Cryogenics 72 (2015) 1–8

Contents lists available at ScienceDirect

Cryogenics

journal homepage: www.elsevier.com/locate/cryogenics

Influence of $YBa_2HfO_{5.5}$ – 'derived secondary phase' on the critical current density and flux-Pinning force of $YBa_2Cu_3O_{7-\delta}$ thick films

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A R T I C L E I N F O

Article history:
Received 9 July 2014
Received in revised form 23 March 2015
Accepted 30 May 2015
Available online 17 June 2015

Keywords:
Critical current density
Flux pinning
Perovskite
Nanoparticles
YBCO

A B S T R A C T

Enhancement in critical current density (J_c) and flux pinning force (F_p) in superconducting thick films of $YBa_2Cu_3O_{7-\delta}$ (YBCO) added with small quantities of nanopowders of HfO_2, $BaHfO_3$ and $YBa_2HfO_{5.5}$, coated on $YBa_2ZrO_{5.5}$ substrate by dip-coating technique is reported. Critical current density measurements were done over an applied magnetic field using standard four probe technique and the results are compared with that of pure YBCO. High critical current density (J_c) of ~4.84 MA/cm^2 at 77 K in self-field was obtained for 2 wt% of $YBa_2HfO_{5.5}$ added YBCO. A systematic increase in J_c observed in YBCO films prepared by the addition of nano HfO_2, $BaHfO_3$ and $YBa_2HfO_{5.5}$, attributed to the formation of a non-reacting 'derived secondary phase' $YBa_2HfO_{5.5}$ (YBHO) in the YBCO matrix. YBCO–$YBa_2HfO_{5.5}$ composite thick films have showed eightfold increases in J_c (3.29 MA/cm^2) at 77 K and 0.4 T compared to pure $YBa_2Cu_3O_{7-\delta}$ film (0.37 MA/cm^2), while maintaining a high transition temperature (T_c). The development of effective pinning centers in nano particle added YBCO thick film have enhanced the flux pinning force from 1.8 GN/m^3 for pure YBCO to a maximum value of 13.15 GN/m^3 for YBCO–$YBa_2HfO_{5.5}$. X-ray diffraction and energy dispersive spectroscopic analysis confirmed the presence of secondary phase, derived in the matrix.

1. Introduction

High Temperature Superconductor (HTS) thick films are being engineered for high current applications in high magnetic fields such as in magnetic shielding, low phase noise oscillators, magnetic resonance imaging receiver coils, flux transformers, fault current limiter, filters, persistent-current and HTS magnets which need high critical current density (J_c) [1–6]. Since J_c decreases with both thickness and magnetic field, artificial pinning centers (APC) in addition to conventional ones are required to keep J_c high. Several nano-engineering techniques were developed to insert and control APCs, thereby dramatically enhancing the electrical transport performance of rare earth barium superconductors [7–14]. Introduction of precipitates of secondary phases or non-superconducting inclusions can act as isotropic, randomly dispersed artificial pinning centers in the superconductor system [15]. There are reports that nanostructured APCs exhibit enhanced pinning of flux in magnetic field, especially at temperatures lower than 77 K due to the effect of randomly dispersed strongly correlated defects [16–18]. However Augieri et al. had reported that $BaZrO_3$ nanorods are less effective flux pinner at low temperature [19].

Understanding the correlation between J_c and the structural defects responsible for the flux pinning in the thick films is vital for improving current carrying capabilities of a coated conductor. Small scale defects, when they are present in large numbers, can effectively pin magnetic flux, thus enabling high critical currents in external magnetic fields. Obviously, defects cannot be added indiscriminately, since they also reduce the effective cross section of the conductor and degrade structural quality of the superconducting matrix, eventually negating the benefits of enhanced pinning [20]. According to Feldmann et al. uniformly added Y_2O_3 nanoparticle layers and short $BaZrO_3$ nanorods can enhance critical current density in YBCO thick films and had reported that 'J_c versus thickness' dependence is not an insuperable intrinsic barrier for producing higher J_c values [21]. A twofold improvement in J_c value by the addition of $BaTiO_3$ in YBCO films was reported by Jha et al. [18]. Similarly, rapid enhancement in flux pinning and critical current density was observed in YBCO films by the incorporation of nanoparticles such as $BaSnO_3$ [9], $BaHfO_3$ [22] Ba_2YTaO_6 [23], Ba_2YNbO_6 [24,25] and Ba_2RETaO_6 [26]. In our earlier works, we had reported the effect of addition of simple oxide, HfO_2 [27], ABO_3 type perovskite, nano $BaHfO_3$ [28,29] and double perovskite $YBa_2HfO_{5.5}$ [30] in bulk YBCO superconductor and observed a

