Growth of all-chemical high critical current YBa$_2$Cu$_3$O$_7$—$\delta$ thick films and coated conductors

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Abstract
The achievement of high critical currents in ‘all-chemical’ YBa$_2$Cu$_3$O$_7$—$\delta$ thick films from low cost and versatile chemical solution deposition (CSD) methodology is still an open issue. Here we report a study of the nucleation and growth conditions to achieve YBa$_2$Cu$_3$O$_7$—$\delta$ films in excess of 1 micron using single pass inkjet printing or multideposition of low-fluorine metalorganic precursors. Growth conditions of thick YBa$_2$Cu$_3$O$_7$—$\delta$ layers are first investigated on LaAlO$_3$, where there is no interfacial chemical reactivity. The second architecture investigated is an ‘all-chemical’ CSDCe$_{0.9}$Zr$_{0.1}$O$_2$(CZO)/YSZ multilayer on single crystal substrates which serves as model system for coated conductors. Finally, the ‘all-chemical’ coated conductor architecture CSDYBa$_2$Cu$_3$O$_7$/CSDCZO/ABADYSZ/stainless steel, where ABAD stands for alternating beam assisted deposition, is investigated. The nucleation conditions of YBa$_2$Cu$_3$O$_7$—$\delta$ films on top of CZO cap layers have been selected to minimize the formation of the BaCeO$_3$ phase at the interface. We demonstrate that by combining the use of Ag additives in the starting YBCO solution and processing conditions leading to low supersaturation (high water pressure and low temperature) we can achieve $\sim$1 $\mu$m thick YBa$_2$Cu$_3$O$_7$—$\delta$ films and coated conductors with high critical currents of $I_{c sf} = 390$ and 100 A/cm-w, respectively, at 77 K and self-field. The achieved control of the interfacial reactivity with CeO$_2$ cap layers opens a route for further increasing film thickness and critical currents in ‘all-chemical’ YBa$_2$Cu$_3$O$_7$—$\delta$ coated conductors.

Keywords: critical current, YBa$_2$Cu$_3$O$_7$, coated conductor, chemical solution deposition, thin and thick film

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

1. Introduction
The development of superconducting power applications and ultra-high field magnets relies, at present, on the use of second generation high temperature superconductor wires, i.e. coated conductors (CCs), based on epitaxial REBa$_2$Cu$_3$O$_7$ (REBCO, RE = rare earth or Y) grown on top of suitably protected metallic substrates [1–4]. Several CC architectures have been widely investigated in recent years, to reach high superconducting performances and the methodology has been extended to other high temperature superconductors [5]. A key objective is to ensure that the manufacturing cost of long length wires remains as low as possible. The success of CCs relies, therefore, on minimizing the cost performance ratio, usually measured as $\€$/kA m, i.e. the cost of producing 1 m of
CC divided by the total self-field critical current $I_c$ of a tape with a width of 1 cm [4–8]. Depending on the working conditions of the power system, the corresponding $I_c(B,T)$, where $B$ is the magnetic field and $T$ is the temperature, needs to be considered to estimate the suitability of the corresponding approach to CCs [4, 7]. The total critical current of a CC is defined as $I_c(B,T) = I_c(B,T)$ where $I_c(B,T)$ is the critical current density and $t$ is the film thickness. A common challenge in all the manufacturing approaches to CCs is, therefore, to increase the REBCO film thickness $t$ as much as possible, while $I_c(B,T)$ remains high through different strategies to optimize vortex pinning [9–13].

Several CC architectures and growth methodologies, which greatly differ in achieved performance, prospective cost and manufacturing scalability, are being investigated worldwide at present. The most widely spread group of growth methodologies are those where deposition and growth occur simultaneously, for example: pulsed laser deposition, metalorganic chemical vapor deposition, e-beam evaporation (EV) or hybrid liquid phase epitaxy [14–20]. A second wide group of growth methodologies is characterized by sequential deposition and growth steps. These are, for example, the techniques based on chemical solution deposition (CSD) of metalorganics, such as trifluoroacetates (TFA), or EV using either BaF$_2$ as an intermediate compound or the reactive co-evaporation by deposition and reaction process [21, 22]. Very different strategies have been followed to increase the REBCO film thickness under these two broad groups of CCs, mainly because the degree of understanding and control of nucleation and growth of epitaxial layers greatly differs. In both groups of techniques YBCO films of several microns have been demonstrated with $I_c$ values at 77 K and self-field in excess of 700 A/cm-w [14, 20, 21, 23–25].

CSD with TFA precursors is an approach to CC manufacturing with the potential for low cost production and capital investment, which has also been demonstrated to be competitive in terms of performance [26–34]. However, the issue of increasing film thickness in CSD still remains a complex problem, which has been poorly investigated and even less for the ‘all-chemical’ CC architectures, where the buffer layers are also grown by CSD [29, 34–36]. CSD relies on preparing metalorganic solution precursors, which are first deposited on top of buffered metallic substrates, either RABiT or ion beam assisted deposition, and then heat-treated following two separate steps [6, 29, 37, 38]. The first step corresponds to a pyrolysis process, where the organic compounds are decomposed into oxides and fluorides (typically at ~300 °C). The second corresponds to the epitaxial film nucleation and growth (typically at 750 °C–800 °C). Up to now, the most widely followed approach is that based on TFA precursors with different amounts of fluorine [37, 39–42]. Solution deposition is performed, in most cases, by spin coating at the laboratory scale and by web coating or dip coating in continuous reel-to-reel processes. Inkjet printing (IJP) has also been shown to be an appealing long length approach [43–45]. All these processes have been primarily used to obtain thin films ($t \leq 500$ nm) and little effort in growing thick films ($t \geq 800$ nm) has been reported [6, 22, 24]. In all cases, a single deposition is made to prepare thin films, while thick films usually require multideposition, i.e. repetitive deposition and pyrolysis steps. In principle, no limitations with respect to increasing film thickness through multideposition should be envisaged. However, multideposition followed by a pyrolysis step is a cumbersome process and so, it is highly desirable to increase the film thickness achieved in a single deposition as much as possible.

