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Long-length YBCO coated conductors for ultra-high field applications: gaining engineering current density via pulsed laser deposition/alternating beam-assisted deposition route

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Abstract

Gaining engineering critical current density \( J_c \) in high-temperature superconducting (HTS) coated conductors based on double-disordered YBCO for operation at high fields is one of the key requirements in upcoming magnet/accelerator projects. Currently, the development of a tape with advanced \( J_c \)-s is performed based on an alternating beam-assisted deposition–pulsed laser deposition (ABAD–PLD) manufacturing route. An obvious way to improve \( J_c \)-s is to reduce substrate thickness. A 40% \( J_c \) increase is expected owing to the thickness reduction from 100 to 50 \( \mu \)m. Nevertheless, the reduction of substrate thickness in case of the applied processing technology that employs relatively thick (2–3 \( \mu \)m) yttria-stabilized zirconia buffer layers leads to the manifestation of considerable strain in the tape resulting in strong tape bowing. Advanced processing routes have been developed to suppress this effect. The highest engineering current density was recorded at well above 1000 A mm\(^{-2} \) under an ultra-high field of 18 T at 4.2 K, \( B/\|c \). Influence of tape bowing and the impact of longitudinal defects are studied via \( V-J \) measurements at 77 K and a moderate magnetic field with intrinsic edge gradients. Potential for further gaining of \( J_c \) was found employing mechanisms of (i) film nucleation from lateral flows that provide material transfer during the PLD process. Further steps include suppressing the instability of instantaneous temperature via quasi-adiabatic pulsed heating of the growing HTS film via (ii) control of laser plume energy, and (iii) energy release during film condensation (condensation enthalpy). These effects disturb the instantaneous temperature of the growing HTS layer, which exhibits very limited capability of heat transfer. Temperature pulses reaching 30–40 K were evaluated via heat transfer modeling. Stabilization of the level of pulsed temperature during layer growth is anticipated to result in a further increase in \( J_c \).

Keywords: high-temperature superconductors, superconducting tapes, yttrium compounds, laser ablation, epitaxial growth, superconducting materials

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

High-temperature superconducting (HTS) coated tapes initially aimed at ‘high-temperature’ applications (e.g. at 77 K) acquired an important additional role as a low-temperature superconductor due to their extraordinary high engineering current density, $J_c$, remaining in ultra-high fields, well above 20 T. Intensive efforts have been made to establish a reliable technology for fabricating long and high-$J_c$ tapes for high and ultra-high fields at Bruker HTS, Fujikura, SuperPower/Furukawa, etc [1–3].

The Bruker HTS technological chain was established and fine-tuned for a 100 μm thick substrate. This relatively high thickness enables favorable mechanical properties that are important for ultra-high field magnets. Despite their excellent critical currents (>1000 A cm$^{-1}$ width at 18 T, B // c, 4.2 K), the thick Bruker tapes yielded high but not the highest level of engineering current density (600–800 A mm$^{-2}$ at the same conditions [3]). Numerous projects, particularly by CERN within the ARIES project [4, 5], have been initiated to increase $J_c$ for our processing route. The reduction of substrate thickness in the employed tape design (as shown in figure 1) to achieve this goal seems to be the simplest. In reality, with a 50 μm thick substrate, major core steps in the employed processing route, such as alternating beam-assisted deposition (ABAD) [6] and pulsed laser deposition (PLD) [3, 7, 8] must be re-tuned because of a substantial variation of mechanical and thermal parameters of a thin substrate.

This work was aimed at (i) exploring recent technological progress in tape production, (ii) finding and analyzing new, advanced conditions for processing HTS-coated tapes based on thin substrates, and (iii) determining the potential for further gaining of $J_c$ by using recently established physical mechanisms and features of a deposition process.

