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Letter

Engineering current density over 5 kA mm^{-2} at 4.2 K, 14 T in thick film REBCO tapes

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Abstract

We report on remarkably high in-field performance at 4.2 K achieved in >4 μm thick rare earth barium copper oxide (REBCO) samples with Zr addition. Two different samples have been measured independently at Lawrence Berkeley National Laboratory and the National High Magnetic Field Laboratory, achieving critical current densities (J_c) of 12.21 MA cm^{-2} and 12.32 MA cm^{-2} at 4.2 K, 14 T ($B||c$), respectively, which corresponds to equivalent critical current (I_c) values of 2247 and 2119 A/4 mm. These I_c values are about two times higher than the best reported performance of REBCO tapes to date and more than five times higher than the commercial HTS tapes reported in a recent study. The measured J_c values, with a pinning force of ~1.7 T N m^{-3} are almost identical to the highest value reported for thin (~1 μm thick) REBCO at the field and temperature, but extended to very thick (>4 μm) films. This results in an engineering current density (J_e) above 5 kA mm^{-2} at 4.2 K, 14 T, which is more than five times higher than Nb_3Sn and nearly four times higher than the highest reported value of all superconductors other than REBCO at this field and temperature. The reported results have been achieved by utilizing an advanced metal organic chemical vapor deposition system. This study demonstrates the remarkable level of in-field performance achievable with REBCO conductors at 4.2 K and strong potential for high-field magnet applications.

Keywords: HTS, YBCO, coated conductor

(Some figures may appear in colour only in the online journal)

Introduction

Rare earth barium copper oxide (REBCO) coated conductors (CC) have a tremendous potential for numerous applications such as fusion reactor magnets, high energy particle accelerators, generators, motors, superconducting magnetic energy storage, and magnetic resonance imaging over a broad temperature range of 4–77 K in high magnetic fields of 2–30 T, due to their high critical temperature, high irreversibility field and high critical current density [1–10]. Several research and development projects are ongoing to develop high-field magnets with insert coils of REBCO, due to its high current carrying capability in high background fields [11–14]. Recently, a 42.5 T magnet has been demonstrated, with 11.3 T contributed by REBCO insert coils [14]. Also very recently, high performance REBCO-round wires with ultra-small diameters of 1.8 mm and other round REBCO wires have been developed for low temperature high-field magnet applications in accelerators [15–18].

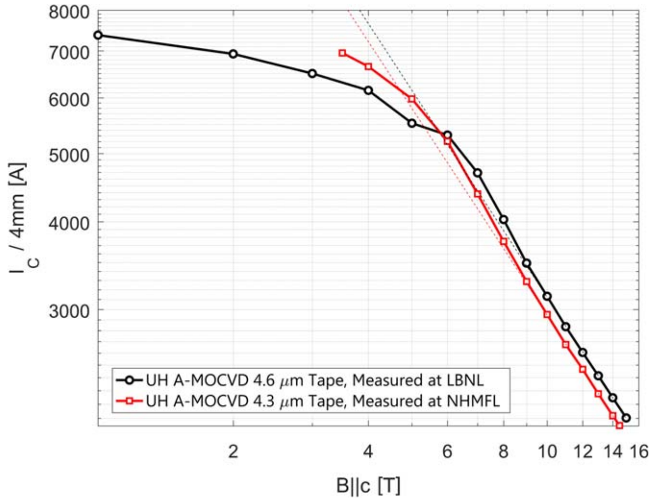

Figure 1. Critical current versus magnetic field applied along the *c*-axis at 4.2 K.

