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Proceedings

Manuscript Draft

Manuscript Number: NIMB PROCEEDINGS-D-16-00529

Title: Effect of annealing high-dose heavy-ion irradiated high-temperature superconductor wires

Article Type: SI: NIMB IBMM 2016

Section/Category: SI: NIMB IBMM 2016

Keywords: superconductor; flux pinning; critical current; ion irradiation; YBCO

Corresponding Author: Dr. Nicholas Strickland,

Corresponding Author's Institution: Victoria University of Wellington

First Author: Nicholas Strickland

Order of Authors: Nicholas Strickland; Stuart C Wimbush, PhD; John V Kennedy, PhD; Patrick Kluth, PhD; Pablo Mota-Santiago; Mark C Ridgway, PhD; Nicholas J Long, PhD

Abstract: Heavy-ion irradiation of high-temperature superconducting thin films has long been known to generate damage tracks of amorphized material that are of close-to-ideal dimension to effectively contribute to pinning of magnetic flux lines and thereby enhance the in-field critical current. At the same time, though, the presence of these tracks reduces the superconducting volume fraction available to transport current while the irradiation process itself generates oxygen depletion and disorder in the remaining superconducting material. We have irradiated commercially available superconducting coated conductors consisting of a thick film of (Y,Dy)Ba2Cu307 deposited on a buffered metal tape substrate in a continuous reel-to-reel process. Irradiation was by 185 MeV 197Au ions. A high fluence of 301011 ions/cm2 was chosen to emphasize the detrimental effects. The critical current was reduced following this irradiation, but annealing at relatively low temperatures of 200°C and 400°C substantially restore the critical current of the irradiated material. At high fields and high temperatures there is a net benefit of critical current compared to the untreated material.

Effect of annealing high-dose heavy-ion irradiated hightemperature superconductor wires

N. M. Strickland^a, S. C. Wimbush^a, P. Kluth^b, P. Mota-Santiago^b, M. C. Ridgway^b, J. V. Kennedy^c, and N. J. Long^a

^a Robinson Research Institute, Victoria University of Wellington, Wellington 6140, New Zealand

^b Research School of Physics and Engineering, Australian National University, Canberra, ACT 2601, Australia

^c GNS Science, Lower Hutt 5010, New Zealand

Abstract

Heavy-ion irradiation of high-temperature superconducting thin films has long been known to generate damage tracks of amorphized material that are of close-to-ideal dimension to effectively contribute to pinning of magnetic flux lines and thereby enhance the in-field critical current. At the same time, though, the presence of these tracks reduces the superconducting volume fraction available to transport current while the irradiation process itself generates oxygen depletion and disorder in the remaining superconducting material. We have irradiated commercially available superconducting coated conductors consisting of a thick film of (Y,Dy)Ba₂Cu₃O₇ deposited on a buffered metal tape substrate in a continuous reel-to-reel process. Irradiation was by 185 MeV ¹⁹⁷Au ions. A high fluence of 3×10¹¹ ions/cm² was chosen to emphasize the detrimental effects. The critical current was reduced following this irradiation, but annealing at relatively low temperatures of 200°C and 400°C

substantially restore the critical current of the irradiated material. At high fields and high temperatures there is a net benefit of critical current compared to the untreated material.

Introduction

High-temperature superconductor (HTS) wires are now a mature technology with multiple companies capable of producing long lengths of high-quality conductor having a low frequency of defects. The second-generation HTS wires based on REBa₂Cu₃O₇ (REBCO, where RE = rare-earth or yttrium) coated conductors in particular are produced through a number of disparate methodologies that generate a variety of microstructures. The superconducting critical current (I_c) that can be achieved in the presence of a magnetic field is an extrinsic property that depends on magnetic-flux pinning by defects in the HTS material. Controlling the density and morphology of defects in the REBCO films is therefore one of the central tasks required to achieve optimized conductors. Point-like, linear, planar and threedimensional defects can all be generated through secondary phase inclusions or growth defects, with the specific growth conditions often determining the dimensionality of these defects.

