Fabrication and Properties of High-$J_c$, Biaxially Aligned YBa$_2$Cu$_3$O$_{7-\delta}$ Thick Films on Metallic Tape Substrates


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FABRICATION AND PROPERTIES OF HIGH-\(J_c\) BIAxIALLY ALIGNED YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) THICK FILMS ON METALLIC TAPE SUBSTRATES

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Abstract
We report the synthesis and properties of high-\(J_c\) biaxially-aligned YBCO films deposited on thermo-mechanically textured nickel tapes. Sharply cube-textured nickel tapes, 125 \(\mu\)m thick, were produced by mechanical rolling followed by a recrystallization anneal. Short segments were coated with epitaxial oxide buffer layers, followed by fully aligned YBCO films to thicknesses of 1 to 3 \(\mu\)m. In-plane textures of 7–10° FWHM are achieved, with c-perpendicular alignment to 1° FWHM. Typical zero-field \(J_c\) values are in the range 5–9×10\(^6\) A/cm\(^2\) at 77 K, with strong behavior in magnetic fields comparable to that of epitaxial films on single crystal oxides. Assessment of properties necessary for a tape conductor technology are discussed.

Introduction
The discovery of high-temperature superconductivity (HTS) offered the promise of new applications, including HTS conductors carrying large currents in substantial magnetic fields, operating at liquid nitrogen temperatures (e.g. in the range 64 K to 77 K). After much practical development and many basic studies of flux pinning, dissipative flux motion, and grain boundary conduction in several HTS material classes, it has become apparent that for such a conductor: (i) the HTS material must have a small electronic anisotropy for potentially large flux pinning energies,\(^1\) and (ii) the current flow must occur in the basal planes across low-angle grain boundaries (<10° misalignment).\(^2\) Previously, we reported a new approach to the development of biaxially-aligned YBCO films deposited on thin metallic-tape substrates.\(^3,4\) Here we consider some systematics in the synthesis and properties that are relevant for potential applications as conductors. While present studies have been made on short segments, the technique is extendible to long lengths, and could point the way to a new coated conductor technology that can satisfy the above requirements.

Experimental Approach
Sharp texture can be achieved in \(\text{fcc}\) metals by appropriate rolling deformation followed by primary thermal recrystallization.\(^5\) In the present work, metal tape substrates were formed by progressive rolling of 99.99% pure nickel at room temperature, followed by annealing at temperatures between 400°C and 1000°C. While rolled tape thicknesses could be well controlled down to 25 \(\mu\)m, 125 \(\mu\)m thick
segments were used in the present work. Well-polished mechanical rolls produce Ni surfaces requiring no further treatment, and having an rms surface roughness of about 10 nm.

Chemically compatible buffer layers were then deposited on the textured base metal using sputtering, thermal or e-beam evaporation, or pulsed laser deposition (PLD). Typical oxide buffer layers are composed of a CeO₂ primary layer (50–400 nm thick), and a YSZ upper layer (100–500 nm). Briefly, an epitaxial CeO₂ film can be grown directly on the Ni surface at temperatures in the range 400°C to 650°C. To accomplish this, the vapor deposition occurs in a reducing background gas of 2×10⁻³ Torr Ar/4%H₂, which removes and prevents the formation of NiO, while leaving the much more stable CeO₂ unaffected. We refer to the net, aligned buffered metal tapes as Rolling Assisted Biaxially Textured Substrates (RABiTS). Further details regarding epitaxy of oxides on metals and of the RABiTS formation conditions are given elsewhere. YBCO films were grown on the RABiTS using PLD at a temperature of 780°C in an oxygen pressure of 185 mTorr. Typical short segment sample dimensions are 3 mm wide x 15 mm long. YBCO thicknesses were chosen in the range 1–3 µm for an assessment of high-current properties, as discussed below. For four-terminal electrical transport measurements, gold or silver current and voltage contacts were sputter deposited, and the sample given a final anneal at 500°C in 1 atm O₂. The gauge length between the voltage terminals was either 3 or 4 mm.

