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Research paper

Thick REBaCuO superconducting films through single-coating of low-fluorine metallorganic solution

M. Boubeche *, C.B. Cai, H.B. Jian, M.J. Li, W.T. Yang, Z.Y. Liu, C.Y. Bai

Shanghai Key Laboratory of High Temperature Superconductors, Department of Physics, Shanghai University, Shanghai 200444, China

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ABSTRACT

A high critical current I_c is crucial for the application of high temperature superconductors $YBa_2Cu_3O_{7-\delta}$ in energy efficient power devices and wires.

In this paper we report the fabrication of thick $(YGd)_{1.3}Ba_2Cu_3O_{7-x}$ films on a metal substrate using low-fluorine metal organic deposition method. The effects of the film thickness on the microstructure, texture and superconductivity properties of the films were evaluated. In order to increase the film thicknesses by single coating, the influence of withdrawal speed during the dip coating on resulting thickness are investigated with the other processing parameters fixed. It is revealed that there is a maximum thickness for a certain starting solution. Here we used 3 different solutions, Conventional Low Fluorine solutions with 2 M and 2.5 M, and super low-fluorine solution with 2.5 M. The maximum thicknesses of about 710 nm, 1280 nm and 1460 nm were obtained, respectively.

1. Introduction

Over the years much research has focused on developing various deposition techniques for thin film superconductors. These techniques can be broadly divided into two categories, namely physical and chemical methods. The physical method consists of magnetron sputtering, pulsed laser deposition (PLD) [1,2] and co-evaporation [3]. It still remains a major challenge for most thin film groups to optimize the deposition method to produce high quality films. The chemical method is composed of the metal organic chemical vapor deposition (MOCVD) [4,5], liquid phase epitaxy (LPE) and metal organic deposition (MOD) methods [6–12]. Metal organic deposition (MOD) is one of the popular solution deposition methods, involving coating of an organic precursor solution on a substrate followed by thermal decomposition to form the final desired compound [6]. Solution deposition is a fast, cost efficient method and is applicable to large-area substrates. The advantages of MOD over other deposition techniques are rapid deposition rates, uniform and ease controlled composition.

In this work we used Conventional Low Fluorine (CLF) and Super Low Fluorine (SFL) MOD to reduce the amount of fluorine in the starting solution to enhance density and uniformity of the produced films as proved else [9,10]. It has been shown to be successful to get thick films of single coating by changing the withdrawal speed during the dip coating, and the starting solution concentration.

* Corresponding author.
E-mail address: rahma@shu.edu.cn (M. Boubeche).

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2. Experimental details

Starting solutions with different concentrations were prepared by dissolving gadolinium, yttrium, barium, and copper salts with the ratio of Y + Gd:Ba:Cu equal to 1.3:2.0:3.0. The solutions were synthesized using the Conventional Low Fluorine (CLF) solution with fluorine content of 54% (CLF2M and CLF2.5M), and a super low-fluorine (SLF) solution with fluorine content of 31% in order to reduce the pyrolysis time to less than 2 h. Details of the process are mentioned elsewhere [11,12]. The solutions concentrations were 2 mol/L and 2.5 mol/L. All the films were prepared by dip-coating method on metal substrate, heated by 5 °C/min up to 300 °C, then by 20 °C/min up to 780 °C, the films where held 120 min on 780 °C: 90 min in humid mixed gas, the following 30 min in dried mixed gas, after rapid cooling to 450 °C kept at 450 °C in dry oxygen for 60 min, finally cooled to the room temperature in a dry oxygen gas.

The texture, the thickness and critical current of crystallized films were examined respectively by X-ray diffraction (XRD), Brooker DEKTAKXT stylus and Cryoscan, THEVA.

3. Results and discussion

Using a concentration of the starting solution equal to 2.5 mol/l, YBCO films with various thicknesses where obtained by changing the withdrawal rate. The texture of the resultant films are shown in Fig. 1, illustrating a well defined *c*-axis orientation of all the studied samples. *a*-axis orientation became intense with increasing

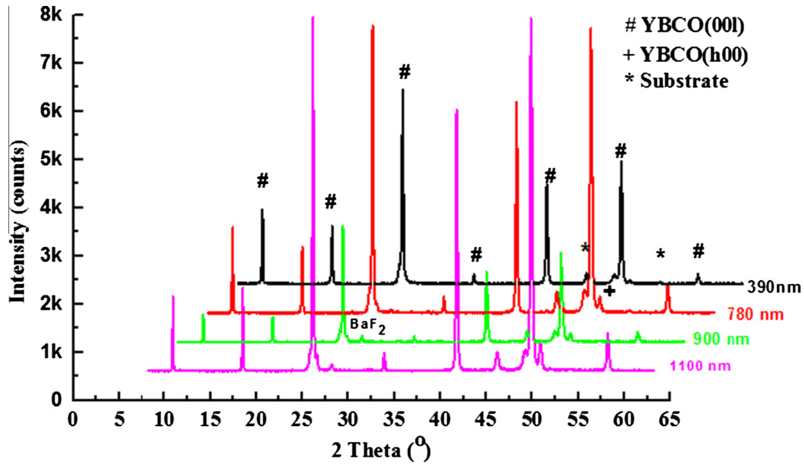

Fig. 1. XRD graphs for different films with different thickness using CLF2.5M solutions.

the thickness. Which implies that there is a certain quantity of *a*-axis grains in the thick films. Other peaks appeared around $2\theta \sim 26°$ it belongs to BaF2 [13], and it becomes more intense with the augmentation of the REBCO thickness. This may due to the insufficient holding time in the crystallization step, hence thick films need long crystallization time.

