to scale with damage energy in other metals and alloys when fission and fusion neutron irradiations are compared. The change in resistivity of pure metals[7-10] and the change in order of Cu_3Au[11] both scale with damage energy in fission-fusion comparisons and we expect such a correlation in Nb_3Sn.

Although it is projected that the neutron spectrum at the magnet position (in a Tokamak, for instance) will be somewhat softer than a fission spectrum with a small component of high-energy neutrons, the existence of "hot spots," or regions where the shielding is not as effective as the average, will make for an enhanced flux of the higher-energy neutron component. The problem of these hot spots has been pointed out by several authors,[12] and the consequences of enhanced fluence above projected lifetime-of-the-reactor estimates and a harder spectrum present are of concern in the materials affected. This is especially true for Nb_3Sn where end-of-reactor-life estimates of ~10^{18} n/cm^2 total fluence do not pose any problems with the critical-properties' degradation, but any part of the magnet that receives a factor of ten higher fluence, with the attendant amplification of damage energy due to a higher-energy spectrum, will suffer severe degradation of properties. The "weak link" nature of the magnet conductor then makes the operation of the magnet dependent upon the damage at the worst spot of the magnet. A quantitative knowledge of the damage-energy cross sections of high-energy neutrons in these materials and the direct consequences of high-energy-neutron damage on their critical properties are essential to the projected utilization of these materials in confinement magnets. Measurements of critical-properties' changes of Nb_3Sn following irradiations at the HFBR, RTNS, and the UC-Davis d-Be neutron source are reported to help provide this information.

EXPERIMENTAL

All of the specimens reported on here were made at Brookhaven National Laboratory by the "bronze process."[13] They consist of 19-core multifilamentary Nb_3Sn and single-filament Nb_3Sn wires. The diameter of the total wire conductors was 0.036 cm. Specimens typically 2.5 cm in length were cut from longer wire after the heat treatment to produce the A15 layer. Specimens from the same batches were used in all three irradiations reported here. Measurements of I_C were performed mainly at the Francis Bitter National Magnet Laboratory using transverse magnetic fields up to 22.5 T, but with most measurements only to 19 T. The T_C's were measured by the resistive technique (4-probe).

Irradiations at the HFBR were performed in hole V15 which has a flux of 1.3 x 10^{14} n/cm^2 sec for E > 1 MeV and 5.6 x 10^{14} n/cm^2 sec for E > 0.11 MeV. All HFBR fluences reported here are in terms of the flux for E > 0.11 MeV. The HFBR specimens were irradiated in sealed quartz tubes filled with helium gas. The temperature of the specimens is estimated to be 400 K during irradiation. For the irradiations at RTNS the specimens were placed side by side in aluminum foil forming a package ~2.5 cm long x 0.6 cm side and irradiated in air. The temperature of irradiation was room temperature. A similar procedure was employed for the specimen set for the d-Be (30 MeV d) neutron irradiation at UC-Davis with the exception that the specimens were encased additionally in Kapton so that the beam position and profile on the specimens could be determined.

Neutron fluence at the RTNS was determined from the ^{93}Nb(n,2n)^{92m}Nb reaction using a cross section at 463 mb.[14] Niobium foils 6 mm in diameter by 0.025 mm thick were placed on both the front and back of the superconductor wire packet to determine fluences.

At the UC-Davis cyclotron the low- and medium-fluence samples were irradiated at positions for which the flux and spectra had been previously determined.[15] The highest-fluence samples were part of a multiple-foil stack from which the spectrum was determined in a source-characterization experiment.[16] For both RTNS and the d-Be source the fluences are averages over a 6-mm-diameter foil at the plane of the wires. Individual wires could see fluences differing from this average value by ±15% at RTNS and ±30% at the cyclotron.