The greatest difficulty in increasing the film thickness in CCs, following the TFA approach, is the poor control of the high temperature nucleation and growth processes, when BaF$_2$ is used as an intermediate compound. The complexity of the process is associated with the control of the gas–solid reaction leading to the growth of YBCO [22, 29, 34, 38, 46]:

$$\text{Ba(O}_2\text{F})_2 + \frac{3}{2}\text{CuO} + \frac{1}{4}\text{Y}_2\text{O}_3 + y\text{H}_2\text{O} (g) \rightarrow \frac{1}{2}\text{YBa}_2\text{Cu}_3\text{O}_{6.5} + 2y\text{HF} (g)$$

(1)

Depending on the selected processing parameters, during the heating stage, the intermediate precursors formed after the pyrolysis of the metalorganics will display a different chemical and microstructural evolution [27, 29, 47–50]. This may induce other intermediate phases to also play a role in the YBCO film formation. For example, if $\text{Y}_2\text{Cu}_2\text{O}_5$ is formed, the growth of YBCO proceeds through a competing reaction and it may remain as a secondary impurity [22, 29]:

$$\text{Y}_2\text{Cu}_2\text{O}_3 + 4\text{BaO} + 4\text{CuO} \rightarrow 2\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$$

(2)

Detailed theoretical and experimental analyses of YBCO epitaxial film development by heterogeneous nucleation and growth, based on reactions (1) and (2), have been performed. The relevance of the different processing parameters has been verified when the YBCO films are in the thick limit [22, 29, 51–53]. On the other hand, in the case of thick films, the definition of the required processing parameters still remains strongly based on empirical analysis [51]. However, it is very important to achieve a good understanding of the influence of processing parameters on the nucleation and growth steps in the thick limit, in order to increase the $I_c$ values of CCs based on this approach [52, 53].

In the thin film limit, the thermodynamic parameters controlling the formation of stable c-axis nuclei by overcoming the nucleation energy barriers $\Delta G^*$ (energy barrier for heterogeneous nucleation) have been previously identified by several authors [29, 46, 51, 54]. In this limit, we can assume that the HF gas can freely diffuse outside the film precursor and then, the supersaturation degree $s$ ($s = \Delta\mu/kT$) and $\Delta G^*$ can be estimated on the basis of purely thermodynamic parameters defining the formation of independent nuclei at the interface [46, 51, 55]. For example, the nucleation barrier of nuclei with a cylindrical shape can be written as:

$$\Delta G^* = \frac{\pi(h\gamma_{int})^2}{-(\frac{\pi}{2})\Delta\mu - (\gamma_{up} + \gamma_{int} - \gamma_{sub})}$$

(3)
Figure 1. A simplified schema of the correlation between gas flow along the film and across the precursors of the YBCO thick films and the YBCO heterogeneous nucleation events at the substrate interface. The already formed nuclei are indicated together with the position of locations of potential new YBCO nuclei. $P(\text{HF})$ at the interface with the substrate is influenced by the previous nuclei already having been formed in the thick limit and so the nucleation events are correlated. In the thin film limit, the HF and H$_2$O gas diffusivity through the film is high enough to avoid any correlation among nucleation events and so they can be modelled individually. Schema derived from [51].

where $v$ and $\gamma$ represent the unit cell volume and surface free energy per unit area (lat = lateral surface, up = upper surface, int = interface, sub = substrate), $\Delta \mu$ is the chemical potential change per molecule between the epitaxial YBCO phase and the precursor random nanocrystalline state (equation (1)). The analysis of the behavior of the energy barrier $\Delta G^*$ indicated that when the supersaturation ($s$) is small the c-axis orientation is preferred [29, 46, 54].

Unraveling how nucleation and growth occurs in YBCO thick films based on the BaF$_2$ intermediate still remains an open issue because additional phenomena need to be taken into account. A remarkable effort has been made related to films where BaF$_2$ deposition was undertaken through EV [22, 26, 27, 51, 56, 57], however, thick films prepared by CSD have not been analyzed as much [24, 29, 32, 58].

In the thick film limit, the supersaturation degree controlling the heterogeneous nucleation of an island at the substrate interface becomes correlated with the nuclei previously stabilized and already growing. Thus, controlling nucleation and growth of these films becomes a much more complex phenomenon [51, 52]. As a general rule, in this process the advancement of the formation of YBCO crystals requires that $P(\text{HF})$ is significantly different from the equilibrium value $P_c(\text{HF})$ at the growing interface (advancement of equation (1)) and so, removal of HF gas is crucial to control the supersaturation degree [52, 53]. In thick films the $P(\text{HF})$ drop across the porous precursor film is determined by the already stable nuclei and influences the supersaturation degree at neighboring sites (figure 1). Therefore, the nucleation process becomes a collective phenomenon. This is the main reason why different deposition methodologies (BaF$_2$ evaporation or CSD), having different solid precursor properties and initial phases, need to choose different processing parameters [50].

The nucleation rate $dN/dt$ ($N =$ number of stable nuclei at the substrate interface) is a kinetic parameter linked to the height of the nucleation barriers $\Delta G^*$ (energy barrier for heterogeneous nucleation), hence, the surface energy of the cap layers and the interfacial energy ($\gamma_{\text{surf}}$ and $\gamma_{\text{int}}$ in equation (2)) will also strongly influence the formation of epitaxial films [22, 59]. The nucleation rate is an important parameter, which determines the choice of cap layers when growing thick YBCO films. For example, CeO$_{2-x}$ cap layers have a low lattice mismatch with YBCO and so, it facilitates heterogeneous nucleation, although nucleation catalytic effects associated with surface oxygen non-stoichiometry have been mentioned as playing a significant role [60, 61].

On the other hand, chemical reactivity of CeO$_{2-x}$ with a BaF$_2$ precursor in solution-based films may lead to formation of BaCeO$_3$ (BCO) layers, which then strongly degrades the YBCO film texture (large lattice mismatch among both oxides). Therefore, tight control of the degree of advancement of reaction (1) versus that of the BCO layers is very important to avoid or minimize this last undesired reaction [61−63].

Overall, it is worthwhile further investigating the influence of several competing processing parameters involved in the growth of ‘all-chemical’ thick YBCO films and CCs, in order to advance in the development of materials with large $I_c$ values.

