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Phase Evolution of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films by all-chemical solution deposition route for coated conductors

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Abstract. In order to understand the all-chemical-solution-deposition (CSD) processes for manufacturing coated conductors, we investigated the phase evolution of $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO) films deposited by a low-fluorine metal-organic solution deposition (LF-MOD) method on CSD derived $\text{Ce}_{0.9}\text{La}_{0.1}\text{O}_2/\text{Gd}_2\text{Zr}_2\text{O}_7/\text{NiW}$. It is shown that the phase transition from the pyrolyzed film to fully converted YBCO film in the LF-MOD process is similar to that in typical trifluoroacetates-metal organic deposition (TFA-MOD) processes even though the amount of TFA in the solution is reduced by almost one half compared with typical TFA-MOD cases. Moreover, we found that the formation of impurities (mainly BaCeO_3 , NiWO_4 and NiO) is strongly related to the annealing temperature, i.e., the diffusion controlled reactions become intensive from 760 °C, which might be connected with the poor structural and superconducting properties of the films deposited at high sintering temperatures. Based on these results, the optimized growth conditions of YBCO films were established, and a high critical current density (J_c) of about 2 MA/cm² (77 K, self field) is achieved in a 200 nm thick YBCO film in the architecture made by our all CSD route.

1. Introduction

Due to the great potential for their use in electric power applications, the YBCO based coated conductors have been extensively studied and developed during the past two decades. Long-length wires with superior performance have been successfully manufactured by several routes [1, 2]. Among those techniques, combining Rolling-Assisted Biaxially Textured Substrates (RABiTS) and chemical solution deposition (CSD) is considered as one of the most promising routes, which could easily be scaled-up and further reduce the costs due to initial investment and production. However, because of the lack of comprehensive understanding of CSD techniques, the industrialization of CSD processes for both buffer layers and superconducting layer (all CSD processes) is still quite challenging and rarely reported [3]. Further investigations of all CSD processes, including compatibility between the materials used in each layer, solution chemistry, as well as nucleation and growth mechanism are needed.

In our previous studies, we reported a CSD process for a full buffer layer architecture [4]. A critical current density (J_c) value higher than 1 MA/cm² (77 K, in self-field) was achieved in a YBCO film grown by pulse laser deposition, demonstrating the high quality of this buffer stack for coated conductors. In this work, we focused on an all CSD process, where a low-fluorine metal-organic



solution was used for depositing YBCO directly on the CSD derived buffer layer. The low-fluorine metal-organic deposition (LF-MOD) route chosen for this work has the purposes of shortening the pyrolysis process and reducing amount of the TFA in the precursor solution. In this work, the phase evolution of YBCO during the firing processes was investigated in order to establish the heat treatment profiles. We paid additional attention on transition of the intermediate phases, formation of YBCO phase and inevitable impurities (BeCeO_3 , NiWO_4 , NiO), all of which have strong influences on the superconducting performance of the final products. With the guideline of phase evolution, the growth conditions of the YBCO films were optimized.

2. Experimental

The propionate-based MOD route for preparing $\text{Ce}_{0.9}\text{La}_{0.1}\text{O}_2$ (CLO) and $\text{Gd}_2\text{Zr}_2\text{O}_7$ (GZO) buffer layer stack on textured NiW substrates was described previously [4, 5]. With optimized coating and heat treatment conditions, high quality buffered substrates were obtained routinely for further YBCO deposition.

The LF-MOD process for YBCO film deposition was similar to standard TFA-MOD, where the only difference in solution is that the copper salt is replaced by a non-TFA precursor. First, a solution (referred to as solution A) was made by dissolving home-made yttrium and barium trifluoroacetates in methanol with a Y/Ba ratio of 1/2, like in standard TFA-MOD solutions [6]. Secondly, copper acetate was dissolved in a small amount of acrylic acid (10% excess) and de-ionized water. After full reaction and removal of residual solvent and by-products by vacuum evaporation, a dried copper-based powder was obtained. The final precursor solution was made by mixing stoichiometrically solution A and copper-based powder at room temperature. The total cation concentration was 1.5 M by adjusting the amount of methanol. The precursor solution was deposited onto substrates by spin-coating with a rotation speed of 3000 rpm. A typical heat treatment procedure, including pyrolysis, sintering and oxygenation, was used for obtaining superconducting YBCO films. The pyrolysis process started from room temperature to 400 °C with a ramp of 1.7 °C/min in humid oxygen (dew point (D.P.) = 20 °C). Then partially reacted samples were quenched from different firing temperatures (400 °C, 600 °C, 700 °C, 740 °C, 780 °C, 820 °C) to study the phase evolution. All the firing processes were performed in a humid atmosphere (D.P.=20 °C, with 100 ppm oxygen). For the fully reacted YBCO films, following oxygenation was carried out at 450 °C for two or three hours in flowing oxygen gas.