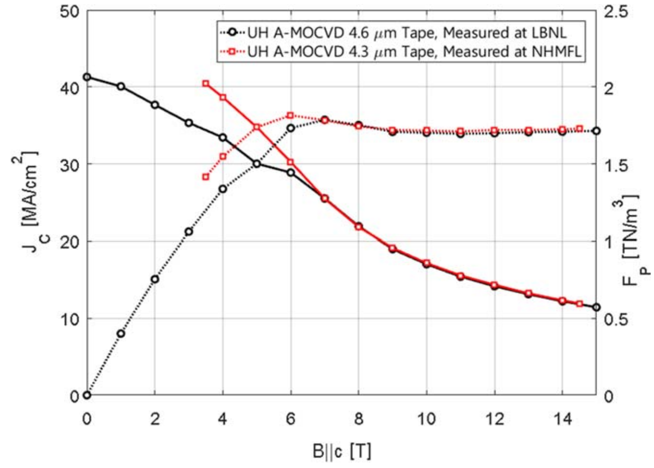

Figure 2. Critical current density (solid lines) and pinning force (dotted line) versus magnetic field applied along the *c*-axis at 4.2 K.

Significant progress in in-field performance has been achieved by introducing nanoscale defects like $BaZrO_3$ (BZO) [19–22], $BaSnO_3$ [23], $BaHfO_3$ [24], and Gd_3TaO_7 [25]. Pinning centers such as RE_2O_3 and BMO nanocolumns (M is metal) have been shown to enhance J_c over a wide range of temperatures (e.g., [26–30]). The BMO nanocolumns provide effective vortex pinning along *c*-axis and at low temperatures, the strain induced by lattice mismatch between BZO and REBCO matrix results in a high density of weak point pins raising J_c at all magnetic field directions [31–35].

A remarkably high pinning force density (F_p) of 1.7 T N m^{-3} has been attained at 4.2 K, 20 T in 0.9 μm thick 15 mol% Zr added REBCO film processed using metal organic chemical vapor deposition (MOCVD) by our group [31]. Recently, $BaHfO_3$ (BHO)-doped 0.26 μm $SmBa_2Cu_3O_y$ and 0.94 μm $EuBa_2Cu_3O_y$ films have been shown to exhibit a comparable F_p of 1.6–1.7 T N m^{-3} at 4.2 K, 15 T. [32, 33].

Significant increase in I_c performance can potentially be achieved if the strong deterioration of J_c with thickness is addressed, which is common to most REBCO growth techniques (e.g., [36–38]). Recently, a 3.2 μm thick, 20 mol% Zr REBCO film has been demonstrated by our group using conventional MOCVD in three passes, with a champion J_e of 1 kA mm^{-2} at 4.2 K at 31 T [35], demonstrating that this level of J_e is attainable in films thicker than the typical 1 μm. The multi-pass approach was used in order to curb severe degradation in J_c with thickness (>1 μm). However, the multi-pass technique significantly complicates the process [35, 39–42], which poses significant problems for scale-up to long length production.

An advanced MOCVD (A-MOCVD) system was developed under the ARPA-E grid-scale rampable intermittent dispatchable storage program, aimed at overcoming the main issues identified in conventional MOCVD reactors, including the J_c degradation with thickness [40]. The reactor utilizes direct ohmic heating of a suspended substrate tape, highly laminar flow and rapid tape temperature control using non-contact light pipe temperature monitoring, which when combined, enabled us to grow high performance thick REBCO films with and without dopants [40–42]. Previously, over 1500 A/12 mm critical current was achieved in 4.4 μm thick undoped REBCO on an ion beam assisted deposition MgO/LMO substrate in a single pass deposition using an A-MOCVD system [35].

Recently, we have explored the feasibility of utilizing A-MOCVD for growing thick Zr doped REBCO films optimized for in-field performance at intermediate temperatures (30–50 K) and fields, which is the operating regime of interest for applications such as motors and generators [42]. The results of this study have demonstrated that growth of very thick films without deterioration of J_c or texture is possible even in the presence of high volume density of $BaZrO_3$ nanorod precipitates. Remarkably, a high critical current density (J_c) of 15.11 MA cm^{-2} was achieved in a 4.8 μm thick 15 mol% Zr doped REBCO film, at 30 K, 3 T ($B\|c$), deposited in a single pass [42].