2. Processing platform

The HTS-coated tapes are based on CrNi stainless steel (or Hastelloy) with thickness of 100 μm (‘thick’ substrates) and 50 μm (‘thin’ substrates) and tape widths of 4 or 12 mm. The tapes were electro-processed, or alternatively mechanically polished and finally ultrasonically cleaned. ABAD [6] was employed for deposition of a bi-axially textured yttria-stabilized zirconia (YSZ) buffer layer, typically 2–3 μm thick. PLD is based on a drum-supported tape with equilibrium heating of the tape via hot wall reactor positioned in the deposition zone [3, 8]. Further details of PLD including a discussion of mechanisms determined growth of double disordered (DD) superconducting layers [3] based on YBa$_2$Cu$_3$O$_{7-δ}$ can be found in [8]. The final steps of tape manufacture include enveloping of the tape with a 1–2 μm thick silver layer deposited via thermal vacuum evaporation and metallization; the final steps of tape fabrication are described in [7, 8].

3. Tape performance

3.1. Critical current and engineering current density

In-field performance of DD-YBCO-based tapes with different substrate thicknesses is shown in figure 2, which plots the critical current versus flux density for a set of samples (end pieces of long tapes). The areas viewed in figure 2 as high-field (HF) and ultra-high-field (UHF) tapes are defined via a simple condition, $I_{c,\text{UHF}}(B) > 2020 B^{-0.73}$, that allows to distinguish the DD-YBCO tapes of HF or UHF class. Such definition is possible because the $I_c(B)$-dependence is well defined via $\alpha$-law [1] in the wide field range. The tapes with lower $I_c$ correspond to HF tapes because even with moderate current level they are able to carry sufficiently high currents (>300 A) in 4–10 T range. UHF tapes must transport high currents in 10–31 T range. Thus, their $I_c$ level in wide field range must be higher.

There is a scattering of $I_c$, which, nevertheless is not degrading in tapes with thinner substrates. In general, the overall scattering of in-field $I_c$ per cm-width, as shown in figure 3, for routinely produced tapes indicates technological progress regarding increase of reproducibility and yield.

In the 4 mm wide tape, Q021-18N1, based on a 50 μm thick substrate, the highest $I_c$ value observed was 454 A at 18 T, $B // c$, and 4.2 K (the red star in figure 2). Considering the Cu plating of $2 \times 20 \mu m$, the total tape thickness is 90 μm, which is 50 μm thinner than that of our thick substrate tapes. This results in a champion level $J_c = 1261$ A mm$^{-2}$ (that nevertheless should not be interpreted as a standard tape quality, more a level that is potentially achievable). However, we can expect an increase in $J_c$ because of the reduction in the tape thickness. The thickness reduction factor of 1.56 (140 μm/90 μm) leads to an increase of $J_c$ from 800 to 1244 A mm$^{-2}$, where 800 A mm$^{-2}$ represents a high level $J_c$ for a 100 μm substrate of HTS-coated tapes [3]. This is in good agreement with the observed value of 1261 A mm$^{-2}$ and
indicates the potential to gain even higher \( J_e \) values because the \( J_e \) of the tapes with 100 \( \mu \)m thick substrates may exceed the 800 \( \text{A mm}^{-2} \) \cite{3} mentioned earlier.

Figures 4(a) and (b) show the reduction of \( I_c \) in the course of tape bending. A new device with a variable bending radius enabled measurements without dismounting the tape sample. The critical bending radius for the 50 \( \mu \)m case corresponded to \( R_{cr} = 4.5 \text{ mm} \). This value is half that of \( R_{cr} = 8.9 \text{ mm} \) observed at 100 \( \mu \)m \cite{9}.