The phase assembly and crystallographic textures were investigated by means of x-ray diffraction (XRD) using $\text{Cu K}\alpha$ radiation in a four-circle diffractometer (Bruker D8). The surface morphology of the films was observed by scanning electron microscopy (SEM, Supra 35) equipped with an inlens detector. The superconducting transition temperature (T_c) was determined by AC susceptibility measurements with AC field amplitude of 0.1 mT and frequency of 21 Hz, while the critical current densities, J_c , were calculated based on Bean model [7].

3. Results and discussion

θ - 2θ theta scans of the samples quenched from different temperatures are shown in Fig. 1. In the pyrolyzed film, we observed $\text{Ba}_{1-x}\text{Y}_x\text{F}_{2+x}$ (BYF, $x = 0.35$) and CuO phases, which are basically the same compositions as those in all TFA solutions [8]. When the sintering temperature reaches 600 °C, the peak shift of barium oxyfluorides can clearly be seen, which means that yttrium starts to dissolve out of BYF. The peaks corresponding to the BYF and CuO phases become narrow at the elevated temperatures up to 780 °C, indicating the grain growth of these phases. The BYF peaks nearly remain at the same positions after 700 °C, implying that almost all Y was released from the BYF matrix. However, since the peak of the Y_2O_3 the most likely Y phase, is too close to the buffer layer peaks, it is difficult to claim if there is any such phase forming at this stage. However, $\text{Y}_2\text{Cu}_2\text{O}_5$ (211) ($2\theta = 31.2^\circ$) clearly appears at 740 °C, probably due to the reaction between the Y_2O_3 and CuO. This could indirectly support the view of the formation of Y_2O_3 formation at an earlier stage. The first appearance of the YBCO (001) peaks occurs at temperatures lower than 760 °C, while the trace of random orientation (103)/(013) implies that the epitaxial growth is predominating under these conditions.

Regarding the impurities formation caused by elemental diffusion at the interlayers, we mainly focus on BaCeO₃ and NiWO₄ which are normally observed when using nickel based substrates. As early as 700 °C, we could observe a small amount of BaCeO₃ and BaCeO₃(110) peak became intense with increasing temperature. Compared with other reports where YBCO^{MOD} films were grown on CeO₂ cap layers made by physical deposition technique (i.e., sputtering or PLD) [9, 10], the formation of BaCeO₃ in our case takes place much earlier. This is probably due to the microstructure of the CLO^{CSD} cap layer, which leads to its high chemical activity [11]. However, it seems that under an optimized firing profile, BaCeO₃ has little negative influence on the superconducting properties in the final YBCO films, as will be shown in the following. The NiWO₄ peaks appear at 700 °C as well and keep developing at higher temperatures.

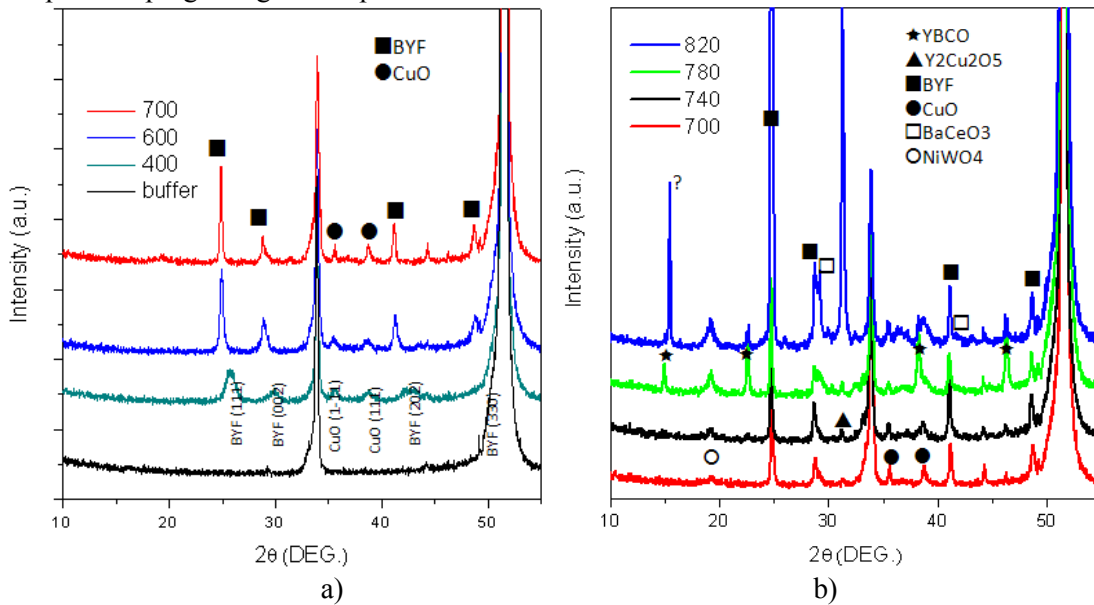


Figure 1. θ - 2θ scans of the buffered substrate and a series of YBCO samples quenched from different temperatures

