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Abstract—We demonstrate a 2-fold increase of the in-field critical current of AMSC’s standard 2G coil wire by irradiation with 18 MeV Au ions. The optimum pinning enhancement is achieved with a dose of 6×10^{11} Au ions/cm². Although the 77 K, self-field critical current is reduced by about 35%, the in-field critical current (H/c) shows a significant enhancement between 4 – 50 K in fields >1 T. The process was used for the roll-to-roll irradiation of AMSC’s standard 46 mm wide production coated conductor strips which were further processed in to standard copper laminated coil wire. The long length wires show the same enhancement as attained with short static irradiated samples. The roll-to-roll irradiation process can be incorporated in the standard 2G wire manufacturing with no modifications to the current process. The enhanced performance of the wire will benefit rotating machine and magnet applications.

Index Terms— Critical current density, flux pinning, high-temperature superconductors, irradiation, roll-to-roll processing.

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I. INTRODUCTION

SECOND GENERATION (2G) high temperature superconducting wires, based on the (RE)Ba₂Cu₃O_{7- δ} (RE = rare earth elements) materials, are being developed for a number of electric power applications and are expected to provide significant benefits to a range of applications [1]-[3]. The major anticipated applications include cables that operate at high temperatures (65 - 77 K) and relatively low fields and rotating machines that operate at a temperature of 30 - 50 K range in fields of 1 – 4 T. Although the performance of the production length 2G wire is nearing the level required for commercial cable applications, the severe drop in critical current density (J_c) in magnetic fields limits its use in commercial rotating machines. Thus increasing the current carried by second generation (2G) high temperature superconducting (HTS) wire in the presence of high magnetic fields is critical for the commercialization of HTS based rotating machine applications (generators for off-shore wind turbines, hydro power and utility applications; compact, light-weight marine propulsion; etc.) and various HTS magnet applications.

Research over the past few years has shown that pinning landscapes based on mixed defect structures are effective in improving the 2G HTS wire performance [4]. Although mixed pinning structures based on self-assembled BaZrO₃ columns and nano-particle defects have been demonstrated in short samples of vapor phase grown (RE)BCO films, it has been difficult to uniformly incorporate these highly engineered defect structures into production length conductors, resulting in significant variation in the critical current (I_c) along the wire length at the operating conditions. This is critically important for commercial 2G wires since performance of the entire long length wire is limited by the section with the lowest performance. Additionally, these self-assembled columnar structures are not viable options for solution-grown REBCO films using metal organic deposition (MOD). Thus we have been exploring alternative routes to fabricate precisely engineered defect structures into production length MOD-based REBCO films.

Nano-particles of various compositions can be introduced into the MOD-based films and have provided significant improvements in the pinning microstructure. The current state-of-the-art MOD REBCO-based 2G wire incorporates a mixed pinning landscape consisting of a dispersion of nano-particles of various sizes, stacking faults and dislocations. Although the development of this pinning microstructure has resulted in a near doubling of in-field J_c in the 20 – 50 K range, the I_c level of actual wires is at least a factor of 2 below that required

for commercial rotating machine applications. Research over the past year has shown that the addition of point defects, produced by proton irradiation, significantly improves the pinning in the state-of-the-art MOD-based REBCO films, with the critical current (I_c) more than doubling at ~ 30 K in magnetic fields > 2.5 T [5]. This earlier work used 4 MeV protons that produced additional ~ 5 nanometer size defects in the existing microstructure, but required long exposure times. Similar performance enhancements have been achieved with much shorter exposure times with 4 – 14 MeV oxygen ions [6]. These results demonstrate these nanometer sized defects are highly effective at pinning vortices in high fields in MOD-based REBCO films. However, they are only relevant to high volume production of 2G wire if the ion species and energies can be optimized to allow very short exposure times.

In this presentation, we will describe the pinning enhancements achieved in AMSC's production wire using Au ions with an energy around 16 MeV. The J_c enhancement was similar to that achieved with the proton or O irradiation. In contrast to proton or O irradiation, the dosages required to achieve the equivalent pinning enhancement were significantly lower, making roll-to-roll irradiation of production length conductors viable using readily available electrostatic generators.

