Development of $YBa_2Cu_3O_7$-Ba_2YTaO_6 nanocomposites by chemical solution deposition

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Abstract. In this work we report the study of chemical solution deposited (CSD) $YBa_2Cu_3O_{7-\delta}$ (YBCO) nanocomposite thin films, with diluted Ba_2YTaO_6 (BYTO) nanoparticles. We studied the influence of thermal treatment on the properties of nanocomposites, with different content of secondary phases (6%, 10% and 20%). We measured the film microstructure, nanostrain and the superconducting properties: critical temperature (T_c) and critical current density (J_c). The films were characterized using X-rays diffraction and SQUID inductive measurements. The use of two steps in the thermal treatment allowed to increase the nanostrain up to 0.30% for high nanoparticle loads (20%BYTO), decrease the nanoparticle size down to 15 nm and lead a smoother J_c(H) dependence, compared with the standard (single-step) thermal process and the pristine YBCO films.

1. Introduction

The applied research on superconductors, in which transport of large currents is required, has led today to the development of tapes, based on high-T_c superconductors (HTS). These kind of tapes are called: second generation – 2G, or coated conductors, and are usually based on an YBCO layer, deposited onto a metallic substrate. These devices present a great potential for applications, given that, in addition to the competitive high values of J_c achieved, higher than 1 MA/cm^2, its manufacture can be easily scaled up to produce long lengths, by chemical methods, at low cost. A usual method of fabrication consists in depositing a superconducting layer, from a chemical solution onto a metallic substrate. To increase J_c different methods are being investigated, in order to enhance vortex pinning. An attractive and economical method is the chemical solution deposition route- CDS, in which a structure of non-superconducting or nanocomposites, is diluted inside the superconducting matrix [1].

YBCO nanocomposite materials have opened a new path to enhance vortex pinning in HTS. Fabrication of nanostructured YBCO thin films with the incorporation of nanosized oxide-secondary phases, has shown strong enhancement of YBCO performance at high magnetic fields, in a wide temperature range, ensuing the potential for use in a broad number of power applications [2-6]. Recently, it has been demonstrated that the presence of metal oxide nanoparticles in the YBCO matrix generates nano-scale defects, which act as vortex pinning centers, improving its performance. Rare-earth niobiates and tantalates (double perovskite $Ba_2RE(Ta,N)O_6$ and pyrochlore Re_3TaO_7 with RE= Yb^{+3}, $Gd^{+3,}$ Er^{+3}) have gained attention as effective artificial pinning centers [7]. CSD is a cost-effective technique for preparation of large area and long length films [6]. Trifluroacetate (TFA) metal-organic decomposition (MOD) has been widely investigated as a chemical solution route [8, 9]. CSD of YBCO-Ba_2YTaO_6 (YBCO-BYTO) system is attractive, since among several nanocomposite systems, this one presents the highest percentage of random

nanoparticles segregated within an epitaxial superconducting matrix [3, 5]. However, at a certain BYTO concentration (≥ 10%M), nanostrain values saturate and nanoparticles tend to agglomerate producing a current blocking effect in the YBCO matrix, degrading J_c [3, 2].Therefore, it is crucial to control nanoparticle formation process in order to engineer nanoparticle size and nanostrain to improve superconducting properties. In order to control these parameters, a two-step thermal process was developed in a previous work [2]. This process allows formation of a highly epitaxial and strained YBCO matrix, with more than 90% randomly oriented nanoparticles. It has been shown that nanoparticle size is reduced from 40-60 nm, with a one step process, down to 15-20 nm, with a two steps process. This work was made for BYTO concentration of 10% M [3].

In the present work we studied the characteristics of CSD- YBCO-BYTO nanocomposite thin films with BYTO concentrations 6%, 10% and 20% M. We found similar results to the previous work. We obtained critical current densities of ~2.6 MA/cm^{2}at 77K and a smooth $J_c(H)$ dependence.

2. Experimental details

YBCO-BYTO films were made starting with a chemical solution precursor obtained by introducing stoichiometric amounts of tantalum (V) ethoxide in a TFA-YBCO solution prepared as described elsewhere [10, 11]. Molar percentages of BYTO 6%, 10% and 20% have been investigated. The metallorganic precursor solution was spin coated on crystalline substrates (100) $LaAlO_3$ (LAO) of 5x5 mm^2. Then a thermal treatment (pyrolysis) was made, in a tubular furnace, under oxygen flow, at 300°C for 30 min. Instead of a one step thermal process used in previous works [2], the crystallization process was achieved following a two steps process, in an atmosphere of N_2, with an O_2 partial pressure of 0.2 mbar, as indicated in figure 1. A water vapor partial pressure P(H_2O) of 22 mbar was added at certain stages of the process, indicated by blue boxes. It is important to note that the second heating ramp (from 575°C to 775°C) has been made under dry atmosphere, to minimize the coalescence of the already nucleated BYTO nanoparticles. After thermal process the superconducting phase was achieved by oxygen annealing at 450 °C.