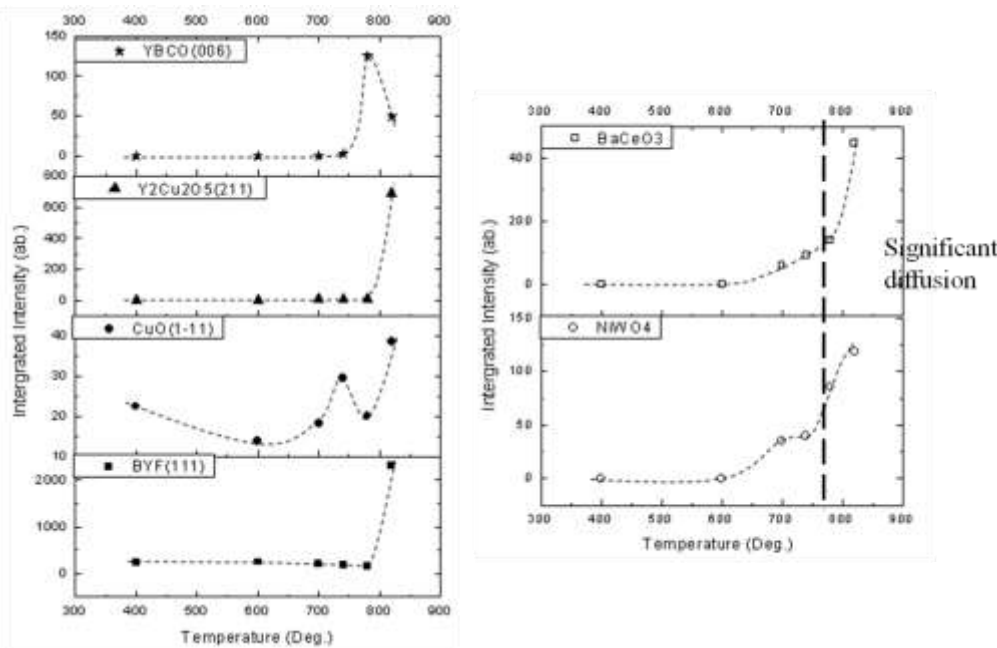


Figure 2 Integrated area evolution versus temperature of XRD peaks of intermediate phases (left) and impurities (right) of films.

The integrated peak area evolution of several phases during the YBCO sintering process are plotted as a function of temperature and presented in figure 2. It allows us to draw the evolution trends when the temperature increases. It can be seen that the $Y_2Cu_2O_5$ and BYF phase are still not converted completely at 820 °C due to the short dwell time. The dramatic increase of BYF (111) peak intensity is due to the texture formation. The drop of the YBCO (006) peak intensity is associated with both unknown phase formation and the complex reactions in the interlayer (as evidence by rapid increase of the $BaCeO_3$ content). Considering these results, we conclude that sintering YBCO films on such CSD-derived substrates at a temperature higher than 780 °C should be avoided. One should notice that the aforementioned phase evolution is based on the kinetics of the firing process up to 820 °C. In the practical cases, longer dwell time will change the starting formation temperatures of the intermediate phases and final products, e.g., YBCO (00l) peaks appear as early as 700 °C after 3 hours dwell.

According to the results of phase evolution, several sintering temperatures (from 700 °C to 780 °C with an interval of 20 °C) were chosen to optimize the growth conditions of YBCO films. Larger amount of grains with undesirable orientations can be discerned on the surface of the films sintered at temperatures above 760 °C, suggesting that the elemental diffusion significantly changes the YBCO nucleation and growth. On the contrary, less oxidation of the metallic substrate was obtained at lower sintering temperatures. Since the growth rate at 700 °C is very low, we could not obtain well crystallized YBCO film at this temperature with reasonable dwell time (e.g., 3 hrs). The films sintered at 720 °C and 740 °C exhibit sharp in-plane and out-of-plane texture quality, and J_c values of 2.1 MA/cm² and 1.0 MA/cm² were obtained, respectively, demonstrating the feasibility of the all-CSD route for coated conductors fabrication.

4. Conclusion

Phase evolution of YBCO grown on $Ce_{0.9}La_{0.1}O_2/Gd_2Zr_2O_7/NiW$ substrates by a low-fluorine metal-organic solution deposition method was studied. It is shown that the compositions in the pyrolyzed film are similar to those in typical TFA MOD films. Besides, we noticed that impurity formation (mainly $BaCeO_3$, $NiWO_4$ and NiO) becomes intensive from 760 °C, which might deteriorate the superconducting performance of YBCO layers. According to these results, the optimized growth conditions of YBCO films were established, and a J_c of 2 MA/cm² (77 K, self field) is achieved in such an all CSD derived architecture.

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