In this study, we used the A-MOCVD reactor to explore the possibility of growing very thick films optimized for 4.2 K in-field performance, The main purpose of this study was to investigate whether the A-MOCVD approach of growing very thick films with high J_c is also suitable for low temperature, high-field operation as well as to investigate the limits of thick REBCO films.

Experimental

In this study, REBCO films containing 15 mol% Zr were grown to a thickness over 4 μm. The composition is defined as 0.15 $BaZrO_3$ + 1.0 (Y, Gd)$_1Ba_2Cu_3O_{7-x}$ + 0.3 ((Y,Gd)$_2O_3$), with equal amounts of Y and Gd. The thick REBCO film samples were deposited in single pass in the A-MOCVD reactor on 12 mm wide Hastelloy/Al_2O_3/MgO/$LaMnO_3$ substrates, over a deposition zone length of 30 cm at deposition rate of 0.192 nm min^{-1}. Critical current measurements were performed in a field parallel to *c*-axis orientation, utilizing the standard 1 μV cm^{-1} criterion. The samples for I_c measurements were cut

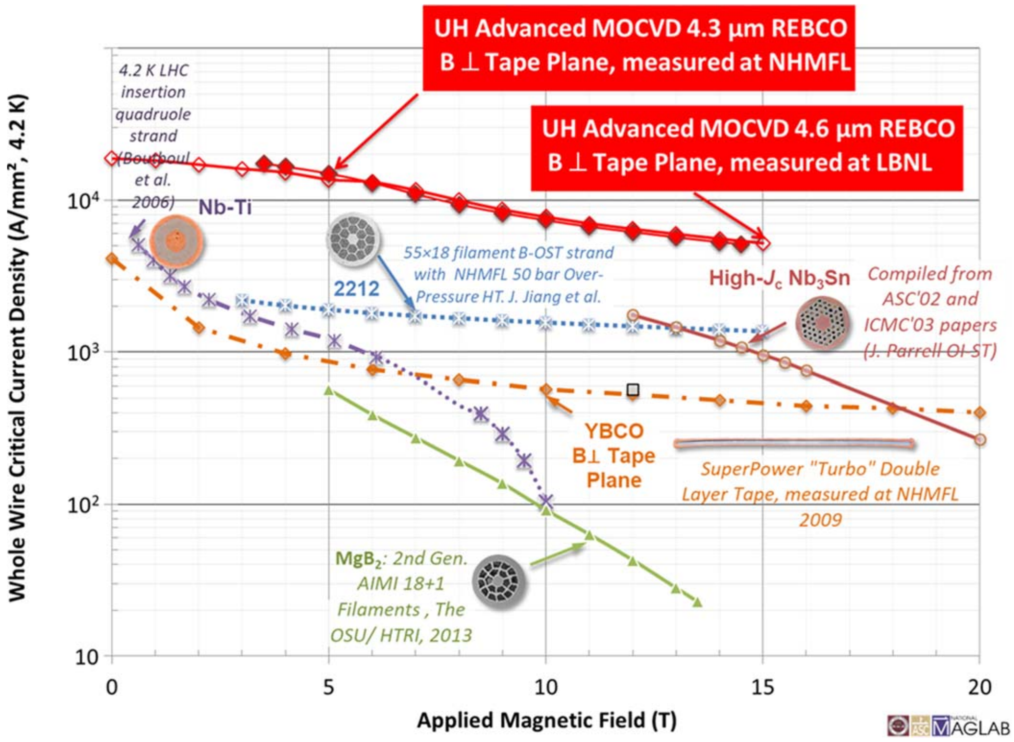

Figure 3. Engineering current density of UH REBCO samples versus magnetic fields along the c-axis at 4.2 K, compared to other superconductor technologies. Reproduced with permission from [44].