II. IRRADIATION PROCESS

A. Experimental Details

The coated conductor samples used in this study were taken from AMSC's 2G HTS wire production line. The samples consisted of a 75 μm thick Ni(5at%)W RABiTSTM substrate with a buffer stack consisting of 75 nm each of Y_2O_3 , YSZ and CeO_2 deposited by reactive sputtering and a 1.2 μm thick YBCO layer, doped with Dy_2O_3 , deposited by an MOD process. Approximately 1 μm of Ag was deposited on both sides of the tape. The samples were processed as AMSC's coil wire formulation which has a 77 K, self-field I_c of around 385A/cm-w and enhanced pinning at lower temperatures. Samples were cut from the 46 mm wide strip after the oxygenation step in AMSC's RABiTS/MOD-YBCO manufacturing process [7].

The ion irradiations were carried out at Brookhaven National Laboratory using the tandem Van de Graaff Facility which consists of two 15 MeV electrostatic accelerators. The ion beam consisted of Au ions with either a 5+ or 6+ charge with an energy of 16 – 18 MeV. The Au ions were generated from a sputtering source and the ion beam had a particle current of approximately 120 nA at the sample. The samples were irradiated with the ion beam oriented along the crystallographic c -axis of the YBCO.

Short samples were irradiated by moving the sample under a static beam for a set time to achieve the targeted exposure. 46 mm wide, long length production strips were irradiated in a roll-to-roll configuration by rastering the ion beam across the width of the moving strip at a frequency of about 40Hz. The line speed of the tape was varied to achieve the targeted exposure.

Critical currents were measured using a 4-point transport technique with 1 $\mu\text{V}/\text{cm}$ voltage criteria.

The normalized relaxation rates (flux creep rate) were measured in a commercial SQUID magnetometer on small square samples [8]. The rates were obtained from the time decay of M_{irr} over a period of 1 hour at fixed temperature and field, after the field was appropriately cycled to create the initial fully penetrated critical state.

B. Short Sample Irradiation

Some of the earlier irradiation studies were carried out on YBCO films with no Ag coating or at most an extremely thin layer [9]. However, studies have shown that the post annealing of the samples at temperatures above 300°C essentially removes the defects from the YBCO film. Thus if the irradiation technique is to be considered for a manufacturing process, it must be carried out after the Ag layer deposition and oxygenation steps, since these processes generally require temperatures in excess of 300°C. Thus the energy of the ions must be high enough to fully penetrate the Ag layer and still have sufficient energy to cause displacements uniformly through the entire thickness of the YBCO layer.

SRIM-TRIM calculations [10] for Au ions indicate that Au ions with an energy of around 16 – 18 MeV are required to uniformly irradiate a 1.2 μm thick YBCO layer through a 1 μm thick Ag layer. If a thicker Ag layer is used, a higher energy for the Au ions is required.

Previous irradiation studies of AMSC coated conductor tapes using either H or O ions showed that a dosage of 2×10^{17} H/cm² and 1×10^{13} O/cm² was required to achieve the maximum pinning enhancement in moderate fields at around 30 K. However, SRIM calculations indicate that Au ions will produce significantly more displacements per collision than either O or H. Thus the required dosage should be significantly less.

Our initial studies were carried out on 1 x 2 cm coupons using Au⁵⁺ ions with an energy of 18 MeV. Samples were exposed to the ion beam for various times to achieve a dose of 1×10^{11} Au/cm² to 2×10^{12} Au/cm². Fig. 1, which plots I_c as a function of magnetic field (H/c) at 27 K for the different doses of Au ions, shows that a maximum enhancement is attained around 6×10^{11} Au/cm². Fig. 2 shows that the I_c enhancement for the optimum dose relative to the unirradiated sample at 30 and 77 K. At low fields and high temperature, the I_c of the irradiated sample is reduced; however, at temperatures and fields relevant to rotating machine applications the I_c enhancement is over 2-fold and increases with increasing field up to at least 6 T.

In order to establish whether similar enhancements can be achieved on a moving sample, the coupon sample holder was modified so that a 1 m x 1 cm tape could be transported through the static ion beam. The tape speed was adjusted to achieve the optimal dose of 6×10^{11} Au/cm². Before the experiment, the beam was shaped and the xy profile determined to establish the irradiation area and beam intensity. Fig. 2 also shows that the I_c enhancement measured on the moving sample was nearly identical to that obtained on the

short static samples.

Fig. 1. I_c as a function of magnetic field (H//c) at 27K for the different doses of Au ions.