The microstructure and phase analysis of the fully converted YBCO nanocomposite films were studied by two-dimensional (2D) X-ray diffraction (XRD) using a Brucker AXS GADDS diffractometer. Strain quantification (ε) was carried out based on the Williamson-Hall method [12, 13], by analyzing the symmetric (00l) 2θ Bragg diffraction integral breath, acquired in a SIEMENS D5000 diffractometer. Quantitative determination of the randomly oriented nanodots fraction (RF) was performed by 2D-XRD analysis, based on simultaneous out-of-plane measurement of both textured and random signals [2]. Both J_c and T_c were obtained by SQUID measurements.

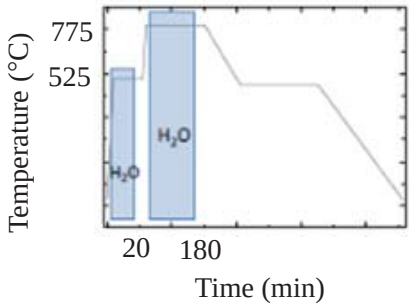

Figure 1. Schematic of the two step thermal process used to prepare YBCO nanocomposites.

3. Results and discussion

Figure 2 shows the diffractogram of YBCO films, with different molar percentages of BYTO, (6%, 10% and 20%). We observe in all films (00l) reflections, indicating c-axis orientation. The peak that appears at $2\theta \approx 43.0°$ corresponds to the (400) reflection of BYTO second phase. The addition of Ta in the precursor solution changed very little the **c** lattice parameter of YBCO ($c \approx 11.683$ Å), indicating no Ta substitution in YBCO [2]. The CuO phase is also identified in all samples.

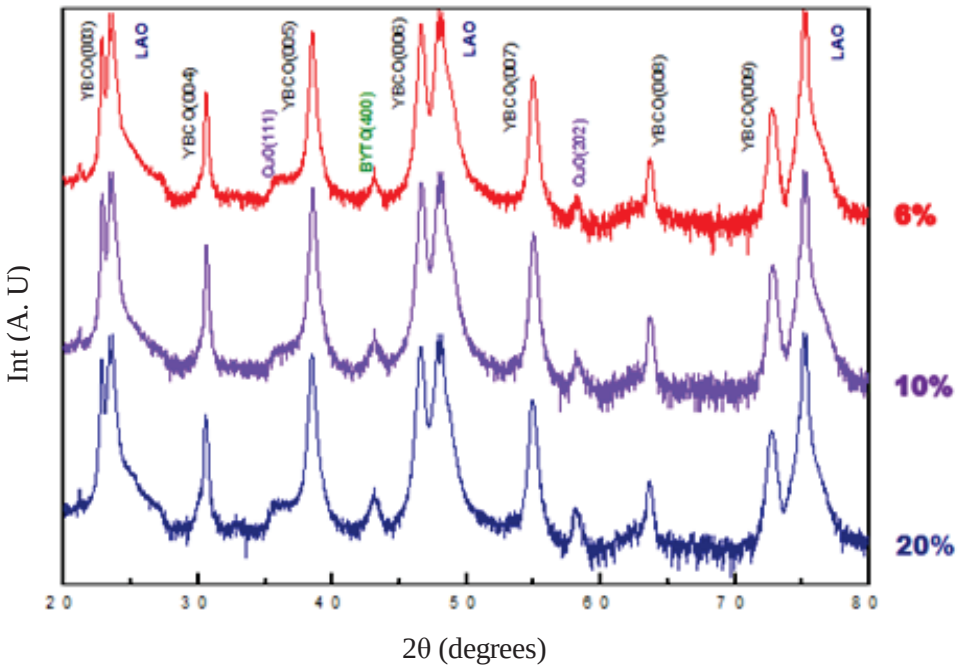

Figure 2. X-ray diffraction of YBCO-BYTO nanocomposites films with different molar percentages of BYTO (6%, 10%, and 20%).

A typical 2D-XRD pattern for YBCO with 10% mol BYTO is shown in figure 3. The (00l) diffraction spots of the YBCO phase confirm c-axis orientation, consistent with figure 2. The BYTO ring from the (220) reflection indicates that this minority phase is mostly randomly oriented, with a random fraction, RF= 95%.

The size of epitaxial BYTO nanoparticles was estimated from the Scherrer formula ($D= 0.9\lambda/\beta \cos\theta$) and ranges from 5 nm to 15 nm, as the BYTO percentage is increased. From previous TEM analysis it has been identified that epitaxial nanoparticles tend to grow at the interface with the substrate. It is important to note that the particle size of the randomly oriented nanoparticles embedded within the YBCO matrix is decreased from 60 nm to 15-20 nm when using the two step thermal process [3].