4 mm wide and critical current was measured over ~1 mm bridge in order to bring the total current to manageable levels for these measurements.

TEM characterization was performed using JEOL 2000FX microscope. Two-dimensional (2D) x-ray diffraction analysis was conducted using a Bruker GADDS system equipped with Vantec 500 detector.

Results and discussion

Two different samples were measured independently at Lawrence Berkeley National Laboratory (LBNL) and the National High Magnetic Field Laboratory (NHMFL) at 4.2 K, in magnetic fields up to 15 T applied along the c-axis. Both samples were of the same nominal composition and 15% Zr addition and were deposited separately in A-MOCVD as two independent samples, resulting thicknesses of 4.6 and 4.3 μm, respectively.

The results are summarized in figure 1 as a function of applied magnetic field parallel to c-axis ($B \| c$). Remarkably high critical current values of 2247 and 2119 A/4 mm width have been measured at 4.2 K, 14 T for the two samples. These values are higher by a factor of >2 than the best reported value in 3.2 μm thick, 20 mol% Zr added GdYBCO film processed in three passes using conventional MOCVD and more than five times higher than the commercial HTS tapes reported in a recent study [35, 43].

The corresponding J_c values are 12.21 MA cm^{-2} and 12.32 MA cm^{-2} respectively, as shown in figure 2 with solid lines. The pinning force (F_p) at 4.2 K, 14 T is 1.7 T N m^{-3}, as shown in figure 2 with dashed lines. This value is the same as the highest value reported in a 0.9 μm thick, 15 mol% Zr added GdYBCO film processed in single pass using conventional MOCVD [31]. This is significant in the sense that the same pinning force is achieved in samples with more than a four-fold increase in thickness. The pinning force has a peak at ~6 T and becomes near-constant at fields above 9 T. The peak in pinning force correlates well with the estimated matching field of 6.1 T obtained from the area and the nanorod count from plane-view TEM micrographs over >300 nanorods. The alpha value of the $I_c \sim B^{-\alpha}$ dependence is $\alpha = 1.03$ (1.02) at fields above 9 T for the two samples measured at LBNL and NHMFL, respectively.

The very high J_c values achieved directly impact the engineering current density (J_e)—one of the major metrics for most 4.2 K applications. The measured samples were deposited on substrates with Hastelloy and buffer stack thicknesses of 50 μm and 0.2 μm, respectively, ~3 μm cap silver layer and ~40 μm of surround copper stabilizer. Utilizing these values, the corresponding engineering current density values for the two samples at 4.2 K, 14 T ($B \| c$) are 5.48 kA mm^{-2} and 5.13 kA mm^{-2}, respectively, which again constitutes more than a two-fold increase compared to the best value of 2.5 kA mm^{-2} reported in the 3.2 μm thick, 20 mol% Zr added GdYBCO film [35]. To put these values on a map, the J_e versus field values of these two samples are plotted against other commercial superconductor technologies available for 4.2 K operation, i.e., on a plot of J_e versus B of various 4.2 K superconductors, as made and maintained by Lee [44]. The