Fig.2. I_c enhancement at 30 and 77K (relative to an unirradiated control sample) of a stationary short sample and a moving 1 meter tape after irradiation with 18 MeV Au to a dose of 6×10^{11} Au/cm²

Figure 3 shows the normalized relaxation rate, $S(T)$, for a sample irradiated with 18 MeV Au ions to a dose of 6×10^{11} ions/cm² and two unirradiated samples, for fields of (a) 0.3 T and (b) 1 T oriented parallel to the c -axis. Below ~ 20 K, the creep rates for the irradiated and pristine samples are similar, suggesting that vortex dynamics is dominated by pre-existing pinning. The increasing $S(T)$ can be understood within the framework of the classic Anderson-Kim model [11] that predicts an approximately linear $S(T)$ -relation of the form $S = k_B T/U_0$, where U_0 is the dominant vortex pinning potential. $S(T)$ does not extrapolate to zero with either sample, indicative of a contribution due to quantum creep at low temperatures [12]. Many previous studies show similar $S(T)$ in this temperature range for YBCO samples with vastly different pinning properties [13,14,15].

At temperatures above 20 K, all samples show a clear departure from the almost linear $S(T)$ dependence suggesting a fundamental change in the vortex dynamics with a crossover in vortex pinning behavior from mostly strong single-vortex pinning at low temperatures to mostly collective pinning at high temperatures [12,13,14]. In the pristine samples, $S(T)$ is strongly non-monotonic and exhibits a minimum around 40 K with values of $S < 0.02$ that are remarkably low for YBCO at these temperatures. A similar minimum appears in other YBCO films with pinning dominated by large nanoparticles and is likely related to the large pinning energy of the individual nanoparticles **Error! Bookmark not defined.**[15].

The creep rate above 20 K is much higher for the irradiated sample, consistent with a scenario where pinning is dominated by smaller defects resulting in the well-known plateau in $S(T)$ [12]-[14].

Fig. 3: Flux creep rate as a function of temperature for an Au irradiated sample at 0.3 T (upper figure) and 1.0 T (lower figure). $S(T)$ for unirradiated controls are included for comparison.

C. Wide Tape Roll-to-Roll Irradiation

The irradiation of a 46 mm wide production coated strip was evaluated next. The irradiation was done using a roll-to-roll transport system attached to one of the beam lines of the tandem Van de Graaff accelerator. For the roll-to-roll irradiation of the 46 mm wide tape, the ion beam was shaped into a rectangle approximately 5 by 1 cm with the long dimension oriented along the length of the tape. In order to uniformly irradiate the strip the ion beam was rastered at approximately over 70 mm at a frequency of 50 Hz. The ion beam in the wide tape irradiations consisted of Au⁶⁺ ions with an energy of 16 MeV and a particle current of 120 nA. In the first experiment the tape transport speed was varied every 3 meters to change the dose from 2×10^{11} A/cm² to 10×10^{11} A/cm² in order to identify the optimum dosage. Between each speed change, the ion beam was blocked to leave an unirradiated control section. After the irradiations of all the dosages were completed, the 46 mm strip was roll slit in to four 10 mm wide tapes.

Figure 4 shows that the 77 K, self-field I_c and T_c both decrease as the dose of Au ions increases. At the previously determined optimal dose of 6×10^{11} Au/cm² the self-field I_c decreases by about 30% and the T_c falls by 2 K. The I_c enhancement as a function of dose was consistent with that measured with the static samples, with the optimal dose at 6×10^{11} Au/cm². The I_c enhancement, measured across the width of the 46 mm tape, was consistent confirming the uniformity of the irradiation as seen in Fig. 5.

Using the optimal Au dose, an 80 m length of 46 mm wide production tape was irradiated in the roll-to-roll configuration. The tape speed was approximately 10 m/hr. After the irradiation, the 46 mm strip was roll slit into 4 mm 'insert wires' that were laminated with a Cu stabilizer forming a standard coil wire. The 77 K self-field transport (1 m

measurement length) I_c of the unirradiated wire along with the short length I_c of samples taken from the ends of the 46mm wide strip before irradiation are shown in Fig. 6. The uniformity of I_c and the consistent decrease from the unirradiated sample confirm the uniformity of the irradiation over the 80 m length.

Fig. 4. I_c and T_c as a function of dose of 16 MeV Au ions after irradiation of a moving 46 mm wide production coated conductor tape.

Fig. 5. I_c enhancement measured in two separate 10 mm tapes slit from the 46 mm strip after the roll-to-roll irradiation.

