NEUTRON INDUCED DISORDER IN SUPERCONDUCTING A-15 COMPOUNDS*

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ABSTRACT

The effect of high energy neutron ($E > 1$ MeV) irradiation at 60°C on the superconducting critical temperature, T_c , the upper critical field, H_{c2} , the lattice parameter, a_0 , and the degree of Long Range Order has been measured for Nb and V based A-15 superconducting compounds. Large reductions in T_c and H_{c2} are observed for fluences up to 5.0×10^{19} n/cm². For Nb₃Al a_0 increases and the degree of Long Range Order is significantly reduced as T_c is depressed. The results are discussed in terms of atomic ordering in the A-15 structure.

INTRODUCTION

One important application of the A-15 compounds will be to serve as conductors in superconducting magnets for plasma confinement in future fusion reactors. In this application the magnet will be subject to neutron irradiation, the exposure depending on the power level of the reactor and the thickness of the magnet shielding. It is therefore necessary to determine the effect of neutron irradiation on the superconducting properties of these materials as such information will be required in the design of any future fusion reactor. To obtain such information experiments have been carried out in fission reactors on the assumption that to a first approximation the effects will be comparable to the environment of the magnet in a fusion reactor.**

We have determined the effects of high energy neutron ($E > 1$ MeV) irradiation at 60°C on the superconducting transition temperature, T_c , and the upper critical fields, H_{c2} , of several Nb and V based A-15 superconducting compounds. The effect of neutron irradiation on the critical current, I_c , for multifilamentary NbTi and Nb₃Sn composite conductors is presented in this volume in a separate paper.¹ We have also studied the relationship between atomic ordering and superconductivity of Nb₃Al using neutron diffraction to study the disorder produced by neutron irradiations.

EXPERIMENTAL TECHNIQUES

The samples were prepared in a variety of ways. Table I summarizes the characteristics of the samples used along with the method of preparation. For more detail concerning the methods of sample preparation appropriate references are included.

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**The yield of α particles is much lower for fission reactors.

TABLE I
Characteristics of Samples Before Irradiation

Sample	Transition Onset T_c (K)	Transition Width (90%) ΔT_c (K)	Phase Present	Lattice Parameter of A-15 Phase a_0 (Å)	Method of Preparation
Nb ₃ Sn	18.1	0.3	A-15	5.290	CVD; ref. (2)
Nb ₃ Al	18.7	0.2*	A-15+15% α Phase	5.183	Arc cast/50 hrs at 750°C
Nb ₃ Ga	20.3	0.1*	A-15+50% α Phase	5.165	Arc cast/15 min at 1850°C/48 hrs at 700°C; ref. (3)
Nb ₃ (AlGa)	19.9	0.6	A-15+	5.165	ref. (4)
Nb ₃ Ga	6.5	0.5	Phase		Arc cast
V ₃ Si	18.9	0.2	A-15		ref. (4)
V ₃ Ga	12.6	2.0	A-15		Multifilamentary wire; 100 hrs at 650°C; ref. (5)

*50% of transition width. 90% transition at 16.9 K.

**50% of transition width. Sample has a tail to 15.9 K.

The irradiations were carried out in the Brookhaven National Laboratory High Flux Beam Reactor (HFBR) where the samples were exposed to the whole spectrum of the reactor.⁶ In the following only the fast flux ($E > 1$ MeV) will be considered. The calculated temperature of the samples during irradiation was $60 \pm 5^\circ$ C. Superconductivity was detected by a standard mutual inductance technique operating at 37 Hz, the temperature being determined by a calibrated carbon thermometer, the error in temperature measurement being $\pm 0.2^\circ$ K. The upper critical field, H_{c2} , was measured in a manner previously described.⁷ The degree of Long Range Order (LRO) for unirradiated and irradiated Nb₃Al was determined by neutron diffraction techniques on powdered samples. A least squares computer fit, including anisotropic Debye-Waller temperature factors, to the observed intensities was used to determine the LRO parameter to $\pm 1.5\%$. The lattice parameters of unirradiated and irradiated samples were determined from x-ray powder patterns using a 114.5 mm Debye-Scherrer camera, specially adapted to handle radioactive powders. The accuracy of the lattice parameters are ± 0.001 Å. A complete description of the (LRO) experiments will be presented in a subsequent publication.⁸