In this work, we report our investigation of the nucleation and growth conditions of YBCO thick films (~1 $\mu$m thick). We first investigated the growth on LaAlO$_3$ (LAO) and CSD/CZO/YSZ single crystal substrates as model systems (figure 2). LAO is a stable substrate that is not reactive with Ba and so, allows us to concentrate on YBCO film growth for large thicknesses. In addition, the CSD/CZO/YSZ substrates need to consider the chemical reactivity with the CZO cap layer. Several multilayered architectures have been investigated, to generate further understanding of the role played by different parameters on the nucleation and growth steps of films with different thicknesses (figure 2). Then, we use this knowledge to grow YBCO films on CSD/CZO/ABADYSZ/stainless steel (SS) metallic substrates from Bruker HTS (ABAD = alternating beam assisted deposition). This architecture has previously been shown to be a suitable CC architecture for thin YBCO films [17, 43]. During growth on LAO substrates, we can concentrate solely on controlling the supersaturation conditions determining nucleation and growth in the thick film limit. In the other two cases, we also need to minimize the interfacial chemical reactivity with the CZO cap layers to keep good control of the nucleation process. We will show that through the use of Ag-TFA additives in a YBCO seed layer, c-axis nucleation becomes more robust, thus, allowing the growth of thick films and CCs on top of CZO cap layers with improved performances (figure 2).

Overall, we define a set of nucleation and growth parameters, which allow achievement of thick YBCO films with high critical currents using both IJP and multipideposition approaches for solution deposition. The microstructure and superconducting properties of these thick films have been characterized and it is found that fairly high critical current densities have been obtained so far: $I_c^{\text{ss}} = 390$ A/cm-$w$ in single crystal substrates and $I_c^{\text{ss}} = 100$ A/cm-$w$ in CCs, at 77 K and self-field. We finally discuss future paths to
further increase the film thickness and the critical currents of ‘all-chemical’ CCs.

2. Experimental details

2.1. Film preparation

For the deposition of thin and thick layers of YBCO, we have used 5 × 5 mm LAO or CSD/CZO/YSZ single crystals and CSD/CZO/ABAD/YSZ/SS metallic substrates (table 1). The metallic substrates have been provided by Bruker HTS, Germany [17]. The tapes are 4 mm wide and 0.1 mm thick. On top of these substrates, CSD/CZO cap layers were deposited by spin coating, although they can also be produced by IJP [44, 45, 64]. These CZO cap layers were grown at 900 °C during one hour in air, they are ∼25 nm thick and atomic force microscopy (AFM) analysis indicates that they have a rms (root mean square) roughness of ∼1.6 nm [61, 65]. In the case of single crystals, an rms roughness of ∼0.9 nm was systematically achieved.

The low-fluorine metalorganic precursor solution for the deposition of thin YBCO layers is composed of two acetates (barium acetate, Sigma Aldrich, and copper acetate anhydrous, Alfa Aesar—Cymit Química) and one TFA (yttrium trifluoroacetate anhydrous, 99.99+% Sigma Aldrich). They are dissolved in methanol and propionic acid in a molar concentration of 1.5 M (sum of metals). The fluorine reduction of this precursor solution is ∼80%, as compared to the all-TFA route [39]. From this precursor solution, ∼200 nm thick YBCO layers are obtained by single deposition spin coating, at 6000 rpm for 2 min. The pyrolysis of this type of solution has been described in detail elsewhere [39]. For the thicker films (t ≥ 700 nm), single pass IJP or spin coating multideposition was used (although multiple pass IJP has also been demonstrated as feasible) (table 1). The precursor solution for IJP had to be modified to achieve suitable properties for IJP deposition [43, 44]. Therefore, the solvent was changed from methanol to butanol and the molar concentration of this low-fluorine precursor solution was decreased down to 1 M (sum of metals). Prior to the deposition by IJP, two additives were added to the YBCO solution: one additive was used to modify the interaction between the ink and the substrate, which was provided by KAO Chimigraf Company (Rubi, Spain). The other, only used when indicated, was 5 molar% of Ag-TFA (99.99%, Aldrich) (table 2). As we will comment later, Ag salts decrease the supersaturation of reaction (1) and so, allow us to

Figure 2. Schema of the different multilayered architectures investigated for model thin and thick films grown on single crystal substrates and in a coated conductor architecture. The single crystals are used as model systems to focus on particular aspects of nucleation and growth. The use of Ag additives allows the promotion of c-axis nucleation at lower temperatures due to a reduced supersaturation.

Table 1. Solution deposition methodologies used for different substrates and the corresponding film thicknesses achieved.

| Substrate | Deposition method | Achieved thicknesses |
|-----------|------------------|---------------------|
| LaAlO₃ single crystals | Spin coating | 200–300 nm |
| | Single deposition | 1000–1200 nm |
| | Multideposition | 1000 nm |
| CSD/CZO/YSZ single crystals | Spin coating | 200–300 nm |
| | Single deposition | 1000–1050 nm |
| CSD/CZO/ABAD/YSZ/SS | Spin coating | 200–300 nm |
| | Single deposition | 1000 nm |

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...an FEI Quanta 200 FEG scanning electron microscope was used. The landscape of the structural defects of these layers was studied by transmission electron microscopy (TEM), using an FEI Titan 60–300 microscope equipped with an X-FEG gun, a CETCOR probe corrector and a Gatan TRIDEM 866 ERS energy filter operated in scanning TEM (STEM) mode at 300 kV. The superconducting properties were measured inductively, using a superconducting quantum interference device (SQUID) magnetometer (Quantum Design, San Diego, CA). The critical current density (Jc) was calculated using the Bean critical state model of a thin disk [66].

Besides XRD, the CZO cap layers were also subjected to a morphological study using AFM. The equipment used for this purpose was an Agilent 5100 with silicon tips and the topography of the samples was obtained using a tapping mode. The images were processed with a Mountains Map Premium 7.2 software. Besides the quantification of the roughness, this software permits a quantification of the flat area fraction of the surface of these CZO cap layers. To do this, we have to first define a threshold height value above which the surface is not considered flat. In our case, we have used a 1.5 nm threshold value, which represents one and a half fluorite unit cell height. Then, using the threshold, value a binary mask is applied to the whole AFM image. The result is a two color image, where the flat areas are represented by the bright sections [65]. For the CZO cap layers used in this work, flat area fractions between 62% (in the $^{53}$CZO/YSZ/SS metallic tapes) and 90% (in the $^{53}$CZO/YSZ single crystals) have been quantified.

