NEW MICROCRACK NETWORK GENERATION IN TSMTG YBCO

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Abstract. Melt Textured YBa₂Cu₃O₇ / Y₂BaCuO₅ (Y123/Y211) and Y123/Y211/CeO₂ composites, have been grown by Top Seeding Melt Texturing growth (TSMTG). The initial powder mixture has been densified, through a pre-melting process carried out at 1000-1050°C under O₂ atmosphere. This process leads to a strongly decreased porosity of the pellet. Afterwards, TSMTG is carried out in air atmosphere. The influence of the pre-melting process on the final microstructure has been analyzed by optical and scanning electron microscopy (SEM). Microstructural analysis reveals a high microcracking density. Three well defined microcrack orientations have been detected, one parallel the (001) planes and a new class of microcrack parallel the {100} planes. The latter is being produced by the pre-melting densification process, while the usual microcrack parallel to (001) planes also prevail. The influence of the oxygenation process and the Ce additives on the development of this new class of microcrack pattern will be discussed. Finally, the influence of the new microcrack network on the critical current density will be presented.

1. Introduction

In YBCO (TSMTG) samples due to their ceramic character and growth method present a high diversity of defects [1-2]. The mechanical properties of YBCO are poor, and they easily cleave along the ab-plane cracks. Different processes have been tried to improve these properties for trapped field applications. For example, macrocracks could be avoided by the addition of submicrometric secondary phases like Y211 particles. Melt textured samples have a high density of microcracks parallel to the ab-plane, with a mean distance characteristic of several μm. These ab-plane cracks form due to large stresses induced during the tetragonal (T) to orthorhombic (O) phase transformation [3]. These microcracks grow around the Y211 particles, as a result of different thermal expansion coefficients of the matrix and the particles during the cooling process [4-5]. Microcracks do not propagate across the Y211 particles thus the mechanical properties of the composites improve by Y211 addition [1]. Melt textured growth (TSMTG) YBa₂Cu₃O₇₋δ / YBaCuO₃ composites having different microcrack orientations are very interesting materials because they offer new opportunities to understand the complex properties of bulk superconductors. We report on new microstructural features such including a new class of micro/macromacraks parallel to the {100} planes being promoted by a pre-melting densification process under O₂ atmosphere prior to the slow cooling crystallization process. We present the development of this new class of micro/macromacraks pattern and the influence of the
oxygenation process, initial concentration of Y211 particles and additions of a secondary phase like CeO₂.

2. Experimental
Melt texturing of YBCO has been carried out by Top Seeding Growth (TSMTG). Two starting compositions of YBCO/Y211 with 75%"Y123+25%"Y211 and (Y123+30%"Y211)+1%"CeO₂ using NdBCO seeds [6] have been chosen. All the explored samples have been prepared using an innovative pre-melting densification process developed at Argonne National Laboratory [7]. This process consisted of heating the pellets in an isothermal furnace in a temperature range of 1000-1050ºC in O₂ atmosphere followed by a fast cooling process. Afterwards the typical growth process consisted of heating the samples in an isothermal furnace up to a pre-sintering temperature of ~960ºC for 24h in air. Subsequently, the temperature was risen ~30ºC above the peritectic temperature (Tp~1010ºC) for 5h, rapidly cooled down until a temperature T₁, characteristic of the heterogeneous nucleation at the seed, and slowly cooled down at 0.5ºC/h within a temperature window [T₁,T₂]~15ºC, where T₁ is a characteristic temperature for homogeneous nucleation without seed. All samples were single domain and grew up to the border of the pellet and the top surface showed the typical cross shape of the ab crystallographic directions. The described phenomenon is characteristic of all the pellet, though for comparison purposes all micrographs shown belong to similar position in the sample. Small pieces few millimeters from the seed were afterwards cut to perform the superconducting characterisation. The effect on the growth process and microstructure was studied by polarized optical microscopy and scanning electron microscopy (SEM-EDX) with prior chemical etching. The (001) planes and cross sections parallel to the [001] axis were analyzed. The superconducting properties were studied by measurements of SQUID magnetometry, Jc(T) temperature dependence of the inductive critical current density for H//c and Jc(H) magnetic field dependence of the inductive critical current density measured at 77K.