EXPERIMENTAL RESULTS

Transition Temperature

Figure 1 shows the results for the transition temperature as a function of fluence for the systems studied. The data are plotted as the reduced transition temperature, T_c/T_{c0} , where T_c is the transition temperature after irradiation and T_{c0} its value before, given in Table I. We see that the depression of T_c for the A-15 compounds studied here can be approximately described by a universal curve of the type shown. Up to a fluence of $\sim 10^{18}$ n/cm² the depression of T_c , relative to its unirradiated value, is about 5%. Above this value T_c is rapidly depressed reaching values of 84.5% and 83.3% for Nb₃Sn and Nb₃Ga respectively for fluences of 5.0×10^{19} n/cm². For Nb₃Al no superconductivity was observed down to 1.4° K for fluences of 1.4×10^{19} n/cm², 1.9×10^{19} n/cm² and 5.0×10^{19} n/cm². In a previous work,⁹ we observed a transition at 3.5° K for arc cast but unannealed Nb₃Al irradiated

Fig. 1 Reduced superconducting transition temperature, T_c/T_{c0} against fluence, $nvt(E > 1 \text{ MeV})$. Arrows indicate sample was normal to temperature shown. Curve is drawn as usual aide.

to a fluence of $5.0 \times 10^{19} \text{ n/cm}^2$. It thus appears that the depression of T_c for annealed Nb_3Al is more sensitive to neutron irradiation than unannealed Nb_3Al . The transition widths after irradiation were of the same order as before irradiation, with the exception of V_3Si whose width increased from 0.15°K to 3.5°K . This broadening for V_3Si is not understood and more irradiations are planned.

The unirradiated value of T_c can be recovered in the irradiated samples by annealing. Figure 2 shows the recovery of T_c for Nb_3Sn and Nb_3Ga irradiated to $5.0 \times 10^{19} \text{ n/cm}^2$. Nb_3Al , which was normal to 1.4 K after an exposure of $5 \times 10^{19} \text{ n/cm}^2$ increased its T_c to 15.9°K after a 10 minute anneal at 750°C . After 20 minutes its T_c was 16.2°K and after 80 minutes T_c had a value of 17.7°K .

Upper Critical Field

The temperature dependence of the upper critical field is shown in Fig. 3 for Nb_3Sn , Nb_3Ga and arc cast Nb_3Al irradiated to $5.0 \times 10^{19} \text{ n/cm}^2$. The temperature dependence of the critical field for Nb_3Al and Nb_3Ga are linear in the range from T_c to 1.5°K , the lowest temperature available. The value of T_c obtained by extrapolating the critical field curves to $H=0$ agrees well with the measured T_c obtained by the mutual inductance technique. H_{c2} for Nb_3Sn , Nb_3Al and Nb_3Ga was $300 \pm 100 \text{ G}$ at 1.6°K , $4700 \pm 50 \text{ G}$ at 1.6°K and $7700 \pm 50 \text{ G}$ at 1.5°K respectively. For purposes of comparison, H_{c2} at 4.2 K for unirradiated Nb_3Sn , Nb_3Al and Nb_3Ga is 235 , 295 , 340 kG respectively.^{10,11} This large reduction in H_{c2} upon irradiation is most likely due to the very low T_c 's of the samples.

Long Range Order and Lattice Parameter

We observe a considerable reduction in the degree of LRO for Nb_3Al upon irradiation. For a fluence of $1.2 \times 10^{19} \text{ n/cm}^2$ the LRO is reduced by 18% relative to its unirradiated value. At the same time T_c is depressed from 18.7°K to 9.6°K , a 48.6% reduction. The lattice parameter increases from $5.183 \pm 0.001 \text{ \AA}$ for unirradiated Nb_3Al to $5.200 \pm 0.001 \text{ \AA}$ for a fluence of $5.0 \times 10^{19} \text{ n/cm}^2$, a 0.33% expansion. A full description of the LRO and lattice parameter experiments will be reported separately.⁸

Fig. 2 Superconducting transition temperature, T_c , against annealing time, T , at 750°C for Nb_3Sn and Nb_3Ga irradiated to $5.0 \times 10^{19} \text{ n/cm}^2$.