Nanostrain was quantified by means of the Williamson-Hall method [9]. In figure 4 it is observed that the two steps thermal process increases nanostrain, reaching values up to 0.30% for the 20% BYTO samples. Two steps thermal process do not degrade T_c ~ 90K, and lead to critical current densities up to 2.6 MA/cm^2 at 77K, as shown in table 1. The YBCO-20% BYTO nanocomposite show lower J_c values, compared to 6% and 10% nanocomposites, although the nanostrain values are very high. It is likely that despite decreasing nanoparticle size, such high concentration of nanoparticles start to be an obstacle to current flow percolation [9].

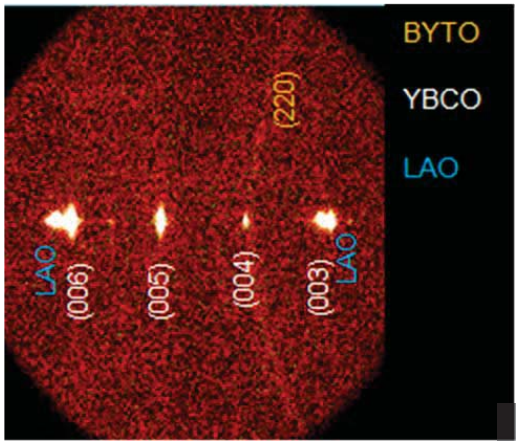

Figure 3. (a) Two-dimensional X-ray diffraction pattern for the YBCO-10% BYTO film.

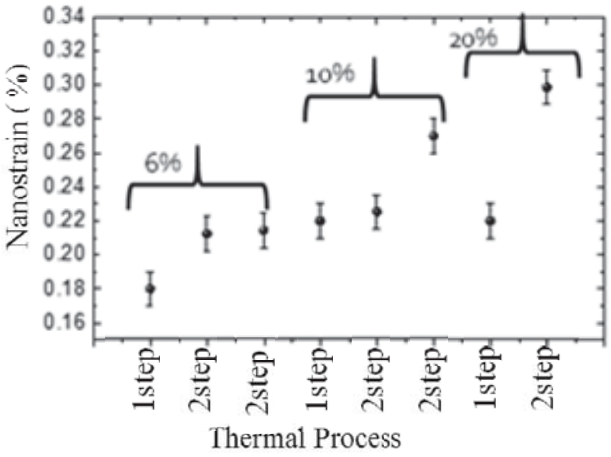

Figure 4. Nanostrain versus thermal process for YBCO nanocomposites with different percentages of BYTO. The first point in each set of data is presented for comparison and corresponds to samples prepared in a previous work with a one step thermal process [2].

Table 1. Physical properties of YBCO nanocomposite films, with different molar percentages of BYTO.

%BYTO	T_c (K)	ΔT_c(K)	J_c (77K) MA/cm^2	Particle Size of epitaxial NP (nm)
6	89	2.8	2.4	5.6
10	89	4.4	2.6	10.5
20	89	3.6	1.9	14.3

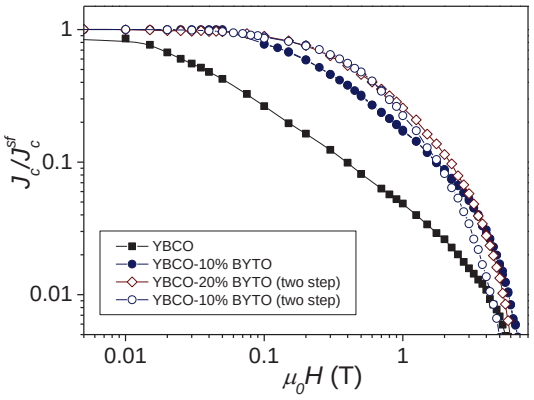

Figure 5. Magnetic field dependence of the critical current density a 77K and H ∥ c, normalized to its self-field value, for: standard YBCO sample, YBCO-10% BYTO nanocomposite made with the standard one step thermal process and two YBCO-BYTO nanocomposites (10%BYTO and 20%BYTO) made with the two step thermal process.

The pinning strength has been evaluated from the J_c (H) behavior at 77K and H ∥ c in a log-log plot, normalized at self-field, as shown in figure 5. At low fields, there is a plateau associated with the single vortex pinning regime, where vortex-vortex interactions are negligible and vortices are pinned individually. At high fields, collective effects become important and J_c starts to decrease with magnetic field [3]. All the YBCO-BYTO nanocomposites show a smooth dependence with magnetic field, compared to pristine YBCO films. Also, the two step processed YBCO-BYTO films show a smoother field dependence, compared to the single-step processed YBCO-BYTO films.

4. Conclusions

The use of a two-step heating profile to prepare YBCO nanocomposites with 6%, 10% and 20% BYTO is an effective approach to decrease nanoparticle size, increase nanostrain and improve the pinning performance while maintaining highly epitaxial YBCO films. These results demonstrate the potential for an industrially scalable chemical solution deposition approach to prepare YBCO nanocomposites.

5. References

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