### 3. Results and discussion

#### 3.1. Growth of low-fluorine thick YBCO layers on LAO single crystals

In this section, we will report an analysis of the growth process and the superconducting properties of thin and thick YBCO films grown on LAO substrates as a model system. The processing conditions to grow YBCO thin films from low-fluorine precursors on LAO substrates (<300 nm) with...
Figure 3. (a)–(c) Results corresponding to low-fluorine YBCO 200 nm thick films deposited on LAO single crystals: (a) the 2D-XRD $\theta$-2$\theta$ pattern, (b) the integrated spectrum of the pattern shown in (a), (c) the SEM image. (d)–(f) Results corresponding to low-fluorine YBCO 800 nm thick films deposited on LAO single crystals and grown in the same conditions: (d) the 2D-XRD $\theta$-2$\theta$ pattern, (e) the integrated spectrum of the pattern shown in (d) and (f) an SEM image.

Figure 4. (a) Low field ($H \sim 20$e) zero-field cooling magnetization versus temperature measurements used to determine $T_C$ and $\Delta T_C$. (b) and (c) Cross section TEM images of the YBCO 200 nm film after growth. The observed lines correspond to stacking faults.
optimized superconducting properties have been previously investigated and they are described in the experimental section (tables 1 and 2). In figure 3(a) we present a typical 2D-XRD pattern of a 200 nm thick YBCO film grown on LAO single crystals and in figure 3(b) the corresponding integrated θ-2θ spectrum is shown.

As it is observed, the YBCO thin layers are highly epitaxial, i.e. no random or a-b nucleation and no intermediate or unreacted phases are present. The lack of a-b grains is also confirmed by the SEM image (figure 3(c)) that reveals a homogeneous surface with low porosity of the YBCO layer. These thin YBCO layers exhibit reproducible excellent superconducting performances: \( J_c(77 \text{ K}) = 5 \text{ MA cm}^{-2} \), \( T_c = 91.2 \text{ K} \) and \( \Delta T_c = 1.8 \text{ K} \) (figure 4(a)). Also, they exhibit a very homogeneous microstructure showing only a few YBaCuO defects (Y248) stacking faults (figures 4(b) and (c)), whose atomic structure and distribution has been recently described in detail [67–69]. This solution becomes, therefore, an excellent candidate for the multideposition of thick YBCO layers by the spin coating technique.

We prepared thick YBCO layers, both by multideposition and by single deposition IJP, as described in the experimental section (table 1). Optical microscopy images recorded after the pyrolysis showed, in both cases, film homogeneities similar to that of thin films. However, when these thick films are grown following the same processing conditions of high quality thin films, a strongly perturbed texture is achieved (see the XRD patterns and SEM image in figures 3(d) to (f)), indicating that some random nucleation occurs under these processing conditions. This means that the degree of supersaturation was excessively high for films with increased thickness. It is clear, therefore, that to achieve high quality YBCO thick films the processing conditions need to be deeply re-examined.

As we have explained in section 1, in the thin film limit, the nucleation process of epitaxial YBCO films can be described on the basis of the formation of independent nuclei stabilized when its energy overcomes the energy barrier described in equation (3).

On the other hand, the theoretical analysis of the heterogeneous nucleation process in epitaxial YBCO thick films based on the BaF2 approach (equation (1)) was first performed by Solovyov et al [22, 51]. These analyses clarified that consideration of the thick limit requires extending the schema of the classical nucleation theory exemplified by equation (3) to the consideration of how the HF gas flow balance induces collective effects in the nucleation events. While in the thin film limit, it could be safely assumed that there was no influence of the HF gas impedance towards the film surface, in the thick film approach this effect needs to be explicitly taken into account [51–53]. The main conclusion in the analysis of nucleation in thick films is that the supersaturation degree \( s = \Delta \mu / kT \) in a high surface energy site, i.e. a site with a retarded nucleation event, is influenced by the already existing stable nuclei, i.e. the nucleation process becomes a collective effect (figure 1). The supersaturation of the sites with delayed nucleation could then be correlated with several processing parameters:

\[
s = \frac{\Delta \mu}{kT} \propto \frac{dr}{P(H_2O)^{1/2}D_s} \tag{4}
\]

where \( d \) is the film thickness, \( r \) is the growth rate and \( D_s \) is the HF diffusion permeability through the film precursor. It is clear from equation (4) that the supersaturation degree \( s \) is directly influenced by the film thickness and so, we can understand that the processing conditions need to be modified to grow thick films. Based on the predicted dependence of supersaturation for the processing parameters, we decided to investigate new nucleation conditions, mainly by increasing the \( P(H_2O) \) values and reducing the growth rate \( r \). The temperature dependence of YBCO growth rates \( r \) has been previously investigated by several authors [70–73] and it was found to follow the thermally activated behavior arising from the equilibrium constant \( K_c \) of reaction (1) [72]. For example, \( r \) was found to decrease by a factor of 2.5 when reducing the growth temperature from 820°C to 770°C. Concerning \( P(H_2O) \), our experimental set-up allowed us to increase it by a factor of 10. Overall, therefore, supersaturation \( s \) is predicted by equation (4) to be reduced by a factor of \( \sim 7 \) in our modified nucleation experimental conditions of thick films (table 2). Although the growth experiments of YBCO thick films under these modified conditions showed a significant improvement concerning texture quality, the presence of a/b grains oriented perpendicular to the substrate was not fully eliminated and so, further improvement of the nucleation process was still required (figures 5(a)–(c)).

To further enhance the texture quality of the YBCO thick films, we decided to implement in our growth process of thick films the use of an additional YBCO-Ag seed layer (50 nm thick) at the substrate interface. In previous investigations, it was shown that, due to the decreased peritectic temperature of the Ag-YBaCuO\(_{7-x}\) system, the supersaturation degree is decreased under non-modified processing conditions, compared to the YBaCuO\(_{7-x}\) composition [43, 74]. As a result, enlarged temperature windows for c-axis nucleation, reduced final YBCO film porosities and enhanced film planarities were achieved [46]. This strategy was found to be indeed successful, as it is concluded from the analysis of the results reported in figures 5(d)–(f) corresponding to a 1.2\( \mu \text{m} \) thick film prepared by spin coating multideposition (five deposition repetitions with the first being a YBCO-Ag seed layer) (table 1). The film texture quality was clearly improved: no a/b nucleated grains were detected in the XRD pattern and only a few a/b axis nucleated grains could be observed in the SEM images. Only minor traces of Y\(_2\)O\(_3\) are visible in both cases (using or without Ag) and so, we expect a near completion of the reaction (1).