3.2. Transverse bowing

A characteristic feature of the ABAD–YSZ buffered tapes is a bowing that originates from compressive stress occurring mainly during the growth of the buffer layer. This is a well-known effect caused by atomic peening \cite{9} occurring in the course of the sputter-based ABAD employed in our processing route. Furthermore, a significant degree of bowing is retained by the tapes after deposition of all layers and intermediate annealing. This bowing effect can be seen in figure 4(c), where a cross-section of a 12 mm wide tape is depicted (left tape). The bowing height may exceed several millimeters depending on the thicknesses of the buffer layer and substrate. Such tapes show inhomogeneity of the in-plane texture, which results in an inhomogeneous \( I_c \) over the tape length. This can be seen in figure 2 for samples Q020-N1 and Q020-N2 which represent different parts of the same tape. The difference in \( I_c \) at 20 T corresponds to \( \sim 6\% \). In some cases, this difference may exceed 10%.

We found that that this effect can be suppressed by the adjustment of the deposition parameters and introduction of additional layers. As a result, the tape has a significantly lower bowing height as is shown in the right-side tape in figure 4(c). Nevertheless, some residual bowing should be considered during tape processing, especially for thin substrates with a width of 12 mm.

3.3. Impact of longitudinal defects

Any technology based on reel-to-reel tape translation \cite{10, 11} may expect intermittent longitudinal defects, e.g. scratches, especially in bowed tapes. In this work, we simulated such defects by scratching the finished tape (without the Cu layer) by using a hard material tool.

We measured the impact of an artificially introduced, 2 cm long surface scratch on the critical current at 77 K. The total dimension of the sample was 12 cm \( \times \) 0.4 cm; the voltage contacts were separated by a distance of 3 cm and the initial critical current was 38.5 A (curve 1 in figure 5).

We used the criterion of 1 \( \mu \)V cm\(^{-1} \) voltage drop between contacts (i.e. 3 \( \mu \)V at 3 cm distance) to define \( I_c \). With a scratch, the critical current (curve 2) degraded to 33.6 A, i.e. by \( \sim 13\% \). This degree of deterioration also seems to be typical of the slitting of the manufactured tapes.

We compared \( I_c \) degradation due to the scratch in a field produced in the gap between 2 disc-like permanent magnets
creating a flux density \( B \sim 0.4 \, \text{T} \). In this particular field applied perpendicular to the tape, \( I_c \) is reduced to 12.4 A, i.e., by a factor of \( \sim 3 \) compared to the initial tape. In scratched tape, \( I_c \) reduces to 11.8 A in the field, yielding a difference of 5%. In contrast to the 13% reduction, this minor influence of a scratch at 0.4 T seems surprising. Here, we must consider the ‘zooming’ effect, which is pronounced in the local magnetic field. Owing to the drastic suppression of the in-field critical current, voltage drop \( U_{\text{tot}} \) in the measured tape is mainly determined by the in-field tape area. This follows from the \( n \)-law, which in the considered case of the coordinate-dependent field, may be generalized as

\[
U_{\text{tot}} = \frac{U_e}{L} \int_0^L \frac{I}{I_c(B(x))} n(B(x)) \, dx, \quad (1)
\]

where the in-field area with the lowest \( I_c \) determines the fraction of the integrand. Here, \( U_e \) is a criterion used for defining \( I_c \). 

\( L \) is the distance between voltage probe contacts, \( B(x) \) is a linear distribution of flux density in the longitudinal direction of the tape, and \( n(B(x)) \) is a coordinate-dependent power value used instead of \( n \)-values employed in the constant field case. The approximation via \( (1) \) (figure 5(a), curve 4a) sufficiently agrees with that obtained through the experiment, similar to the approximation suggested in [12]. However, the approximation predicts a 7% reduction of \( I_c \) instead of the observed 5%. This suggests that further features of tape behavior within field gradients should be considered. Nevertheless, the dominating influence of defects remains in the field was confirmed.