Strong flux-pinning centers can also be introduced after the growth of the HTS material by heavy-ion irradiation. The ions themselves do not play an active role, usually passing all the way through the film; rather it is the damage tracks consisting of columns of amorphized material that subsequently act as pinning defects. The damage tracks have radii up to a few nanometres [1]–[4] and can be continuous or discontinuous depending on the electronic energy loss of the ion [2][3]. The radius of the damage tracks is similar to the coherence length in the *a-b* plane which makes them near ideal pinning centers for magnetic fields oriented parallel to the irradiation direction, thus significantly enhancing the in-field I_c [1][4][7]. These damage tracks are of great interest in fundamental studies of flux pinning as they can be introduced into a wide range of samples with different initial microstructures independently of the fabrication methodology or conditions, and they can be well controlled with respect to density and to a lesser extent with respect to continuous section length.

The I_c enhancement is most prominent when the magnetic field is applied parallel to the damage tracks [4],[6]–[9]. On the other hand, a high density of irradiation tends to reduce current percolation and the superconducting transition temperature T_c , with the result that I_c can often be reduced for low magnetic fields or for magnetic fields applied in other directions [7],[8],[10]. A splay in the direction of the tracks, or a combination of multiple irradiation directions, can be used to widen the angular range over which I_c is enhanced [4],[5][8][11]. The angular range of benefit also tends to widen for shorter, more discontinuous tracks [12]–[14] as these better preserve current percolation and accommodate meandering flux lines.

Systematic studies of irradiation are facilitated by the commercial availability of long-length coated conductor wires with very consistent properties along the length. It is pertinent to note that these conductors have already been engineered for high flux pinning and any improvements in I_c demonstrate the headroom available for performance increases using strong engineered pinning centers. Recently, it has been demonstrated that long lengths of conductor can be irradiated in-line at energies and rates that make incorporation of an ion irradiation treatment into an industrial process plausible [15]–[17].

The anisotropy of flux pinning with respect to the magnetic field orientation can be measured conveniently with a transport method. This method shows very clear enhancements of I_c when the field is oriented parallel to extended planar or linear defects, including ion damage tracks [7],[8],[18]. I_c is often measured at 77 K due to the relative simplicity of making measurements in liquid nitrogen and due to the high relative effectiveness of flux pinning at temperatures near T_c . This does have the disadvantage of making the results very sensitive to any reduction in T_c arising from the irradiation treatment.

We have previously shown that annealing samples after irradiation can partially restore T_c while still maintaining flux pinning by damage tracks, thereby increasing I_c at 77 K [8]. In this

work we extend this result and show that I_c of a heavily irradiated sample can be improved even at 30 K through annealing for short times at relatively low temperatures.

2 Experimental

The virgin coated conductor samples were cut from a long length of wire insert taken from the HTS wire production line at American Superconductor Corp. (Devens, MA) [19] prior to lamination. In these coated conductors, a 1.4 μ m thick (Y,Dy)BCO layer was deposited by metal-organic deposition on a Ni-5at.%W foil substrate buffered with 75 nm thick layers of Y₂O₃, Y₂O₃-stabilized ZrO₂ and CeO₂. Crystallographic texture was imparted to the substrate by the RABiTS deformation-annealing process and the buffer and HTS films grew epitaxially on that surface, with the *c*-axis of (Y,Dy)BCO lying perpendicular to the substrate surface. A 1 μ m thick Ag capping layer was deposited on top of the HTS layer. The process conditions were those of the standard coil wire product of American Superconductor, which optimizes the conductor for high *I*_c at 20–40 K and 2–5 T.

Irradiation with 185 MeV ¹⁹⁷Au ions was performed at the Australian National University's Heavy Ion Accelerator facility. In this work the irradiation was performed with the ion beam directed perpendicular to the surface, so the damage tracks were on average parallel to the (Y,Dy)BCO *c*-axis. An irradiation time of 2 minutes was sufficient to accumulate a total fluence of 3×10^{11} ions/cm².

The Ag capping layer on top of the HTS is somewhat thinner than that usually employed in the American Superconductor process to facilitate penetration by irradiating ions with minimal energy loss. The capping layer significantly reduces the energy of the irradiating ions entering the HTS film thus affecting the average electronic energy loss and the damage track morphology. In Fig. 1(a) the lower dashed curve depicts the energy profile of the Au ions entering at 185 MeV and passing through silver, as calculated using SRIM software [20].