Even if the new supersaturation conditions lead to full c-axis nucleation and growth, we could still go further and investigate whether there is any influence of the solution deposition methodology on the final film quality. For example, it is known that during the pyrolysis there is a tendency to segregate a CuO layer at the film surface and thus, when using a multideposition approach to prepare thick films we are facing the problem of compositional inhomogeneity.
Figure 5. The growth of thick YBCO films under modified supersaturation conditions: (a) the 2D-XRD θ-2θ pattern of a thick YBCO film grown on LAO single crystals without Ag additives; (b) the integrated XRD spectrum of the pattern shown in (a); (c) the SEM image of the thick YBCO film presented in (a) and (b); (d) the 2D-XRD θ-2θ pattern of a YBCO film (1.2 μm thick) grown on LAO single crystals including Ag additives; (e) the integrated spectra of the pattern shown in (d); and (f) the SEM image.

Figure 6. (a) The FIB image of a multilayered pyrolyzed YBCO thick film (~1 μm after growth), (b) the high-angle annular dark-field (HAADF)–STEM image of the same film after quenching it prior to the nucleation temperature of YBCO, (c) the FIB image after pyrolysis of a single deposition IJP YBCO thick film (~1 μm after growth), (d) the STEM image of a YBCO film deposited by IJP after quenching from the nucleation temperature before starting the growth process.
This is clearly visible in figures 6(a) and (b), where we display a focused ion beam (FIB) cross section image of a film with five pyrolyzed layers (final film thickness \( \sim 1 \mu m \)) and a cross section TEM image of the same film quenched after heating it up to the growth temperature, respectively.

As we can see from the FIB image, between each deposited and pyrolyzed layer some CuO interlayers are formed. Therefore, the high temperature processing conditions chosen to grow these films need to promote Cu atomic diffusion out of these interlayers to be able to form YBCO. Eventually, this diffusion process may lead to the formation of some layered porosity associated with the phase transformations and shrinkage occurring during the heating ramp (layered pores in figure 6(b)) [23, 31, 56]. This effect can be minimized, however, by following different strategies. For example, if the layer thickness of each deposition is small the Cu diffusion length is large enough to overcome the initial inhomogeneous atomic distribution. In addition, through careful intermediate high temperature annealing treatments, the films can be compacted and homogenized [26, 27]. Therefore, achieving optimized nucleation and growth conditions of thick YBCO films involves careful processing requirements, to minimize the effect of the undesired CuO interlayers and the associated porosity that, eventually, may be created after the film growth.

An alternative approach to minimize this source of film inhomogeneity is to perform a single pass solution deposition and pyrolysis step for the thick films. This is, in principle, possible by using IJP, where the total amount of salts deposited can be controlled electronically (table 1). The use of this deposition methodology for CSD YBCO thin films has been previously described [43–45], while its extension to thick films and the corresponding optimization of the pyrolysis process will be presented elsewhere. Applying this deposition method, we were able to achieve homogeneous films, from one single deposition, up to 1 \( \mu m \) thick onto LAO single crystals, without any CuO interlayer phase separation after the pyrolysis process and so with reduced film porosity after the heating stage (figures 6(c) and (d)). The FIB image confirms that the microstructural and compositional homogeneity is indeed preserved following IJP and a single pyrolysis process. Actually, the difficulties with keeping thick homogeneous films in a single deposition are associated with both the solution deposition and drying steps and the pyrolysis treatment, where buckling or cracking should be avoided [75]. Using the IJP technique and applying the new supersaturation nucleation and growth condition for c-axis nucleation in thick YBCO films we were able to grow YBCO layers with thicknesses in the range of 0.6–1 \( \mu m \) having high texture qualities and good superconducting performances. In figures 7(a)–(c) the 2D-XRD patterns of a 600 nm thick YBCO layer on LAO single crystals and the corresponding SEM image are shown. We see that at this film thickness the YBCO layers are highly epitaxial. When the YBCO film thickness is increased \( \sim 1 \mu m \) the texture begins to degrade and a small amount of random grains is observed (figures 7(d)–(f)). Besides the texture degradation, also confirmed by some broadening of the (001) XRD Bragg peaks, we can also detect the presence of some unreacted crystalline phases (in this case Y\(_{2}\)O\(_3\)). This indicates that the supersaturation conditions for c-axis nucleation at these high film thicknesses still requires some improvement, even if no a–b nucleation is detected in the XRD patterns and SEM images (figures 7(d)–(f)).

Our success in achieving films with a thickness in the range of \( \sim 0.6–1 \mu m \) approaches the best results registered so far in other functional CSD films [76]. In figures 8(a)–(c) we present TEM cross section images and energy dispersive x-ray microscopy (EDX) compositional analysis of a 1 \( \mu m \) thick single deposited IJP layer after the growth process. The TEM image shows that the porosity has been drastically reduced, as compared to the thick YBCO layers deposited by the multideposition method and grown using a conventional high temperature annealing process. The TEM and EDX images in figures 8(a)–(c) show that some secondary phases persist (BaO, BaCuO\(_x\)). This defect structure, however, does not show any sign of the layered porosity observed in the multideposition process (figure 6(b)), thus suggesting that further optimization of the nucleation and growth processes of IJP films should allow further improvement of the microstructural homogeneity.

An overall view of the superconducting performances of the YBCO layers with different thicknesses deposited and grown on LAO single crystals is reached looking at the thickness dependence of \( J_c \) and \( I_c \) at 77 K and self-field (figure 9). Thin films in the range of 200–300 nm display \( J_c(77 K) = \) values around 4–5 MA cm\(^{-2}\) and \( I_c(77 K) = \) \( \sim 120 \) A/cm-w. Thick films prepared using multideposition by spin coating or single pass IJP, with thicknesses in the range 0.6–1.2 \( \mu m \), preserve fairly good critical current densities: \( J_c(77 K) = \) 2–2.7 MA cm\(^{-2}\) and \( I_c(77 K) = \) 240 A/cm-w is the highest value registered on LAO substrates, corresponding to a 1 \( \mu m \) thick film. These \( I_c(77 K) = \) values were obtained using inductive SQUID magnetization measurements. We should note that these values of the critical currents are close to those achieved by other vacuum-based film deposition techniques [14, 15, 18, 20, 21]. Finally, figure 10 displays the measured temperature dependence at self-field \( I_c(T) = \) for several films, which summarizes the success in developing thick YBCO films on LAO substrates. A detailed analysis of the in-field, temperature dependence and orientation of \( J_c(H,T,\theta) \) is an issue related to the vortex pinning landscape of CSD YBCO films. This topic goes beyond the scope of the present work, but it has been recently analyzed in detail in relationship with the development of CSD nanocomposite YBCO films [77, 78].