RESULTS AND DISCUSSION

A. RTNS Irradiations

The irradiation of the Nb_3Sn 19-core wires at the RTNS began in 1973. Measurements of I_C to 4 T were made on specimens irradiated to fluences of 1 x 10^{18} n/cm^2. Reductions of I_C were seen at low fields with a slight increase observed at 4 T.[6] Measurements at higher fields including results for the specimens reirradiated to 2 and 3 x 10^{18} n/cm^2 are presented in Fig. 2. Here, and in the other figures, lines are drawn through the data points only as guides for the eye. For reactor irradiation fluences below 6 x 10^{18} n/cm^2 (E > 0.11 MeV), I_C above 4 T is increased because H_{C2} is increased. Here we see that this is true for the 1 x 10^{18} n/cm^2 fluence, but for the higher fluences I_C (and H_{C2}) falls below the virgin value at all fields. These data correspond to continued irradiation of the specimens for which data were presented in ref. 6. Similar data have been taken for single-core Nb_3Sn wires and are included in the analysis.

B. d-Be Neutron Irradiations

Comparison irradiations have also been carried out using the UC-Davis d-Be (30-MeV-d) neutron source. Results of measurements of I_C following three fluences of d-Be neutron irradiation are shown in Fig. 3. For the lowest fluence, 1.4 x 10^{17} n/cm^2, there is essentially no change. For 7.0 x 10^{17} n/cm^2, increases in I_C at high field are observed along with a somewhat larger decrease in T_C and a substantial increase in H_{C2}. The data for this fluence are for either specimen, 1-3 or 1-4, as the scatter between the two sets was negligible. (These two specimens were adjacent in the sample package so that flux differences should be at a minimum.) The highest-fluence data almost duplicate the medium dose values. At first glance one might conclude that the fluences were identical. The crossover to decreasing behavior at the low fields for the higher nominal fluence, and the substantially larger decreases in T_C argue, however, that between the medium and high fluences, the high-field I_C has increased to a maximum value and has decreased back to a level comparable to that of the medium fluence result. The difference is in the derivative of the $dI_C/d\Phi$ where Φ is the

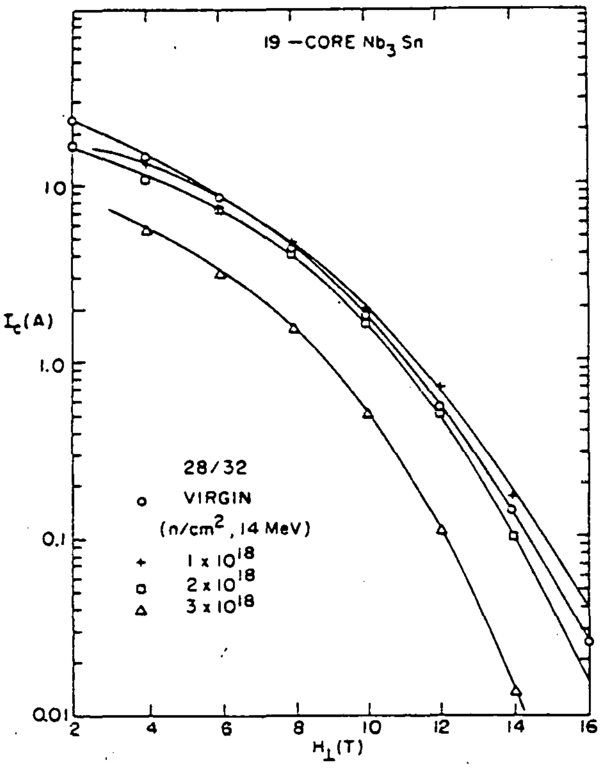

Fig. 2. The critical current of 19-core Nb_3Sn is plotted vs. applied field up to 16 T as a function of fluence up to 3 x 10^{18} n/cm^2 (14 MeV). The 1 x 10^{18} n/cm^2 fluence shows a slight enhancement of I_C at high fields, whereas the two higher fluences exhibit decreases below the unirradiated values.

fluence: positive for low and medium fluence; negative for the high fluence. In this regard the results of the d-Be irradiation are qualitatively similar to that of the 14-MeV and HFBR neutron irradiations.

