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Fluorine-free propionate route for the chemical solution deposition of $YBa_2Cu_3O_{7-x}$ superconducting films

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Abstract

The environmentally friendly fluorine-free propionate chemical solution deposition route has been developed for the epitaxial growth of $YBa_2Cu_3O_{7-x}$ (YBCO) superconducting films. In the present work, metal acetates are used as reagents to prepare a YBCO fluorine-free precursor solution. In order to determine the thermal decomposition behavior, the precursor powder obtained by drying the precursor solution was investigated by thermogravimetric analysis coupled with mass spectrometry and differential thermal analyses. The precursor films were deposited on $SrTiO_3$ single crystal substrates by spin coating and subjected to a single-step thermal treatment. Fourier Transformed Infrared spectroscopy was used to study the decomposition of the films. The films have been structurally and morphologically characterized by X-ray diffraction and atomic force microscopy, respectively, and the superconducting properties have been measured by SQUID. The film composition was investigated by Auger electron spectroscopy. The as-deposited YBCO films show a sharp texture and good superconducting transport properties with $T_c=90$ K ($R=0$) and $J_c=0.8$ MA/cm^2, respectively.
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1. Introduction

Due to its high critical current density (J_c) and irreversibility line (H_{irr}), the high-temperature superconducting $YBa_2Cu_3O_{7-x}$ (YBCO) films are the most used one for the second-generation (2G) high-temperature superconductor (HTS) wires/tapes fabrication. The clue towards the fabrication of 2G-HTS tapes is the epitaxial deposition of YBCO thick films on a biaxially oriented oxide template built on flexible metallic substrates [1–5]. The final YBCO film must meet stringent requirements, such as: a high degree of epitaxy, uniformity across the width and along the length ($\sim 10^3$ m) of the tape, as well as excellent mechanical properties. To achieve these properties, many efforts have been focused on the development of chemical solution deposition (CSD) route for YBCO deposition over the last decade. The CSD route has several advantages since, no vacuum equipment is needed, permits a good stoichiometry control, is low-cost and it is suitable for the long length superconducting tapes fabrication process [6]. The CSD of the YBCO films mainly involves four steps: precursor synthesis, coating, precursor decomposition and film crystallization. The understanding and the control of the precursor deposition and its conversion to YBCO are critical for the reproducible manufacturing of uniform, high-performance, HTS wires required for commercial applications.

The trifloroacetates metalorganic deposition (TFA–MOD) of the YBCO films is the most investigated route because it can produce high J_c superconducting films [7,8]. The main disadvantage of the TFA–MOD is the long duration of the pyrolysis step that limits the overall production rate and the liberation of highly corrosive and toxic HF during the YBCO crystallization step. To overcome this disadvantage, several research groups are trying to reduce the fluorine content using low-fluorine [9–12], and fluorine-free coating solutions [13,14] starting from different precursors and

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solvents, which can avoid the HF formation. Metal acetates, acetylacetonates, nitrates and triethanolamine, ethylene glycol, propionic acid chelating agents were used for the preparation of the precursor coating solutions. For example, triethanolamine (TEA) is used to increase the solubility of acetates in methanol/water [14], since the –OH groups provided by the TEA molecules can be deprotonated and the formation of mixed carboxylate–TEA complexes occurs [15].

The formed complex ensures the homogenous distribution of the metal ions. Moreover, the propionic acid is an excellent solvent for dissolving acetates because it can be obtained in high concentrations and has good wetting behavior at the substrate surfaces. However, the fluorine-free solution has a major drawback. The formation of $BaCO_3$ phase during the heat treatment can degrade the superconductivity of the YBCO films. Several authors report on the successful precursor decomposition during the heat treatment and obtained YBCO films with high critical current [16–18]. These studies have revealed that $BaCO_3$ can react with CuO to generate the $BaCuO_2$ phase which then reacts with Y_2O_3 to form YBCO, thus achieving the elimination of the $BaCO_3$ in the final films. This process takes place at high temperatures in low oxygen partial pressure environment.