After 1 μ m passage through Ag, the energy drops to 145 MeV. In our earlier work using the same ions and a similar coated conductor, the Ag layer was instead the more standard 2.6 μ m thickness and in that case the Au ion energy drops to 90 MeV before entering the YBCO layer. The two solid line segments represent the subsequent passage of the ions through the 1.4 μ m thick YBCO film. These energies then translate to a range of electronic energy loss *S*_e in the HTS layer as per Fig. 1(b); for the 1 μ m Ag cap used in this work *S*_e ranges from 28.5 keV/nm to 24.5 keV/nm while for the 2.6 μ m cap used previously *S*_e ranges from 22.5 keV/nm to 17.5 keV/nm. We therefore expect that the damage tracks should be nearly continuous for the 1 μ m cap used in this work while they would be more discontinuous for the 2.6 μ m cap of the previous work [3].

Post-irradiation annealing was carried out in a tube furnace with an O_2 atmosphere at 200°C and 400°C for 1 h each. The sample was removed from the hot zone of the furnace at the completion of the anneal and allowed to cool rapidly in the O_2 atmosphere.

 I_c was measured using a four-probe transport method in a bespoke measurement system [21]. This is a fully-cryocooled system with an HTS dipole magnet and gas-cooled insert providing sample temperatures to below 15 K. Automated transport I_c measurements can be made at currents up to 875 A. Measurements were made in the usual maximum Lorentz force configuration with the magnetic field applied perpendicular to the transport current direction and using the standard electric field criterion of 1 μ V/cm for I_c . The measurement region was defined by a 5 mm × 0.5 mm current-transport bridge created using photolithography and wet chemical etching. Values for I_c plotted in this paper are normalized to unit width of conductor, *w*. The angular convention adopted has 0° perpendicular to the plane of the sample (parallel to the *a-b* planes).

Results

The temperature dependence of the self-field (zero applied field) I_c is shown in Fig. 2 for a single sample before irradiation, after irradiation, and after each of two successive anneals. The irradiation fluence of 3×10^{11} ions/cm² corresponds to a matching field (where the number density of quantized magnetic flux lines equals the number density of irradiating ions) of $B_{\phi} = 6.2$ T. This fluence is therefore expected to be greater than the optimum for fields in the range of 0-5 T measured here and was chosen to clarify the benefit of annealing. Self-field I_c is not expected to benefit at all from irradiation since flux pinning is not the determining mechanism in this regime, and in general will decrease at high fluences due to reductions in both current percolation (superconducting cross section) and T_c resulting from oxygen depletion and disorder. Both of these effects are apparent in Fig. 2. After irradiation T_c drops from 87.5 K to 80.5 K and in addition the variation in I_c with respect to temperature is reduced by a factor of 3–4. Annealing at just 200°C is enough to partially recover T_c up to 83 K and increase I_c over the full temperature range. A subsequent anneal at 400°C further improves T_c and $I_{\rm c}(T)$. The significant recovery of $T_{\rm c}$ following a low-temperature anneal is evidence that the $T_{\rm c}$ reduction arises from disorder of the oxygen ions in the lattice that is widespread and not limited to the damage tracks. Presumably this arises effectively from localized heating during irradiation. 200°C annealing would normally be associated with short-range motion of the oxygen ions only, and not the cations, so has the potential to be beneficial in repairing the disordered (Y,Dy)BCO without reversing the amorphization in the damage tracks. $I_{c}(T)$ remains well short of the original pre-irradiation values, which suggests that at this fluence and energy the density of columnar defects is approaching a percolation limit.

The field-dependences of I_c for fields applied perpendicular to the sample, and therefore parallel to the *c*-axis and the damage tracks, at 70 K and 30 K are shown in Fig. 3. The 70 K data shows the effect of being close to T_c with the low-field I_c diminished by a factor 10

following irradiation. Even at 5 T I_c is diminished for this fluence. Annealing at 200°C increases I_c significantly, by almost a factor 2 at 3–4 T. At 5 T I_c of the annealed sample is higher than the virgin sample despite the clear reduction in percolation implied by the low-field data. A 400°C anneal continues the improvement at all fields, suggesting that annealing at 200°C for 1 h was not sufficient to completely re-order the oxygen ions and annealing at 400°C is not so much as to remove the pinning benefit of the damage tracks through recrystallisation. The 30 K data is remarkable in that for fields above 2 T there is essentially no change in I_c following irradiation. Our earlier work [10] demonstrated that I_c enhancements could be achieved in this temperature / field regime at a lower irradiation fluence, so it is likely that this lack of change is a coincidence of the balance between reduction in percolation and enhancement in flux pinning. After annealing at 200°C and 400°C, I_c remains unchanged at the higher fields, despite there being some improvement in the low-field regime.