C. Damage-Energy and Critical Dose Analysis

In a previous study of the changes in critical properties of Nb_3Sn by RTNs neutrons[6] we derived a value of the damage-energy cross section of 313^{+50}_{-80} b keV based on irradiations to a fluence of 2 x 10^{18} n/cm^2. This determination was based on comparing decreases in T_C with fluence for samples irradiates at the HFBR and RTNS. The T_C data were used because it was believed to be the most reliable. Further, both T_C changes and damage energy can be related to radiation-induced disorder, and the observed property changes are not multivalued as is the case with I_C. Subsequently, we have obtained more data and pushed the irradiations to higher fluence, and have obtained a more-accurate characterization of the HFBR flux and spectrum.[17]

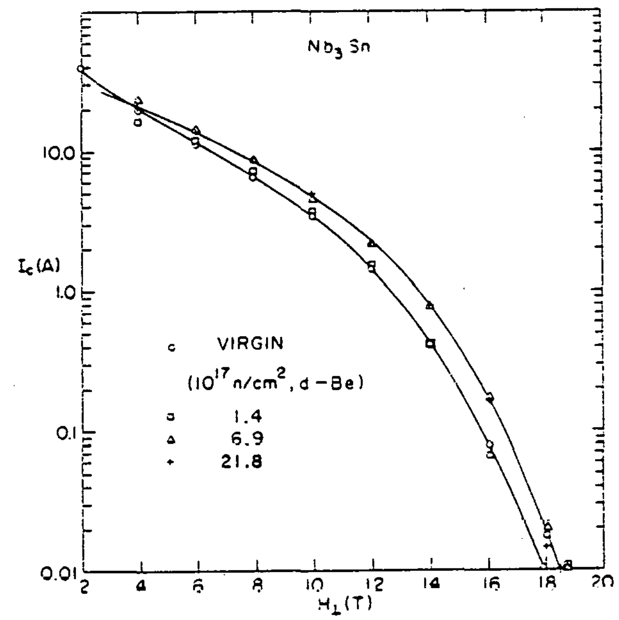

Fig. 3. The critical current of single-core Nb_3Sn is plotted vs. applied field to 20 T for 3 fluences of d-Be neutrons. The increases observed argue that the specimens are in the low-fluence regime for these nominal doses.

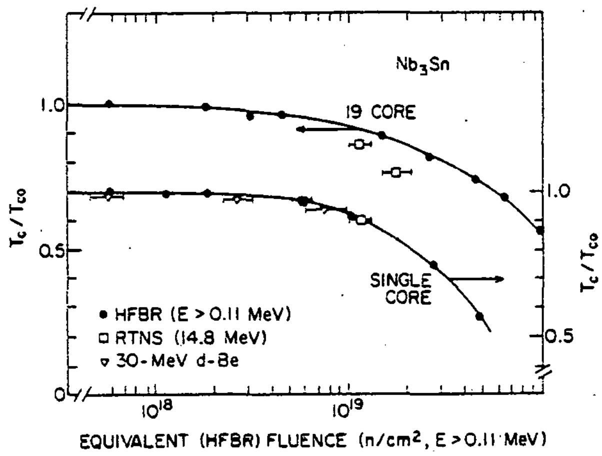

Fig. 4. The reduced T_C's (midpoint) of single-core and 19-core Nb_3Sn wires are plotted as a function of neutron fluence (HFBR). The curves are not fitted but drawn by eye. Reduced T_C values for RTNS fluences are plotted for equivalent HFBR fluences based upon damage-energy cross-section ratios of the two spectra. Horizontal error bars represent fluence uncertainties.

Figure 4 shows the reduced T_C vs. fluence results for both the 19-core and single-core Nb_3Sn for HFBR irradiations (solid circles). The open data points are for values of T_C/T_{CO} for the various 14-MeV-neutron fluences plotted for equivalent HFBR fluences calculated using the ratio of the damage-energy cross sections (the relative cross sections of Table I). The calculations in Table I were obtained using the code developed by Logan and Russell[18] which was modified to use Parkin and Coulter's[19] binary damage functions. Neutron cross sections were then taken from the ENDL (1978) file.[20] By multiplying the high-energy fluences by the relative damage-energy cross sections given in Table I we obtain the damage-equivalent HFBR fluences.