Fig. 2 represents the thickness dependence on the withdrawal speed as well as starting solution. Using 3 different solutions and by increasing the withdrawing speed the thickness trend is the same, with increasing withdrawal speed (*P*), thickness first increases, then reaches a peak, then drops. The thickest resultant films using CLF2M, CLF2.5M and SLF2.5M solutions are respectively 710 nm, 1280 nm and 14,600 nm. Interestingly, for each solution, a maximum thickness is experimentally found with corresponding withdrawal speed *P* which differ from the previous reported [7,8,11]. Some models have been proposed to describe the formation of a homogeneous fluid layer by dip-coating. All of them are based on the Landau-Levich model [14,15], which predicts that the equilibrium thickness depends on the density, the surface tension, and the viscosity of the fluid and is proportional to the withdrawal speed at the power of 2/3. An extended Landau-Levich model, predicts a dependence on withdrawal speed at the power of 1/2 [14,15].

However, these models do not agree with our results especially when the withdrawing speed is high. It may be due to the physical–chemical properties of solutions used, the substrate surface, the heat treatment conditions (the high crystallization temperature), and also the chemical reaction involved in obtaining the final crystallized films, as well as some pores may generated in the films that enlarged the thickness. These results suggest that further work is needed to develop a model that can explain the observed dependence on the withdrawal speed.

Fig. 3 shows the dependence of the maximum thickness obtained when the solution concentration is simultaneously increased and the corresponding withdrawal speed *P* is decreased. The results indicate that one key to improving the thickness by one-time coating is to select an appropriate withdrawal speed based on starting concentration. The relationship between the precursor film thickness and withdrawal speed of the dip-coating using different starting solutions was reported previously [10–16].

The thickness and critical current ratio (I_c/I_{cmax}) dependence on the withdrawal speed for the films produced from CLF2.5M solution is shown in Fig. 4. The I_c ratio initially increases as expected as the film thickness increases. However, I_c ratio value appears to decrease with a thickness over 780 nm. One possible reason is that as the withdrawal speed increases the film becomes too thick for the film to grow well. In the plateau region, as withdrawal speed increases, increasing the thickness seems to not affect I_c. Referring to the thickness variation with withdrawal speed, the I_c is the same for the samples of equal thickness. Commonly if the films grow well, I_c would increase with thickness, and then reach a saturation. However, in our work, the highest I_c value does not correspond the thickest film. That may due to the highest withdrawing speed may

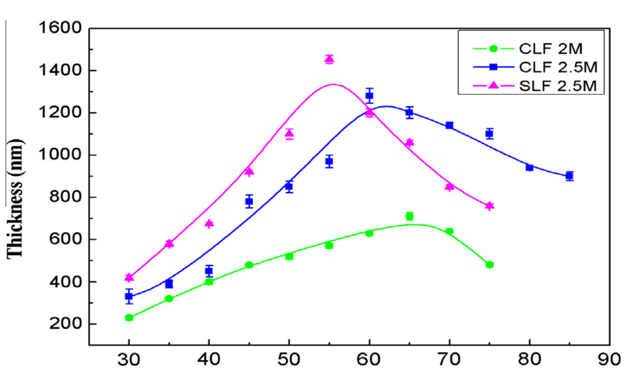

Fig. 2. Thickness dependence on withdrawal speed using the 3 various solutions.

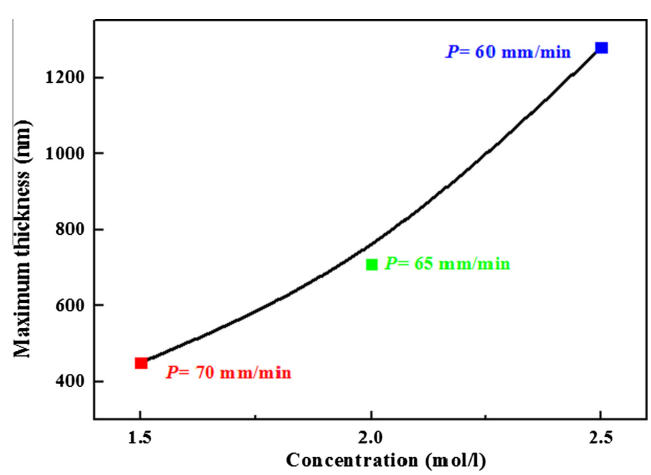

Fig. 3. The maximum thickness variation with the concentration derived from CLF solution.