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http://dx.doi.org/10.1016/j.cryogenics.2015.05.011
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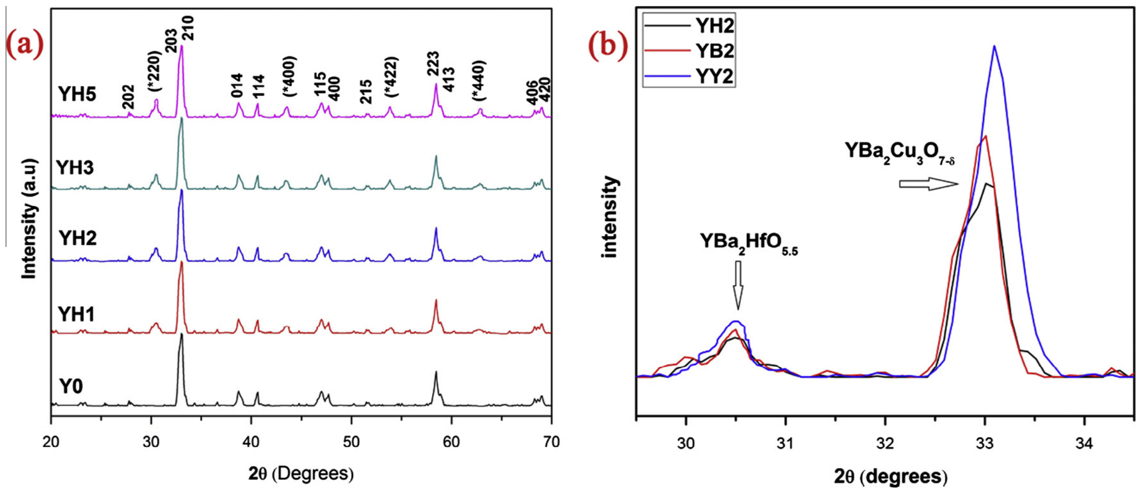

Fig. 1. (a) XRD spectrum of HfO_2 added YBCO thick film sample (b) peak intensity variation with 2 wt% addition of nanoparticles.

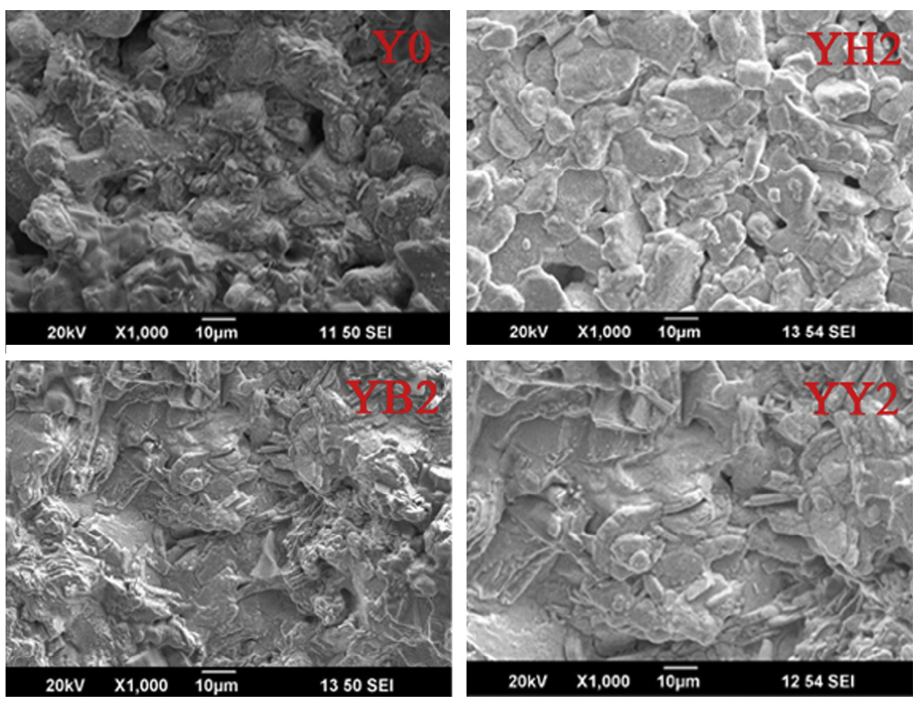

Fig. 2. SEM images of pure and 2 wt% of nanoparticles added thick films.

tremendous improvement in flux pinning force and critical current density.

Osamura et al. had suggested that perovskites could be beneficial for incorporation into YBCO due to their similar crystal structures relative to the host YBCO phase [31]. Thereafter double perovskites were being largely considered also as substrate materials for YBCO films. High flexibility in lattice parameter and magnetic behavior had made double perovskites, of general formula $A_2(BB')$-O_6, a potential candidate. And Paulose et al. had reported that $YBa_2ZrO_{5.5}$ [32] as a good substrate material for the deposition of $YBa_2Cu_3O_{7-\delta}$ films. In this work we report the influence of nanoparticles of HfO_2, $BaHfO_3$ and $YBa_2HfO_{5.5}$ as artificial pinning centers on the critical current density and flux pinning force of YBCO thick films on an $YBa_2ZrO_{5.5}$ substrate, in applied magnetic fields. Through detailed characterizations, we have correlated different levels of nano particle content with microstructural as well as electrical transport properties of YBCO films.

2. Experimental

Conventional technique of solid state route was used for the preparation of YBCO superconductor, by which high purity Y_2O_3, $BaCO_3$, and CuO were thoroughly mixed in the stoichiometric ratio of Y:Ba:Cu = 1:2:3. The mixture was then calcined at 930 °C for 72 h with two intermediate wet grindings. The nano particles of HfO_2, $BaHfO_3$ and $YBa_2HfO_{5.5}$ were prepared through a modified auto-igniting combustion process. In a typical synthesis, aqueous solution containing ions of Y, Ba and Hf was prepared by dissolving high purity Y_2O_3 in dilute HNO_3, $Ba(NO_3)_2$ and $HfOCl_2$ (99%) in