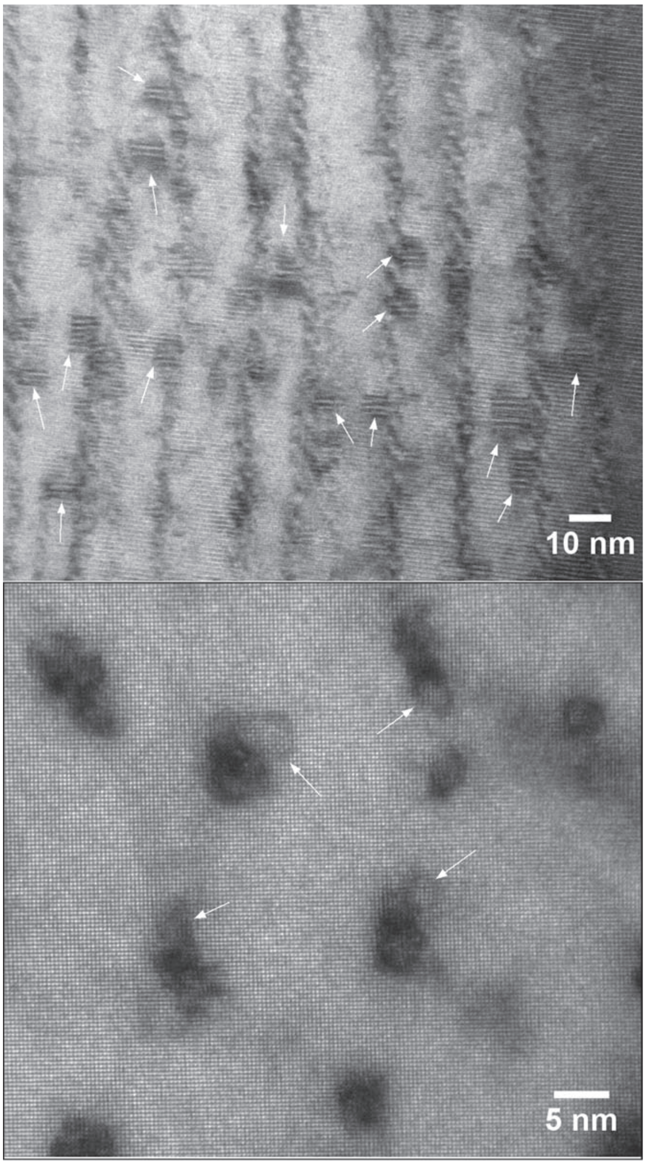

Figure 4. Cross-section (top) and plane-view (bottom) TEM microstructure of the 4.3 μm thick REBCO tape, showing aligned $BaZrO_3$ nanocolumns growing along the *c*-axis and interspersed with small RE_2O_3 precipitates. The average $BaZrO_3$ diameter of both micrographs is 3.7 nm.

results are shown in figure 3. At 15 T, the J_e of the thick film REBCO is over five times higher than the best reported J_e value of Nb_3Sn which is the primary superconductor used now in high-field applications. These results clearly demonstrate the potential of REBCO coated conductors for use in 4.2 K in-field applications.

Figure 4 shows a transmission electron microscopy (TEM) cross-section, as well as plane-view micrographs of the 4.3 μm thick sample, revealing both continuous BZO nanorods and small RE_2O_3 precipitates attached to the nanorods. The average BZO nanorod diameter determined from both cross-section and plane-view micrographs is 3.7 nm. A high density of vertically-aligned BZO nanorods along the *c*-axis and the presence of RE_2O_3 precipitates along the ab plane have been observed as homogeneously distributed over the whole film cross-section, with the selected micrographs being representative of the entire areas examined by TEM. We attribute such a uniform and continuous growth of BZO nanorods along the *c*-axis, without any interruption from RE_2O_3 precipitates over the entire 4.3 μm of thickness, to the high level of temperature and flow control in A-MOCVD [40–42]. This finding is different from that of the 3.2 μm thick 20 mol% Zr added GdYBCO film made in three passes by conventional MOCVD by our group, in which the length of the BZO nanorod was found to be reduced with increasing REBCO layer thickness and a low density of thick and short BZO nanorods was observed at the 100–200 nm interface between two passes [35].