As the figure shows, the reductions in T_C with neutron fluence are somewhat greater than expected for the 19-core specimens. The single-core wires, on the other hand, exhibit T_C changes that are consistent with the disorder induced scaling with the damage energy. Although the T_C changes are small, the same scaling with damage energy is exhibited by the d-Be neutron irradiations also. The anomalously large reductions of T_C for the 14-MeV-neutron irradiated 19-core wires suggest that in those wires the damage-energy scaling underestimates the disorder produced. This inconsis-

Table I. Damage Energy Cross Sections for Nb_3Sn

Spectrum	Max. Fluence	σ_{DAM} (b keV)	Relative Damage Energy Cross section
HFBR (E > 0.11 MeV)	6.7×10^{19}	43	1.00
RTNS (14.8 MeV)	3.0×10^{18}	247	5.74
d-Be(1) (30 MeV)	1.4×10^{17}	179	4.16
d-Be(2) (30 MeV)	7.0×10^{17}	167	3.88
d-Be(3) (30 MeV)	21.8×10^{17}	147	3.42

tency between the two types of specimen is puzzling at present. It is hoped that with higher-fluence results, this problem can be solved.

The variation in derived damage-energy cross sections reflects two important factors. First are the experimental uncertainties introduced by the irradiation procedures. The RTNS source has flux gradient that are large over the sample gauge length which combine with the weak-link nature of the superconductor to cause errors in property change vs. fluence data. The second factor is the effect of stress as reflected by the difference between the 19 core and single core data. Assuming that the two data sets represent two extreme (19 core, low stress; single core, high stress) of the actual stress status in a magnet due to fabrication and magnetic stresses, the response of a real magnet conductor will include behavior spanning both sets of data with the most responsive region (fluence and stress) being the weak link.

For the engineering application in fusion magnets, a maximum permissible dose or critical dose is needed to aid in magnet design. It seems appropriate to define the critical dose ϕ_C as the fluence at which the change in I_C is zero at an applied field of 10 T $(\Delta I_C(10\ T) = 0)$, being that the maximum field at the conductor in the ETF is 11.4 T. This point occurs after I_C has increased and degradation of the superconductor has started (the point where I_C is a maximum). Using the HFBR results $\phi_C \sim 4.4 \times 10^{18}\ n/cm^2$ (E > .11 MeV). Table II shows the corresponding 14-MeV fluence, HFBR fluence for E > 0.11 MeV, and the damage energy and dpa levels. The reduction in T_C is about 4%. There are large uncertainties in the value of ϕ_C as it applies to a fusion reactor magnet spectrum due to the factors discussed for determining the damage-energy or the equivalent dose for equal property change, and the fact that the data were obtained for irradiations at 400 K not 4 K. These uncertainties are not expected to be more than a factor of two but are quite large where the implications of this error is factored into magnet-design parameters.

Table II. Critical Dose for Nb_3Sn $(\Delta I_C(10\ T)=0)$

Spectrum	Fluence
HFBR (E > 0.11 MeV)	$4.4 \times 10^{18}\ n/cm^2$
RTNS	$7.5 \pm 2.5 \times 10^{17}\ n/cm^2$
Damage Energy	0.19 eV/atom
dpa	0.0019

CONCLUSIONS

(a) Changes in critical current and critical temperature in irradiated Nb_3Sn scale with damage energy for fission-reactor, 14.8-MeV, and d-Be (30 MeV d) neutron damage.

(b) A critical dose for irreversible reduction of critical currents in Nb_3Sn is determined to be 0.19 eV/atom damage energy or 0.0019 dpa.

(c) 14.8-MeV-neutron damage appears to be greater in multifilamentary Nb_3Sn than in single-core wires, and exceeds T_C degradations predicted by scaling based upon damage-energy analysis. This aspect needs further scrutiny.

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