4. Potential for further \( J_e \) gain and discussion

4.1. Impact of lateral flows in PLD

In [3], we showed that the distribution of the deposited material that originates from the stationary (i.e., fixed) laser plume is extremely wide, considerably exceeding the diameter of the laser plume. Figure 6 shows the fragment of the interferogram (with fringes of equal thickness) of the pulsed-laser-deposited DD-YBCO on the Al-foil substrate attached to the PLD drum; the deposition was at 25 °C–35 °C. Despite the drum curvature (100 mm\(^{-1}\)), fringes of equal thickness on the flattened foil are circular (not elliptical). Corresponding thickness distribution is shown in figure 8.
‘jumps’ of the ultrasonic flow. A rough estimation of the speed of the initial flow yielded \( \sim 5 \text{ km s}^{-1} \) (for a target-to-substrate distance of \( \sim 5 \text{ cm} \) and plasma propagation time of \(< 10 \mu\text{s})\). Thus, \( M \sim 15 \) implies that the process is within the high hypersonic range.

Deposition in plume-scan mode at standard deposition temperature confirmed this wide distribution remains under actual process conditions [3]. In figure 8, we plotted this distribution together with the critical currents measured at thickness slope area (i.e. over \(-20 \text{ mm} < Z < 30 \text{ mm}\)) they exhibit relatively good agreement.

The observed wide distribution of layer thickness implies that nucleation of DD-YBCO starts far away from the scan area of the laser plume. Considering that film nucleation starts at the nanometer thickness range, the distance from the center of the laser plume may be considerably higher than the radii shown in figures 6 and 7. This offers a good opportunity to independently control layer nucleation via creating different conditions (e.g. temperature and oxygen pressure) in the nucleation zone. As such, the DD-nanostructure and as a result the \( J_c \) performance may be further improved.

4.2. Instantaneous temperature at YBCO surface

The substrate temperature varies rapidly for any PLD process [16]. By using a drum-based tape support with fast drum rotation in the tubular heating zone, temperature oscillations are basically suppressed but not completely eliminated [10]. Especially, the amplitude of residual temperature oscillations is much more pronounced in the case of thin substrates (50 \( \mu \text{m} \)), where this amplitude may exceed 5°C with a transient time of tens of milliseconds. This seems to be tolerable for PLD processing, as HTS layers with the highest \( J_c \) were prepared in this way.

Nevertheless, the process window for substrate temperature is not very wide, possibly less than 10°C, especially in the case of DD-YBCO.

Recently, we found that there is another, even more powerful source for temperature perturbation. This source is the energy release of ions impinging upon the growth surface of the DD-YBCO layer. The key factor that motivated us to analyze this case was that the thermal conductivity of the YBCO layer is extremely poor in the \( c \) direction, i.e. perpendicular to the growth direction.

Owing to the short lifetime of the laser plume, the integrated incident ion energy of \(~0.05 \text{ J cm}^{-2}\) was delivered to the interface within 5–10 \( \mu\text{s} \). Conditions for thermal accumulation and release for the given case are schematically shown in figure 9. Owing to the low thermal conductivity of \(~1 \text{ W K}^{-1}\) at \( T > 300 \text{ K} \) [17, 18], YBCO represents a ‘bottleneck’ for heat release in the normal direction to the substrate that, in this case, together with the buffer layer plays the role of a heat reservoir. The alternative route for heat release via IR emission, which increases with increasing temperature, was shown to be negligible in comparison with heat conduction.

The heating–cooling kinetics evaluated for a heating pulse with a 10 \( \mu\text{s} \) duration are shown in figure 10 for an
The kinetics of temperature saturation and temperature release could have been a result of two factors: increasing heat capacity of the layer and increasing heat resistance with growing thickness. At an intermediate thickness of 0.75 μm, the maximal increase of the instantaneous temperature exceeded 33 °C. Because the pulse a temperature increase above 42 °C occurs, and the YBCO temperature decays much more slowly, taking tens of μs, because (i) much more energy is stored in a thicker layer, and (ii) of increased heat resistance introduced by the HTS layer. Thus, the evaluated pulses of the instantaneous temperature are shown to be rather high, exceeding a tolerance for deposition temperature by a factor of 4 at least. Because the temperature pulses are dependent on layer thickness, one may assume that energy flow originating from the laser plume is constant during the 10 μs pulse (see curve 1 in figure 10). Power delivered to the growth interface during the deposition pulse. Because of the short (<10\(^{-2}\) s) migration time of adatoms on the surface of the growing layer [20, 21] a rapid condensation should take place. Calculation of the energy released via condensation enthalpy is shown in figure 11 that similarly to figure 10 evaluates the kinetics of instantaneous temperature assuming a 10 μs long PLD ‘condensation’ pulse.