### 3.2. Growth of low-fluorine thick YBCO layers on \( \text{cspd Ce}_0.9Zr_{0.1}O_2/YSZ \) single crystals

As we have previously mentioned, when growing YBCO films on CeO\(_2\)-related cap layers there is an interfacial reactivity issue which needs to be properly handled (figure 2) [62, 63]. This reactivity takes place between the BaF\(_2\) precursor of equation (1) and the CZO cap layer and it leads to the formation of BaCeO\(_3\) (BCO). The formation of this
Figure 7. (a)–(c) The 2D-XRD $\theta$–$2\theta$ pattern, integrated spectra and SEM image of a 600 nm thick single deposition IJP YBCO layer; (d)–(f) the 2D-XRD $\theta$–$2\theta$ pattern, integrated spectra and SEM image of a 1 $\mu$m thick single deposited IJP YBCO layer on LAO single crystals.

Figure 8. (a) The HAADF—STEM image of a grown thick IJP YBCO layer (final thickness of 1 $\mu$m), (b) the cross sectional HAADF image showing the YBCO/LAO interface, (c) the cross section the STEM image and corresponding electron energy loss spectroscopy—spectrum image compositional maps of O (K edge), Ba (M edge) and Cu (L edge) in red, green and blue, respectively. BaCuO$_x$ and BaO secondary phases are identified.
Figure 9. The evolution of $I_c(T)_{77K}$ with YBCO film thickness using different deposition and growth techniques for single crystalline and metallic substrates. Dashed lines indicate the slopes corresponding to different critical current densities $I_c$ at 77 K.

Figure 10. The temperature dependence of $I_c(T)_{77K}$ for different YBCO films having modified thicknesses and grown on different single crystalline substrates.

perovskite and the nucleation of YBCO on CZO are two competitive processes. If YBCO nuclei appear first, then we have chances to induce c-axis nucleation on top of the CZO cap layers. The formation of an epitaxial BCO layer will take place later underneath the YBCO layer and will not affect the texture of the YBCO in any way [61]. However, if BCO is formed first at the interface a significant amount of nucleation with random orientation occurs for both the BCO and YBCO layers and so the superconducting properties of the film are degraded.

Previous works on YBCO thin film growth on CSDCZO/YSZ single crystals showed that a key parameter to achieve high $I_c(77K)$ values is the planarity of the CZO cap layers [61]. High performance was achieved with CSD-grown YBCO thin films (thickness $\sim$300 nm): $I_c(77K)_{c} = 4$–5 MA cm$^{-2}$ [61, 65]. These optimal performances are achieved when the surface planarity of the CZO cap layer is above $\sim$70%. The reactivity issue can then be properly handled and the supersaturation conditions can be selected to nucleate c-axis oriented YBCO islands. Typically, the growth conditions for these YBCO thin films were $P(H_2O) = 22$ mbar and annealing temperature $T \sim 770$ °C–780 °C [61, 65] (table 2). When Ag-TFA additives are used in the solution, the nucleation temperature can be reduced down to $\sim$700 °C (table 2).

Now, our objective here is to use the knowledge generated when growing YBCO thick films ($\sim$1 μm) on LAO substrates (section 3.1) to adapt the process to CSDCZO/YSZ single crystal substrates, as a model system for ‘all-chemical’ CCs. The strategy was to mimic, as much as possible, the processing conditions used for LAO and to introduce fine adjustments when necessary. To ensure that the crystallization window of YBCO remained as wide as possible, we included the Ag-TFA additive, either in a YBCO seed layer or in all the YBCO layers of a multideposition process. We focused our nucleation and growth analysis on YBCO films with a final thickness of $\sim$1 μm obtained after five sequential depositions by spin coating with the corresponding pyrolysis processes, as described in the experimental details in section 2 (table 2). Our previous nucleation analysis of YBCO thin films on CSDCZO/YSZ substrates determined that interfacial reactivity was low enough at $T < 780$ °C. We therefore kept this temperature while the low supersaturation $s$ (equation 4) was kept by increasing $P(H_2O)$ (table 2), i.e. $P(H_2O) = 200$ mbar.

Figures 11(a)–(c) show a GADDS XRD pattern, the corresponding integrated XRD pattern and an SEM morphological image, respectively, of a 1.050 nm thick YBCO film grown on top of a CSDCZO/YSZ single crystal. The XRD patterns show that the YBCO film is highly epitaxial while the CZO cap layer has been fully converted to epitaxial BCO. As it is observed, the microstructural quality of the film is fairly good and with just some small pores observed in the SEM image. Figures 12(a)–(e) show several cross section TEM images of such YBCO thick films, grown under the above-mentioned conditions. The low resolution cross section TEM image (figure 12(a)) displays a lower residual porosity than the YBCO thick films grown on LAO substrates (figure 8(a)), while a few secondary non-superconducting phases (BaCuO$_2$, Y$_2$O$_3$, Y$_2$Cu$_2$O$_5$) still remain (figures 12(b)–(d)). Figures 12(b)–(e) also show that the concentration of Y248 stacking faults is similar to that observed in YBCO films grown on LAO single crystals (figures 4(b)–(c)). We suggest that the nucleation rate of YBCO films is higher when using CZO cap layers owing to its lower lattice mismatch ($\varepsilon = -0.52$%) than with LAO ($\varepsilon = -1.56$%) because under these unstrained conditions the nucleation barrier (equation 3) usually decreases [59]. When the nucleation rate is increased, merging of individually nucleated YBCO grains is easier because the density of the nuclei is larger and thus the required atomic diffusion lengths are shorter. As a consequence, the microstructures tend to be more homogeneous. This is an issue that has already been previously discussed by other authors, who suggested that secondary phases could be accumulated at the merged YBCO grain boundaries [79].
Very competitive superconducting performances have been achieved so far in these CSDCZO/YSZ thick films (∼1 μm) (see figure 9): $J_{c}(77\,\text{K}) = 3.7\,\text{MA}\,\text{cm}^{-2}$ and $I_{c}(77\,\text{K}) = 390\,\text{A/cm-w}$ or $J_{c}(65\,\text{K}) = 7.2\,\text{MA/cm}^{-2}$ and $I_{c}(65\,\text{K}) = 750\,\text{A/cm-w}$ (figure 10). These values are very close to the best performances achieved so far in vacuum-deposited YBCO films with similar film thicknesses [14, 15, 18, 20, 21]. Therefore, the competitiveness of the ‘all-chemical’ CC approach is confirmed. As we mentioned before, TEM images show that some residual porosity and secondary phases still remain. This suggests that further improvement of the microstructural and superconducting characteristics of the all-chemical CSD-YBCO/CSD-CZO/YSZ architecture should be achieved through optimized processing conditions of the CZO cap layer and the YBCO layer.