Results and Discussion

X-ray diffraction analysis showed that the buffer layers and YBCO deposits are biaxially aligned with both in-plane and c-axis perpendicular texture. Rocking curve widths of the YBCO (005) peaks yield c-axis alignment in the range of 1–4° FWHM. This value is actually somewhat better than the typical out-of-plane Ni-texture of 6–8° FWHM. In-plane texture taken from φ-scans through off-axis reflections indicate that all subsequent layers typify the 7–10° FWHM in-plane texture of the Ni.

A series of superconducting films were deposited onto YSZ/CeO₂-buffered Ni tapes, along with control samples deposited under similar conditions onto single crystal SrTiO₃ substrates. A focus of the effort was to compare the field and temperature dependent critical current density among different YBCO/RABiTS samples, and with properties of the "bench-mark" materials deposited on SrTiO₃. In addition, a preliminary test of the bend-strain tolerance was conducted, both in compression and tension, and an assessment made of the prospects for achieving practical current levels through this type of coated-conductor approach.

For some samples, initial measurements of the resistive transition and high-temperature Jₑ were made on the full, 3 mm width. To minimize the heating effects at the current contacts, and to reduce the overall currents required, thereby extending
Figure 1. The magnetic field dependent $J_c$ at 77 K, of a YBCO/RABiTS, compared to other HTS epitaxial thin films that have been deposited on single crystal oxide substrates. Most data are shown for $H||c$, the applications-limiting orientation. The characteristics with $H||ab$ is typical of the strong intrinsic pinning observed for all HTS materials.

The measurements to lower temperatures, the samples were subsequently wet-etch patterned to a bridge width of 1 mm. In addition, in some cases pulsed current measurements extended the measurements to the low-field, low-temperature regime. In all cases, the measurements yielded identical results in the regions of overlap, demonstrating that the samples are macroscopically homogeneous and that the measurements are well controlled.

Figure 1 shows the magnetic field dependence of the critical current density $J_c$ at 77 K. Here, the results for a 1.4 μm thick YBCO deposit on a RABiTS is compared with high-$J_c$ epitaxial thin films deposited on single crystal oxide substrates, for different HTS material classes. In all cases, the films on single crystal substrates have zero-field $J_c$ values in excess of 1 MA/cm². While all the films had excellent, similar characteristics with the field parallel to the film plane ($H||ab$ planes, the so-called "intrinsic pinning" case), Fig. 1 emphasizes the characteristics with $H||c$, which is the applications-limiting orientation where the flux pinning is limited by the microstructure and the effects of intrinsic material anisotropy. Figure 1 reinforces that not only is YBCO the HTS material of choice for in-field conductor applications, but the high-field properties of YBCO/RABiTS may exceed those of the prototype, YBCO/SrTiO₃ films. Indeed, the somewhat suppressed $J_c(H=0)$ for YBCO/RABiTS may suggest a high density of crystalline defects produced naturally from the growth process on RABiTS.

Figure 2 supports this idea in a comparison of the Rutherford Backscattering Spectra (RBS) for a YBCO/RABiTS, in both the random and channeling orientations. Analysis of the random spectrum reveals that the YBCO/RABiTS composition is yttrium rich, and slightly barium poor. These results may explain the slightly reduced $T_c$ of 86–88 K. Figure 2 also shows that the RBS channeling minimum yield
The channeling minimum yield $x_{\text{min}}$ is 36% for this YBCO/RABiTS. Measurements on other YBCO/RABiTS samples show that $x_{\text{min}}$ falls in the range 30–50%, while a typical value for YBCO/SrTiO$_3$ is $x_{\text{min}}$ ~4%. The relatively suppressed channeling characteristics of YBCO/RABiTS imply crystalline defects that may be responsible for the enhanced pinning at high vortex densities. Detailed cross-section TEM characterizations will be required to help confirm this conjecture.