The vaporization enthalpy of the DD-YBCO composition was evaluated and employed in modeling of the condensation process in which the energy should be released. The energy was evaluated as about 5% of the energy transferred by ions. However, the pulse temperature was significantly lower, determined by only 2% of the energy triggered pulse, due to LFL which distributes the energy over a large substrate area. Because of the pulsed nature of PLD, energy and temperature effects occur as a sequence of short pulses [3, 22]. Temperature pulses originating from condensation enthalpy should be added to the temperature pulses caused by ‘condensation’ of ion energy.

Thus, the evaluated pulses of the instantaneous temperature are shown to be rather high, exceeding a tolerance for deposition temperature by a factor of 4 at least. Because the temperature pulses are dependent on layer thickness, one may
employ a compensation of the perturbation effect via decrease of the background substrate temperature during deposition process. This may be employed as a tool for improvement of DD-YBCO growth kinetics and, finally, increase of critical current density.

### 4.3. Expected improvement of critical current

The DD-layer growth employing the PLD with LFLs has already been used in the processing of ultra-high field tapes, which yield a very high engineering current density at a high current density.

| Tape # | $T_{src} - T_{dep}$ K | $J_c$ (4.2 K, 5 T, $B//c$) MA cm$^{-2}$ | $J_c$ gain % |
|--------|----------------------|----------------------------------------|------------|
| Q047C  | 0                    | 14                                     | 0          |
| Q068V  | 17                   | 16                                     | 15%        |

Table 1. Gain of critical current and current density in the tape deposited with increased temperature during nucleation of DD-YBCO layer.

The variation of nucleation temperature $T_{src}$, which yield a very high engineering current density at a high current density, is necessary according to new results. Following the variation was needed according to new results. In this case is reproducible enough to confirm the presence of the gain effect. This can be observed despite insufficient amplitude of temperature variation (17 K instead of 35 K). Moreover, the temperature during the major part of layer deposition remained almost constant, while a further variation was needed according to new results (figure 10). Following the ‘sensitivity’ of $I_c$ to the deposition temperature, a 20%–30% $I_c$ improvement should be reachable when the foreseen temperature variation is introduced during DD-YBCO growth. In our drum technology, with a one-directional path of the tape through the deposition zone, the implementation of such a variation could be possible in future studies.

### 5. Conclusion

High performance DD-YBCO coated conductors of reduced substrate thickness (from 100 to 50 μm) of 4 and 12 mm width, are routinely processed in lengths up to 600 and 100 m, respectively. A champion level of engineering current density of well above 1000 A mm$^{-2}$ at 18 T, $B//c$, 4.2 K was observed in DD YBCO coated tape based on 50 μm thick substrate. We developed and implemented a means for suppressing tape bowing that is especially pronounced in thin tapes. The impact of longitudinal defects (scratches) on the $U-I$ dependencies in the magnetic field exhibits a nontrivial effect, where the influence of partial damage to the tape is reduced in a gradient field area.

Potential for further gain of $J_c$ is shown via (i) independent control of nucleation temperature: that may be achieved by activation of lateral flows responsible for PLD nucleation, and (ii) compensation of instantaneous temperature instability (of 20°C–40°C) introduced by impinging ions from the laser plume. The minor impact of condensation energy (enthalpy) released during pulsed crystallization of YBCO is shown. The experimental results confirmed, at least partly, our expectations on the 20%–30% gain of the critical current via temperature variation during layer growth under lateral flows. The implementation of this art of deposition will be accomplished in future studies.

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