3. Results and discussion
The analysis of the high quality single domains obtained showed two main morphological effects. First, the spectacular porosity reduction; only some punctual small remaining porosity was localized in the c-Growth Sector. Second, a new high macro/microcracking density network appeared with three well defined macro/micro orientations. The well known microcracks, parallel to the (001) plane (ab-microcracks) are observed on the surfaces parallel to [001] axis [8] (see figure 1b and 1a) and in addition a new class of macro/microcrack planes parallel to the {100} planes (c-microcracks), generating a perpendicular network. The three families of macro/microcraks planes formed a quasy-pseudocubic network in three dimensions (figure 1a and 2a). Diko and Krabbes [9] refereed to three

![Figure 1. SEM micrographs showing ac-planes of chemically etched MTG YBCO composites. (a) Sample with 25%wt. of Y211 grown with pre-melting densification process. This sample is characterized by extremely regular cracks parallel and perpendicular to the ab-planes. (b) Sample with 30% Y211 and 1%CeO₂ in wt. grown without pre-melting densification process. Only the typical ab-microcracks, parallel the (001) plane are observed.](image-url)
types of cracks, the typical ab-microcracks, ab-macrocracks (macroscopic stresses due to inhomogeneous Y211 particle distribution [8]) and c-macrocracks, which are more or less parallel to the c-direction contained within the ac-plane. The macro/microcracks families presented here are, in addition, oriented parallel to the $\{100\}$ planes (c-microcracks). It is important to notice the correlation of this new macro/microcrack network with the pre-melting densification process. The new family of macro/microcracks, (c-microcracks) is generated only in YBCO composites with a previous pre-melting densification process which initial composition did not contain CeO$_2$ addition. Samples without this pre-melting process, with CeO$_2$ did not show the new macro/microcrack network see figure 1b and 2b. In MTG YBCO composites with a low content of Y211 particles and no CeO$_2$ addition, the distribution of the Y211 phase is not uniform in size ($\leq 10\mu m$) and is not homogeneously distributed in the matrix independently of the densification process used, see figure 2a and 2c. This composition is known to promote macrocracking due to macroscopic stresses [8]. The large Y211 particles are not effective crack stoppers and are ineffective vortex pinning centers[10]. CeO$_2$ addition is used to increase the liquid viscosity avoiding coarsening of the Y211 particles [11] and consequently the distribution of the Y211 particles is more homogeneous in distribution and size ($\leq 1\mu m$). Analysis of the samples before and after oxygenation demonstrated that the new crack pattern is generated during the oxygenation process in samples with pre-melting densification and without Ce addition see figure 2c versus 2a or 1a. In figure 3, $J_c^\ast (T)$ values demonstrate the high quality of the non-porous single domains even with the presence of the macro/microcracks network. The new family of macro/microcracks perpendiculars to the ab-planes do not strongly decrease the $J_c$ critical current
values since cracks do not cross completely the full sample and hence currents percolate because they are not continuous along the c-axis. Microstructural analysis of surfaces cut parallel to the [001] axis reveals the non-continuous character of the new family of cracks parallel to the \{100\} planes across the matrix, see figure 1a. The inset of figure 3a shows the normalized $J_c(T)$ dependence where it is revealed that only samples with CeO$_2$ addition and thus Y211 particles size <1μm, with or without the pre-melting densification process, show a strong contribution of correlated vortex pinning disorder induced by the Y211/YBCO interfaces [12]. This is reflected by a lower $J_c(T)$ dependence (in terms of a “bump”) above 40 K. Finally in figure 3b, the $J_c(H)$ dependence of pre-densified samples with and without the new microcrack network may indicate important changes in the kinetics of the oxygenation process. Samples with the pre-melting densification process (i.e. with almost no porosity); samples without the new microcrack network show a strong decrease of the irreversibility line probably induced by insufficient oxygenation. We conclude that the vortex defect structure of the single domains with the new pre-melting densification process is a complex issue where an interplay between the low porosity, the microcrack network and associated stress relaxation should occur.

4. Conclusions
An innovative pre-melting densification process has been developed that shows two main effects: a spectacular porosity reduction and a high macro/microcrack density network in YBCO samples without Ce additions. The microstructural analysis reveals three well defined macro/microcrack orientations forming a quasy-pseudocubic network with cracks parallel to the (001) plane (ab-microcracks) and two classes of macro/microcrack planes parallel to the \{100\} planes (c-microcracks). The new family of macro/microcracks, parallel to the \{100\} planes (c-microcracks) are generated only in YBCO densified composites without CeO$_2$ addition and thus large Y211 particles and they are formed during the oxygenation process. The macro/microcracks perpendicular to the ab-planes do not strongly damage the $J_c$ critical current values, probably because they extend for only short distances.

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