The angle dependences of I_c at 1 T, 30 K and 1 T, 70 K are shown in Fig. 4(a). At this field, for all temperatures and angles shown the I_c is significantly reduced by irradiation and partially but never completely recovered by annealing. The relative benefit for fields applied parallel to the damage tracks versus fields applied perpendicular to those tracks is clear. At 70 K the virgin conductor was almost isotropic, but after irradiation prominent peaks appear for fields parallel to the tracks (0° and 180°). Similarly at 30 K where the virgin sample had a sizeable "conventional" anisotropy with a peak parallel to the *a-b* plane, after irradiation it becomes completely isotropic. In both fields, annealing increases I_c approximately uniformly with little change in the anisotropy. The same angle dependences of I_c at 5 T are shown in Fig. 4(b). In this case at 70 K we see a slight enhancement over the virgin sample for fields applied parallel to the damage tracks, corresponding to that seen in Fig. 3. The angle dependence emphasizes that this enhancement is restricted to a very small region of the temperature–field–angle parameter space. At 30 K we have already seen that irradiation and

 annealing had no net effect for fields applied parallel to the damage tracks in the 5 T range. Here we see that there is, nevertheless, a reduction and then recovery in I_c for fields applied perpendicular to the tracks. This produces an unusual shape of the angle-dependence I_c and an unusually low anisotropy for this temperature and field regime. With the field applied perpendicular to the damage tracks (90°) the changes in I_c tend to mirror those of the self-field I_c across all the temperatures and fields shown here.

Annealing at 200°C and 400°C results in significant recovery of I_c over the whole parameter space except for one corner, that of low temperature, high field parallel to the damage tracks, where I_c remains unchanged. This suggests that this annealing regime is effective in reordering the (Y,Dy)BCO lattice without recrystallizing the amorphized damage tracks.

4 Conclusions

We have investigated heavy-ion irradiation of state-of-the-art (Y,Dy)BCO coated conductors with 185 MeV Au ions at a high fluence of 3×10^{11} ions/cm². For this ion energy and for the thin 1 µm Ag capping layer on our coated conductors this level of fluence is too high to obtain a significant net I_c improvement below 5 T at any temperature. This is not surprising since the fluence corresponds to a matching field of 6.2 T. It might be expected that greater enhancements would exist for fields higher than that. Pinning enhancements most likely have occurred, but are countered by the reduction in T_c due to oxygen depletion and disorder and by reduction in current percolation through the destruction of too much superconducting material. Annealing at just 200°C for 1 h mostly improves, but at least always retains, the I_c of the irradiated material over the wide parameter space tested: T = 30 - 77 K, B = 0 - 5 T, $\theta = 0$ $- 180^\circ$. We interpret this as restoring oxygen ordering in the otherwise still ordered (Y,Dy)BCO in the regions between the ion damage tracks. Oxygen disorder presumably arises as a result of rapid localized heating. Annealing at 400°C for 1 h continues the trend and, again, does not reduce I_c in any part of the parameter space tested here. Therefore we

suggest that the amorphous ion damage tracks remain stable on this time scale at that temperature. I_c of the irradiated material improves by up to a factor of 2 after the two annealing processes, indicating that annealing should always be an important part of optimising ion-irradiated HTS wires.

Acknowledgments

The authors wish to thank M. Rupich of American Superconductor for supplying the coated conductor material for our irradiation trials and for useful discussions. PK and MCR thank the Australian Research Council for financial support. We acknowledge access to the Heavy Ion Accelerator Facility at the Australian National University funded through the NCRIS grant.

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Figures



Figure 1. (a) Reduction in Au ion energy on passage through silver and YBCO, highlighting the relevant energy ranges of the ions in this and earlier work. (b) Electronic energy loss in dependence on Au ion energy, again highlighting the ranges of energy loss in this and earlier work.



Figure 2. Self-field critical current per unit width as a function of temperature for a single sample in the pristine state, after irradiation, and after two successive anneals.



Figure 3. Field dependences of the critical current per unit width at 30 K and 70 K, for fields applied perpendicular to the sample, of a single sample in the pristine state, after irradiation, and after two successive anneals.



Figure 4. Field angle dependences at (a) 1T, and (b) 5 T of the critical current per unit width at 30 K and 70 K of a single sample in the pristine state, after irradiation, and after two successive anneals.