In this paper a fluorine-free propionate coating solution was used for YBCO superconducting films. The non-fluorine precursor solution was prepared by dissolving Y-, Ba-, Cu-acetate in propionic acid using methanol as a solvent. Glycerol was introduced to the final solution as a chelating agent, in order to increase the pH and to prevent segregation and precipitation. The films have been characterized by X-ray diffraction (XRD) and atomic force microscopy (AFM). The magnetization versus magnetic field measurements, $M(H)$, were performed using a SQUID magnetometer. Film chemical analysis and depth compositional profiling were performed using Auger electron spectroscopy (AES).

2. Experimental procedure

The non-fluorine precursor solution was prepared by separately dissolving the Y-, Ba- and Cu-acetates in methanol and propionic acid (C_2H_5COOH), were neutralized with NH_4OH until the solution became clear. The three solutions were mixed together under stirring and concentrated by the removal of solvents under vacuum (153 mbar, 75 °C for water) resulting in a blue–green coating solution. The chelating agent, glycerol, was added to the final solution. The addition of glycerol improves the precursor solution viscosity and stability. Moreover, glycerol addition increases the pH value up to 5.4–7. The stock solution was stable in air, with a shelf-life longer than two months. The concentration of the final precursor solution was 1.5 M. The coating solution was deposited on $SrTiO_3$ (STO) using a spin coater with a rotation speed of 3000 r/min followed by drying the precursor film at 80 °C for 5–10 min. The as-deposited precursor films were subjected to a single-step thermal treatment, as follows: up to 300 °C the films were thermally treated at a heating rate of the 3 °C/min; between 300 and 600 °C the heating rate increases up to 5 °C/min under humid oxygen atmosphere. The pyrolysis step at 600 °C for 30 and 60 min was made to ensure the complete decomposition of the organic parts. The crystallization treatment has been performed at 850 °C, at a rate of 10 °C/min, for one hour both in humid oxygen, and nitrogen environment. The film was cooled down to 450 °C in the same gas at a rate of 10 °C/min, held at this temperature for one hour in oxygen atmosphere and cooled down to room temperature.

The precursor powders were analyzed by thermogravimetric and differential thermal analyses (TG–DTA) to determine their decomposition mechanism. For the gases evolved during the thermal decomposition processes, TG–DTA has been coupled with mass spectrometry (MS) with m/z=10–90. The precursor powder has been obtained by drying the coating solution on a hot plate at 60 °C and afterwards heated up to 800 °C with a heating rate of 5 °C/min.

Fourier Transform Infrared (FTIR) spectroscopy for the quenched films was performed using a Bruker Tensor 27 FTIR Spectrometer in the ATR (Attenuated Total Reflection) mode. All spectra were normalized with respect to the highest band in the 400–4000 cm^{-1} range.

The structural properties and the texture of the YBCO films were analyzed by means of the X-ray diffraction technique. The X-ray θ–2θ and ω-scans were performed with a Bruker D8 Discover X-ray diffractometer using the Cu $K\alpha$ radiation. The film chemical analysis and the depth compositional profiling were performed by using Auger Electron Spectroscopy. The morphology of the final films was examined by atomic force microscopy (AFM—Veeco). The electrical characterization of the YBCO films was performed by the four points method. The magnetization versus magnetic field measurements, $M(H)$, were performed by SQUID magnetometry. The critical current density, J_c, was derived from the $M(H)$ curves using the Bean critical state model.

3. Results and discussion

For the optimization of the conversion process of the precursor film to the final highly textured films is necessary to carefully investigate the decomposition of the precursor powder. The thermal analysis of the precursor powder was performed under humid oxygen to avoid the sublimation of the copper precursor which could lead to off-stoichiometry in the final films. Araki et al. [19] reported that the humid atmosphere during pyrolysis process restricts cooper sublimation.

The TG–DTA curves of the precursor powder annealed in humid oxygen are shown in Fig. 1a. The TG curve shows two significant weight losses in the temperature ranges 20–260 °C and 240–550 °C, respectively. The weight loss below 260 °C is due to the water evaporation and coordinated solvent molecules. The MS signal for m/z=17 (H_2O) (Fig. 1c) is represented in a logarithmic scale in order to emphasize the small variations of the p_{H2O} background due to the thermal decomposition of the sample. In this stage the DTA curve indicates an endothermic peak at 150 °C and the TG curves show a weight loss of 35%. In the second step (260–550 °C) the DTA shows a strongly exothermic process (465 °C) indicating the decomposition of the precursors. The MS (Fig. 1b) has revealed that the evolved gases

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