3.3. Growth of $\text{YBa}_2\text{Cu}_3\text{O}_7$ films on metallic substrates

The ‘all-chemical’ CC architecture we selected to develop is CSD-YBCO/CSD-CZO/ABAD-YSZ/SS (figure 2). The SS metallic substrates with YSZ deposited by ABAD on top were provided by Bruker HTS from Germany and are described in the introduction of this article.

The CSD-CZO cap layers were deposited using spin coating, although IP can also be used for long length CC production [43, 44] and grown according to the thermal treatment described in the experimental section (table 2). In order for these cap layers to be suitable, they have to comply with some requirements: they have to be epitaxial, they have to exhibit a low roughness and a high degree of surface flatness. The influence of the CZO cap layer flatness on YBCO film growth was previously investigated and it was concluded that the critical current density $J_{c}$ and the film texture quality is improved when the film flatness is enhanced [61, 65]. Therefore, we performed a complete analysis of all these features (epitaxy, roughness and flatness) in our CZO cap layers [44, 61]. The CZO cap layers exhibit a high quality epitaxy as only (00l) Bragg peaks are observed. The AFM measurements reveal rather low values of roughness, ∼1.6 nm for an area of 5 × 5 μm² and ∼1.3 nm for an area of 1 × 1 μm². For the same areas, we notice that the surface flatness of the CZO layer (measured as the percentage of the area with heights differing by less than 1.5 nm) is higher than 60% [43, 80].

The reactivity issue discussed in the previous section meant that the growth conditions of thick YBCO films on CSD-CZO/YSZ single crystal substrates needed to be revised when growth is performed on metallic substrates. This is because the enhanced surface roughness of the CZO cap layers increases the interfacial chemical reactivity and, very likely, it also modifies the heterogeneous nucleation barriers $\Delta G^*$ for YBCO islands and thus, their nucleation rate.
To comply with all the requirements for obtaining high critical current CCs we first investigated the temperature where the interfacial chemical reactivity is slow enough in YBCO thin films (∼200 nm). To promote c-axis nucleation we included YBCO-Ag seed layers in our architecture (figure 2). Two annealing steps at different temperatures were used, one at 700 °C to nucleate the film and another at 770 °C, to complete the film growth. The two-step annealing process becomes necessary to minimize the residual porosity in the YBCO films [27, 46]. As is shown in the XRD patterns of figures 13(a) and (b), a fairly good YBCO film texture was achieved, i.e. only sharp single (001) Bragg peaks are observed in the GADDS XRD pattern. There is no evidence of a-b or random YBCO nucleation, an issue also confirmed in the SEM images (figure 13(c)). The typical values of the texture quality determined by XRD are $\Delta \omega = 2.5^\circ$ and $\Delta \phi = 6−7^\circ$ [45]. Once again, the CZO cap layer has been fully transformed to epitaxial BCO with two orientations perpendicular to the substrate [(200) and {110}]. These YBCO CCs exhibit high critical temperature, $T_c = 91$ K, and fairly high critical current density, $J_c(77K)^f = 1$ MA cm$^{-2}$, a value usually limited by granularity effects in these CCs [43, 80] and similar or slightly lower than those usually displayed by other vacuum deposition techniques [18, 20]. These nucleation and growth conditions were found to be robust, and allowed us to achieve a high reproducibility in the film qualities and the corresponding superconducting performance. We should stress that these results confirm that the use of YBCO-Ag seed layers allows developing c-axis nucleation at sufficient low temperatures.

The next step was to combine the knowledge generated to grow thick YBCO layers on LAO and $\text{CZO/YSZ}$ single crystal substrates and the knowledge about the kinetics of interfacial reactivity forming BCO layers, to achieve thick (∼1 μm) YBCO CCs with the ‘all-chemical’ architecture on $\text{YSZ/SS}$ substrates (figure 2). Owing to the advantages that the IJP deposition method provides for scaling-up the CC production process and grounded in the good results reported in section 3.1, we decided to deposit thick YBCO CCs using this deposition method. A single pass IJP deposition was used leading to a final YBCO film thickness of ∼1 μm on top of $\text{CZO/YSZ/SS}$ substrates. As we have described in sections 3.1 and 3.2 when the thickness of the YBCO layers is increased the supersaturation degree $s$ tends to increase (see equation (4)) and so the nucleating conditions need to be modified. At the same time, however, we need to take into account the modified kinetics of the interfacial reactivity reaction, when the processing conditions of thick films are used. Therefore, we need to handle simultaneously, both the control of the supersaturation degree $s$ and the interfacial reactivity. The increase in the YBCO film thickness by a factor of ∼5 has a strong influence on the percentage of c-axis nucleation achieved in our CC architecture. As in the case of single crystalline substrates, the supersaturation conditions used to grow thin films are again no longer valid here. We adjusted the supersaturation degree $s$ (see equation (4)), following the same approach used in single crystals, i.e. by increasing the water pressure to $P(H_2O) = 200$ mbar and by decreasing the nucleation temperature which directly influences the growth rate $r$. To disclose the optimum nucleation temperature in our CCs we performed short isothermal treatments followed by a quenching process to mainly analyze the nucleation stage.