Fig. 3 Upper critical fields, H_{c2} , against temperature, T , for Nb_3Sn , Nb_3Ga and Nb_3Al irradiated to $5.0 \times 10^{19} \text{ n/cm}^2$.

DISCUSSION

Our results show the superconducting properties of high T_c Nb and V based A-15 compounds are degraded when exposed to high energy neutron radiation at 60°C . An apparent threshold at 10^{18} n/cm^2 for a 60°C irradiation with fast neutrons exists in the T_c vs fluence curve above which T_c is rapidly depressed. This kind of information will have a bearing on the designs for CTRs when these materials are used for

containment of the plasmas. Data on low temperature irradiations are clearly needed as the magnets will be operating at cryogenic temperatures. However, it seems unlikely that the threshold value of 10^{18} n/cm² will be shifted to higher fluences for low temperature irradiations as the neutron induced defects that cause the degradation of the superconducting properties are stable to at least 60°C and probably higher. It's quite possible that degradation will take place more rapidly at low temperatures due to the stability of vacancy-interstitial pairs that anneal out at higher temperatures. It should be noted that low temperature facilities with sufficient neutron fluxes are not yet available because of the gamma heating produced in the cryostats.

We believe that the depressions of T_c observed here are primarily the result of rearrangements of the atoms in the unit cell brought about by displacement collisions between the target atoms and the incident high energy neutrons. This results in a decrease in the degree of LRO, which lowers T_c . The results of the LRO measurements on neutron irradiated Nb₃Al clearly support this model. In an earlier work we calculated the initial depression rate of T_c as a function of Nb sites occupied by Sn atoms for neutron irradiated Nb₃Sn based on a proposed radiation damage model. We obtained a value for the initial depression rate of 3.3°K for a 1% replacement of Nb sites by Sn atoms. On the basis of our measured LRO parameter for Nb₃Al we find a depression rate of 2.2°K for a 1% replacement of Nb sites by Al atoms. These depression rates are quite large and are usually associated with magnetic impurities in superconductors. The underlying mechanism for the destruction of superconductivity in this case, however, is not known and heat capacity measurements are planned to shed some light on this question.

The lattice expansion observed can be explained on the basis of the neutron induced disorder. As the radius of the Nb atoms in the A-15 structure is larger than that of the Al atoms, 1.51 Å as compared to 1.39 Å,¹² the lattice expands as the larger Nb atoms begin to occupy the Al sites. The disorder mechanism also gives some insight into the different recovery rates for Nb₃Sn and Nb₃Ga upon annealing. The radius of Sn in the A-15 structure, 1.41 Å,¹² is closer to that of Nb than that of Ga, 1.38 Å.¹² Thus when a Nb atom is interchanged with a Ga atom, more lattice strain results than when a similar exchange takes place with a Sn atom. This excess strain energy provides the driving force for the reordering to occur. The greater the strain energy difference the faster will be the reordering and hence the recovery of T_c at a given temperature. The Al radius is intermediate between that of Sn and Ga and the recovery time for Nb₃Al is intermediate between that of Nb₃Sn and Nb₃Ga consistent with the above reasoning.

SUMMARY

Our results can be summarized as follows:

1. The superconducting properties of high T_c A-15 compounds are degraded when exposed to high energy neutron irradiation at 60°C.
2. An apparent threshold fluence of $\sim 10^{18}$ n/cm² is required to bring about severe depressions in T_c at an irradiation temperature of 60°C. Below this value T_c is depressed about 5% relative to its unirradiated value, above this value depressions of

80% are observed and in some cases no superconductivity is observed at all for fluences in the 10^{19} n/cm² range.

3. H_{c2} is severely depressed for fluences above 10^{19} n/cm², being only a small fraction of its unirradiated value.

4. T_c depressions are most likely caused by neutron induced disorder of the lattice. For Nb₃Al, direct measurement indicates a substantial reduction in the degree of atomic ordering due to neutron irradiation. A significant increase in the lattice parameter of neutron irradiated Nb₃Al is also observed.

5. These results must be considered in future CTR design utilizing these materials as conductors for magnets.

6. Neutron induced disorder can serve as a useful technique in the study of the effects of atomic ordering on superconductivity.

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