Figure 7 shows the I_c of short samples of the optimally irradiated wire as a function of field (H//c) and temperature. The Au irradiation results in an enhancement of 2-fold or more at fields over 1 T. For comparison, the I_c of an unirradiated control sample at 30 K is included in the plot.

Fig. 6. I_c (77 K, self-field) as a function of length for a coil wire made from a 46 mm strip after irradiation to a dose of 6×10^{11} Au/cm². The pre-irradiation value is estimated from pieces cut from the ends of the original 46mm strip.

III. SUMMARY

We demonstrated the feasibility of enhancing the pinning in production length 2G wire using a roll-to-roll irradiation

process. Irradiation of the 2G wire with a dose of 6×10^{11} Au ions/cm² results in a doubling of the critical current of AMSC's standard coil wire in the 4 – 50 K operating regime targeted for rotating machine applications and high field magnet applications. The roll-to-roll irradiation was carried out with ion energies under 20 MeV, which are readily accessible with commercial electrostatic generators. Further optimization of the ion species and energy promises to lead to additional improvements in I_c and process rates required for incorporation into low-cost roll-to-roll manufacturing.

Fig. 7. I_c as a function of temperature and field (H//c) for a Au irradiated wire. The I_c for an unirradiated control wire at 30 K (open circles with crosses) is included for comparison.

REFERENCES

- [1] A.P. Malozemoff, "The power grid and the impact of high-temperature superconductor technology: An overview", in *Superconductors in the Power Grid*, C. Rey, ed. Elsevier, Amsterdam (2015) ch1.
- [2] X. Obradors and T. Puig, "Coated conductors for power applications: materials challenges," *Supercond. Sci. Technol.* **27**, 044003 (2014).
- [3] Y. Shiohara, T. Taneda, and M. Yoshizumi, "Overview of Materials and Power Applications of Coated Conductors Project," *Jpn. J. Appl. Phys.* **51**, 010007 (2012).
- [4] V. Selvamannickam, M. H. Gharahcheshmeh, A. Xu, Y. Zhang and E. Galstyan, "Critical current density above 15 MA cm² at 30 K, 3 T in 2.2 μ m thick heavily-doped (Gd,Y)Ba₂Cu₃O_x superconductor tapes," *Supercond Sci. Tech.* **28**, 072002 (2015).
- [5] Y. Jia et al., "Doubling the critical current density of high temperature superconductors through proton irradiation," *Appl. Phys. Lett.* **103**, 122601 (2013).
- [6] M. Leroux et al., "Rapid doubling of the critical current of YBa₂Cu₃O_{7- δ} coated conductors for viable high-speed industrial processing," (submitted for publication)
- [7] M.W. Rupich et al., "Second generation wire development at AMSC," *IEEE Trans. Appl. Supercond.* **23**, 6601205 (2013).
- [8] A. M. Campbell, and J. E. Evetts, "Flux vortices and transport currents in type II superconductors," *Adv. Phys.* **21**, 199 (1972).
- [9] H. Matsui et al., "Enhancement of critical current density in YBa₂Cu₃O₇ films using a semiconductor ion implanter," *J. Appl. Phys.* **117**, 043911 (2015).
- [10] J. F. Ziegler, J. P. Biersack and, M. D. Ziegler, SRIM, The Stopping and Range of Ions in Matter.
- [11] Y. Yeshurun, A. P. Malozemoff, and A. Shaulov, "Magnetic relaxation in high-temperature superconductors," *Rev Mod. Phys.* **68**, 911 (1996).
- [12] G. Blatter, M. V. Feigel'man, V. B. Geshkenbein, A. I. Larkin and V. M. Vinokur, "Vortices in high-temperature superconductors," *Rev. Mod. Phys.* **66**, 1125 (1994).
- [13] A. P. Malozemoff and M. P. A. Fisher, "Universality in the current decay and flux creep of Y-Ba-Cu-O high-temperature superconductors," *Phys. Rev. B*, **42** 6784 (1990).
- [14] J. Thompson, et al, "Effect of flux creep on the temperature dependence of the current density in Y-Ba-Cu-O crystals," *Phys. Rev. B* **47**, 14440 (1993).
- [15] H. Haberkorn et al., "High-temperature change of the creep rate in YBa₂Cu₃ O_{7- δ} films with different pinning landscapes," *Phys. Rev. B* **85**, 174504 (2012).