The as-obtained samples were analyzed by XRD. Typical XRD patterns of such quenched films are presented in figures 14(a) and (b). It can be seen that at 700 °C YBCO nucleates randomly while a large fraction of unreacted crystalline phases is still present after 30 min of annealing at this temperature (figure 14(a)). We also noticed that no BCO has formed yet, i.e. at this temperature and under the selected processing conditions no BCO formation is detected. This is also confirmed by the presence of an intense (200) Bragg peak of the CZO film, which shows that no cap layer reactivity occurred. So, at this temperature the reactivity issue is controlled but we have random nucleation of YBCO due to an excessive supersaturation $s$. It is necessary, therefore, to increase the annealing temperature to optimize the nucleation process. If we look to the XRD pattern of a film quenched after annealing during 30 min at 730 °C (figure 14(b)), we can...
see that the fraction of randomly oriented YBCO grains ((103) YBCO Bragg peak) has drastically diminished while the unreacted precursor crystalline phases are no longer present. Taking into account all this information, we designed a double step annealing treatment for growing thick YBCO coated conductors. The first step was the nucleation stage at 730 °C (60 min) followed by the growth step at 770 °C–800 °C (135 min) (table 2).

Figures 15(a)–(c) show XRD patterns and a morphological image (SEM) of a fully grown 1 μm thick YBCO CC based on the new designed nucleation and growth treatment (table 2). Even though a small amount of randomly oriented YBCO grains are still present ((103) YBCO Bragg peak), it is noticeable that c-axis nucleation and growth is strongly favored under these new supersaturation conditions. Again, no a-b grains seem to be present, neither in the XRD patterns nor in the SEM image. These thick YBCO superconductors exhibit fairly good superconducting properties. For a single pass IJP deposition (~1 μm) a YBCO film was achieved exhibiting a critical temperature $T_c = 90$ K, a sharp transition with a width of $\Delta T_c = 6.5$ K (see figure 16(a)), and $J_c(77 \text{ K}) = 1 \text{ MA cm}^{-2}$ which corresponds to $I_c(77 \text{ K}) = 100 \text{ A/cm-w}$ (see figure 9). STEM cross section images of a 1 μm thick YBCO CC (figures 16(b)–(d) show that some porosity and secondary phases still remain and so our analysis is valid as a ‘proof-of-principle’ for how to grow ‘all-chemical’ CCs but there is still further room for improvement for both the film texture and the final microstructure. Figures 17(a)–(c) show a STEM cross section and EDX analysis of the interface in this CC where the formation of a BCO layer is also confirmed. As we have demonstrated in section 3.2, optimizing the nucleation and growth conditions on CSD/CZO/YSZ single crystals, where the cap layer planarization is higher, allowed us to reach much larger critical currents, i.e. $I_c(77 \text{ K}) = 390 \text{ A/cm-w}$. The refinement of the YBCO nucleation and growth conditions, as well as an increase in the CZO cap layer planarization, should allow us to further minimize the concentration of misoriented YBCO grains and decrease the residual porosity. Consequently, the highest performance achieved with single crystalline substrates would be within reach.

Finally, we would also like to stress that all the processes reported here to prepare ‘all-chemical’ CCs are essentially valid for preparation of CSD nanocomposite films and CCs. This enhances the performance in an extended range of magnetic fields and temperatures by introducing preformed nanoparticles in the initial metalorganic solutions [64].

**Figure 14.** (a) An XRD pattern of a YBCO coated conductor (IJP) treated at 700 °C for 30 min and (b) at 730 °C for 30 min and quenched in both cases. The YBCO layers correspond to thick films of 1 μm treated with $P_{\text{air}} = 200 \text{ mbar}$.

**Figure 15.** (a) A 2D-XRD $\theta$-2$\theta$ pattern, (b) integrated spectra of (a) and (c) SEM image of a 1 μm thick YBCO coated conductor with a two-step treatment, at 730 °C and 770 °C–800 °C, and including a YBCO–Ag layer.
is an issue, however, that will be reported elsewhere in the future.

4. Summary and conclusions

Understanding the influence of the processing parameters upon the supersaturation conditions for c-axis nucleation of YBCO layers derived from CSD, using low-fluorine metalorganic precursors, is crucial for the design of optimum growth treatments for both thick YBCO layers on single crystal substrates and on metallic substrates, to form ‘all-chemical’ YBCO coated conductors. In this work, we have described the whole process of solution deposition, pyrolysis, nucleation and growth of YBCO thick films (t ∼ 1 μm). In particular, we have analyzed the difficulties to overcome
during the nucleation and growth processes and we have proposed novel strategies to succeed in achieving high quality epitaxial thick films. Useful parameters have been described for all the steps, to achieve thick CCs with high $I_c$ values. A summary of the flow of the requirements, the typical hurdles found and how to achieve adequate film qualities at all the preparation steps, is indicated in figure 18.

Low-fluorine metalorganic solutions compatible with spin coating and IJP were defined. As a solution deposition method, IJP is presented as a very useful tool, with the potential to scale-up CC production, to achieve high film thicknesses ($\sim$1 $\mu$m) from one single pass deposition (table 1). Regarding the growth conditions of thick YBCO layers on LAO and ‘all-chemical’ CZO/YSZ single crystals, we have found that highly textured films can be prepared when Ag-doped YBCO layers are used, either as a seed or for the full film (table 2). These additives allow us to reduce the nucleation temperature, thus decreasing the interfacial reactivity and hence keeping a high quality c-axis nucleation for thick ‘all-chemical’ YBCO films. The best superconducting performance for an ‘all-chemical’ multilayer architecture was achieved in CSD CZO/YSZ single crystals, where we have demonstrated $J_c(77 K)^d = 3.7$ MA cm$^{-2}$ and $I_c(77 K)^d = 390$ A/cm-w in a 1.050 nm thick YBCO film. These total critical currents are very close to those achieved in vacuum-deposited films at similar film thicknesses.

During the growth of YBCO CCs, the interfacial reactivity with CSD CZO cap layers was found to increase due to a larger surface roughness. However, we could still define modified YBCO nucleation conditions using Ag–YBCO
films, which preserve a high texture quality. For 1 μm thick YBCO CC we have achieved $J_c(77 K)^0 = 1 \text{ MA cm}^{-2}$ and $I_c(77 K)^e = 100 \text{ A/cm-w}$ using the single pass IJP approach. XRD patterns and STEM images have shown that thick ‘all-chemical’ CCs still have room for microstructure improvement. Some porosity, secondary phases and film texture degradation was observed. This is an issue which we associate with the reduced surface planarity and texture quality of the substrate cap layers. It is very likely that the reduced performance, when compared to vacuum-deposited cap layers, arises from the reduced planarity. Further improvement in the superconducting performances of the all-chemical thick YBCO CCs should be achieved through improved surface finishing of the metallic substrates.

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