Figure 5 shows a 2D x-ray diffraction (XRD) pattern of the 4.3 μm thick REBCO film. The sample is tilted by $\sim 23°$ in order to capture the REBCO 103 and BZO 101 peaks, and the spacing between peaks is near-linear in terms of reciprocal space vectors q_a and q_c. The sample reveals very sharp *c*-axis oriented REBCO peaks (00L and 10L series) indicating a very good out-of-plane texture. The pattern also reveals BZO 101 and RE_2O_3 004 and 222 peaks, indicating the presence of BZO nanorods and RE_2O_3 precipitates respectively in the REBCO matrix. The streaking of the BZO 101 peak is not in a constant 2θ direction but rather has a component perpendicular to the 00L direction, indicating small diameter nanorods [41]. Film thickness can also be estimated from the intensity of Hastelloy substrate rings, as was discussed in [35, 41, 42], which is almost negligible here, indicating a very thick REBCO film.

Summary

An A-MOCVD reactor has been used to deposit over 4 μm thick, 15 mol% Zr doped (Gd,Y)BaCuO tapes in a single pass, with fine, continuous $BaZrO_3$ nanocolumns and sharp texture. Critical currents of these samples have been measured at low temperature and high fields at LBNL and NHMFL. Remarkably high critical currents of 2247 A/4 mm and 2119 A/12 mm have been obtained at 4.2 K, in a magnetic field of 14 T ($B \| c$), which are approximately a factor of two higher than the best value reported in the literature. High critical current density of over 12 MA cm^{-2} and pinning force of 1.7 T N m^{-3} have been achieved. The engineering current density (J_e) value (considering a typical 40 μm thick copper stabilizer) of over 5 kA mm^{-2} has been achieved at 4.2 K, 14 T ($B \| c$) which is more than five times higher than Nb_3Sn and nearly four times higher than the highest reported value of all superconductors other than REBCO at this field and temperature. Such a remarkable performance reveals potential for the HTS technology to be utilized in future magnets for various applications requiring 4.2 K operating temperature and very high fields.

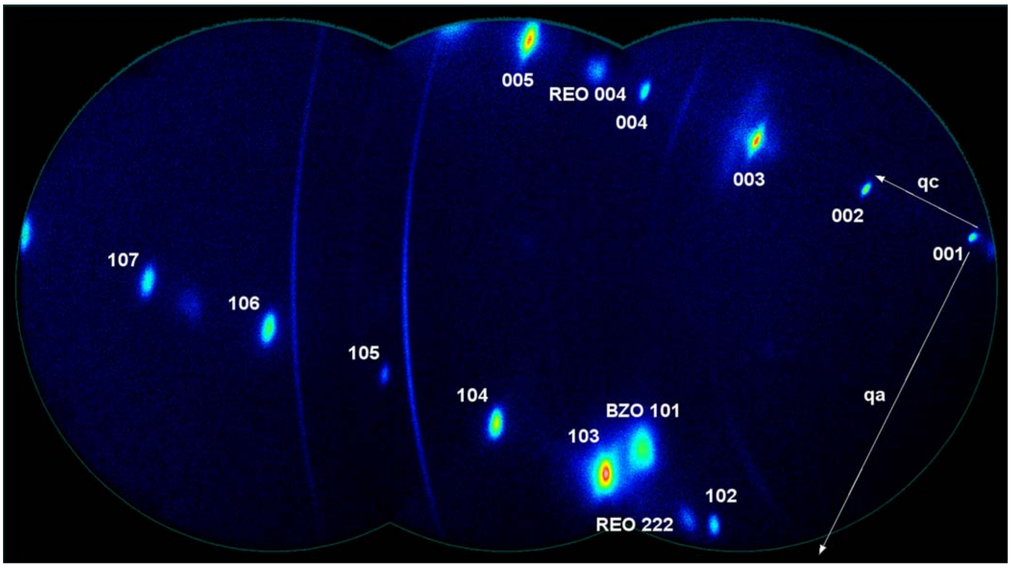

Figure 5. 2D-XRD pattern of the 4.3 μm thick REBCO tape, revealing the sharp out-of-plane texture of the REBCO phase. The BZO (101) peak is streaking in the direction perpendicular to the nanorod length indicating a small nanorod diameter.

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