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Microstructures and phase evolution in $YBa_2Cu_3O_{7-x}$ films grown on various substrates fabricated via a non-fluorine sol–gel route

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Abstract

$YBa_2Cu_3O_{7-x}$ films were grown on polycrystalline Ag and single crystalline YSZ, $SrTiO_3$, and MgO substrates using non-fluorine sol–gel and spin coating methods. The effects of heat treatment conditions on the phase evolution and microstructures were investigated using optical microscope, X-ray diffraction, and atomic force microscope. For Ag substrates, Y123 phase started to form at 750 °C and higher sintering temperatures improved the degree of (0 0 1) texture. Mirror-like surfaces without any cracks were achieved for sintering at 750–900 °C. However, voids were observed for films grown on the Ag substrates at temperatures higher than 810 °C and their size and density increased as the temperature increased. For the films grown on single crystal substrates sintered at 800 °C, numerous microcracks with large crack widths were observed, while cracking is less of a problem for films grown on Ag substrates. Epitaxial films without any cracks were achieved for films grown on single crystalline substrates under optimized conditions. Possible mechanism for the formation of cracks is discussed.

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Keywords: YBCO; Sol–gel; Thin film; AFM; XRD; Optical microscope; Microstructures

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1. Introduction

The chemical solution deposition methods, such as metalorganic deposition using the trifluoroacetates method (TFA-MOD method) [1] or the sol–gel spin coating method have various advantages in forming thin film of $YBa_2Cu_3O_{7-x}$ (Y123) superconductor compared to physical techniques. These include a low cost non-vacuum approach, precise controllability of metals composition, wide flexibility as to the shape of the object to be coated [2,3]. In addition, for Y123, the chemical solution method has high reproducibility of the critical current density [4,5] due to the fact that the crystallization mechanism of Y123 by TFA-MOD method does not take place via solid phase epitaxy from the amorphous precursor as has been confirmed in the conventional MOD route, but it is a chemical conversion process involving water and fluorides [6]. Large areas Y123 films with high-J_c can be easily produced from a purified coating solution, prepared by the solvent-into-gel (SIG) method [7]. Since fluorides attack substrates during heat treatment [8], a chemically compatible buffer layer, for which CeO_2 is mostly used, is indispensable for protecting the metallic substrates [9].

The possibility of obtaining *c*-axis grain orientated Y123 thick films on silver alloy substrates via high-temperature melt processing has also been reported [10,11]. Such a thick film approach may serve as an alternative to the vapour deposition methods. Recently, a non-fluorine sol–gel method was developed for making Y123 thin film [12]. Because Ag is the only flexible metal supporting material which does not react with Y123, it is very desirable if the Y123 can be grown directly on the Ag without any buffer layers. The non-fluorine sol–gel method, which does not attack Ag, may be the best candidate for making large-scale Y123 films or tapes on Ag substrate. Furthermore, we have noted from early reports that Y123 thin films grown on YSZ single crystal substrates exhibit a serious cracking problem with big crack widths [1]. These films were produced by MOD using precursors consisting of mixed metal (2-ethylhexanoates) and have no preferred orientations after heat treatment at temperatures up to 900 °C. Understanding the crack formation mechanism and elimination of cracks are important issues for the preparation of high quality films for coated conductors using solution route. The purpose of this study was to investigate the crack formation in Y123 films made by the non-fluorine sol–gel method, and to understand the factors, which control the cracks formation and the *c*-axis alignment of the film. In this paper, we presented our detailed studies on cracking microstructure, and phase formation of Y123 films grown on (100) oriented single-crystal substrates of yttrium-stabilized zirconia (YSZ), MgO and $SrTiO_3$ (STO), and on polycrystalline Ag substrates.

2. Experimental

Our non-fluorine-based sol–gel Y123 solutions were developed in-house. For the precursor solution, stoichiometric (1:2:3) yttrium trimethyl acetate, barium hydroxide, and copper trimethyl acetate powders were dissolved in a mixture of propionic acid/amine (amylamine) solvent with an oxide concentration between 0.1 and 0.5 mol/l. The addition of amine was important because it greatly improved the solubility of the precursor powders in propionic acid. The stock solution is stable in air with a shelf life longer than two years. Xylenes was used for dilution and for controlling the solution viscosity at between 10 and 100 cp. The films were deposited on different substrates by spin coating at 2000–3500 rpm and were dried at 200–300 °C for several minutes. This process was repeated several times until the desired thickness of the film was achieved. The films were sintered in air between 600 and 900 °C for 0.1–0.5 h, then slowly cooled down to 400 °C and annealed at this temperature for 24 h in a pure O_2 atmosphere then furnace cooled down to room temperature. DTA/TGA curves were recorded in air for the sol–gel solution to gain the information on the solution evaporation, decomposition and phase formation. The substrates used in this study were polycrystalline silver, single crystalline YSZ(100), MgO(100) and $SrTiO_3$(100). The phases, morphologies, and microstructures were investigated in terms of X-ray diffraction, optical microscope, and atomic force microscope.