While the field-dependent $J_c$ levels in YBCO/RABiTS prototypes are adequate to enable new applications in the liquid nitrogen temperature range, the fraction of superconductor in the present architecture is quite small (e.g. ~3 $\mu$m/126 $\mu$m < 3%). An important issue for scale up is achievement of a sufficient overall current density, presumably obtained through the combination of both thinner RABiTS and thicker YBCO deposits. For the latter objective, it is important to assess superconducting properties of thick HTS deposits on RABiTS.

Figure 3 shows such results for applied fields of zero and 1 Tesla, applied parallel to the c axis. Most data are given at 77 K, but some results are presented at 64 K, the temperature of pumped liquid nitrogen just above its solidification point. From Fig. 3, for YBCO thicknesses of ~1–3 $\mu$m, $J_c$ (1T) exhibits little systematic thickness dependence, with values in the range ~100–150 kA/cm$^2$ at 77 K. For perspective, the curve represent $J_c$ values required to satisfy a proposed operations criterion of $K_c$=10 A/mm-width for a tape conductor (e.g. for the present overall thickness $\tau$=100 $\mu$m, this criterion corresponds to an engineering current density $J_c=K_c/\tau$=10$^4$ A/cm$^2$). Even for the present low superconductor fraction, the data at 64 K suggest that practical levels are accessible in the liquid nitrogen temperature range.
Figure 3. The dependence of $J_c$ on YBCO thickness are compared. Open symbols are values in self field, while closed symbols represent $J_c$ in an applied field $H_{||}=1$ Tesla. The curve represents $J_c$ levels required to satisfy an operating criterion of 10 A/mm-width.

It is noteworthy to mention the YBCO/SrTiO$_3$ control sample in Fig. 3 represented by the inverted triangle. This 1.2 µm thick YBCO film was fabricated by an approach that involves the deposition of a stable, non-superconducting precursor film, followed by the formation of YBCO ex situ during a post-deposition furnace anneal. This deposited-precursor technique could provide an important component for the development of a YBCO coated-conductor technology, since the deposition process requires neither high substrate temperatures nor an (activated) oxygen source, and should be insensitive to arbitrarily fast deposition rates. The prospect of reacting large quantities of wire in a batch annealing process is also appealing. Important remaining issues include the suitability of RABiTS substrates for these types of films, as well as the effects of thicker films on reaction times, other processing variables, and on the final properties.

Conductor applications will require strain tolerances that permit bending to form coils for magnets, motors, etc. Figure 4 illustrates results of bend-strain tolerance measurements for a sample placed in compression. Measurements were made sequentially by first bending the sample around a specified diameter mandrel, followed by straightening, and then measurement of $J_c$ at 77 K in zero field. The repeated mechanical cycling associated with this technique resulted in a noticeable work hardening of the Ni. Also shown in Fig. 4 is the effect on the sample resistance $R$ at room temperature. In compression, $J_c$ degrades rapidly near a strain of 0.5%; the abrupt increase in $R$ at this same strain level indicates the likely formation of cracks. For the present RABiTS dimensions, this 0.5% strain level corresponds to a bend...
diameter of about 2.5 cm. Measurements in tension yield similar results, except for degradation at a smaller strain level of ~0.2%.

Summary
If these preliminary results hold for future configurations having thinner RABiTS and thicker YBCO, there appears to be a basis for optimism that several criteria for real applications can be met. Future developments must focus on issues of reproducibility, long-length demonstrations, rates, and problems presented by the magnetic substrate for applications requiring temporally changing currents and fields (ac hysteresis loss issues).

Figure 4. The dependence of $J_c$ on the bend strain produced in compression, for a YBCO/RABiTS sample. Also shown is the room temperature normal state resistance, which serves as an indication of a breakdown by cracking.

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