Proceedings of the 16th Czech and Slovak Conference on Magnetism, Košice, Slovakia, June 13–17, 2016

# Trapped Field of YBCO Bulk Superconductors Prepared by Infiltration Growth Process

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Single grain  $Y_1Ba_2Cu_3O_{7-x}$  (YBCO) bulk superconductors were prepared from different starting precursors. In the first case,  $Y_2Ba_1Cu_1O_5$  (Y211) powder as the solid phase and the liquid phase of mixture of  $YBa_2Cu_3O_{7-x}+BaCuO_2+CuO$  powders were used. In the second case, the solid phase was prepared from the mixture of  $Y_2O_3+BaCuO_2$  instead of the conventionally used Y211, and as the liquid phase a mixture of  $Y_2O_3+BaCuO_2+CuO$  powders was used. The trapped field measurements at 77 K were performed and microstructure of samples was studied by polarized light microscopy. The positive influence of  $Y_2O_3+BaCuO_2$  starting precursor on the oxygenation, Y211 particle refinement and measured trapped field is shown.

DOI: 10.12693/APhysPolA.131.1048

PACS/topics: 74.72.-h, 74.81.Bd, 61.50.-f, 74.25.Ha, 81.10.Fq

#### 1. Introduction

YBCO bulk superconductors prepared by top-seeded infiltration growth (TSIG) method are perspective materials for practical applications. The main reason for using this method is relatively small volume change of resultant product.

Trapped magnetic field of YBCO superconductors is strongly dependent on their microstructure obtained in the process of single grain production. Desirable microstructure of YBCO superconductors should consist of  $Y_1Ba_2Cu_3O_{7-x}$  (Y123) matrix with small homogeneously distributed Y211 particles serving as pinning centres. Addition of CeO<sub>2</sub> or BaCeO<sub>3</sub> is a common way how to refine Y211 particles [1, 2].

Yang et al. [3] published a paper where they proposed a new method for production of GdBCO bulk superconductors by TSIG technique with small 211 particles without Ce addition. They replaced Gd-211 precursor with a mixture of  $Gd_2O_3$  and  $BaCuO_2$ .

In this work, we compared properties of the sample prepared by classic TSIG method with Y211 as a precursor and CeO<sub>2</sub> addition with the sample prepared by new TSIG method, where Y211 is replaced with mixture of  $Y_2O_3$  and BaCuO<sub>2</sub> powders and CeO<sub>2</sub> is not added. The preparation of Y211 pellets by this new method should improve properties of YBCO single crystals refining the size of Y211 particles trapped during peritectic solidification and subsequently to enhance trapped magnetic field of these bulk superconductors.

## 2. Experimental materials and methods

Two kinds of samples were prepared by TSIG method. In the first case, the sample was prepared by known method [4, 5], where as an initial material of solid phase Y211 powder (Solvay Fluor GmbH, Germany, maximum particle size  $d_{\text{max}} = 5 \ \mu\text{m}$ ) with 0.5 wt% addition of CeO<sub>2</sub> (Aure Remy) was used. Commercially available powders of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub> and Ba<sub>3</sub>Cu<sub>5</sub>O<sub>8</sub> (035, i.e. a mixture of BaCuO<sub>2</sub> and CuO powders in molar ratio of BaCuO<sub>2</sub>:CuO = 3:2) were weighted in molar ratio of Y123:035 = 1:1 and used as liquid source.

In the second case, Y211 phase as a solid state precursor was produced from the mixture of  $Y_2O_3 + xBaCuO_2$  powders, where x = 1.2 at.%, and as a liquid phase the mixture of  $Y_2O_3$  : BaCuO<sub>2</sub> : CuO = 1 : 10 : 6 powders was used. After pressing into pellet with diameter 20 mm, the mixture was annealed at 910 °C for 20 h to form Y211 by reaction:  $Y_2O_3 + xBaCuO_2 = Y211$ .

The solid phase for both methods was prepared in the form of 12 g batches uniaxially pressed into pellets with diameter 20 mm. The liquid phase pellets with diameter 32 mm were pressed from batches of 20 g in sample A and 16.5 g in sample B.

The solid phase pellet was placed on the top surface of the liquid source pellet, which, in turn, was placed on the Yb<sub>2</sub>O<sub>3</sub> plate positioned on MgO single crystal plate. On the top of the samples Nd<sub>1</sub>Ba<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub> (Nd123) thin film seeds were placed. The entire arrangement was supported by aluminium oxide plate (Fig. 1a,b).

Both samples (sample A and sample B) for infiltration growth process were heat treated in the same way. First, the samples were heated to  $T_1 = 910$  °C, and stabilized for 20 h. Then the temperature was increased to the melting point T = 1045 °C and kept constant for 1 h to ensure infiltration of liquid phase to solid phase pellet. Cooling process consisted of several steps. After infiltration samples were cooled to temperature  $T_p = 1020$  °C with the rate of 60 °C/h. From 1020 °C to 990 °C they were slowly cooled in three steps for 180 h (rate 0.3, 0.2, 0.1 °C/h) and then samples were furnace cooled. The time temperature schedule for both samples is presented in Fig. 1c. Prepared bulk crystals were oxygenated in the tubular furnace with oxygen flow atmosphere at temperature 410 °C for 300 h.

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Fig. 1. Schematic illustration of pellet layers of the TSIG process with different starting precursors: (a) sample A — method with Y211 solid-phase precursor, (b) sample B — Y211 formed from  $Y_2O_3$  and  $BaCuO_2$ , (c) heat treatment profile used for growth of Y123 bulk single crystal by infiltration growth process.

Microstructure of the samples was studied after polishing by polarised light microscopy and the size of Y211 particles was measured by image processing.

Trapped magnetic field was measured by a home made Hall probe scanner. The samples were cooled in liquid nitrogen and magnetized in an electromagnet with B = 1.4 T applied parallel to their *c*-axis for 5 min. The distance between sample and pole cone at measurement was less than 1 mm.

# 3. Results and discussion

# 3.1. Results of single crystal growth

In Fig. 2 the macroscopic images of the top surfaces of samples A and B are demonstrated. The fourfold growth facet lines starting from the seed are clearly visible on the top surface of both samples indicating that the crystal growth was well controlled.



Fig. 2. The top surface of the single-domain YBCO bulk superconductors ( $\emptyset$ 20 mm) with different starting precursors prepared by the TSIG technique.

It is well known that  $CeO_2$  addition to samples increases the growth rate of Y123/Re123 single crystals [6, 7]. By comparing our samples treated with the same time-temperature regime, it can be seen that the single crystal in sample B without  $CeO_2$  addition is bigger than that in the sample A with  $CeO_2$  addition (Fig. 2). We assume that the main reason is in the difference of Y211 particles size in the samples at peritectic reaction. In the sample A, Y211 powder with average particle size 5  $\mu$ m was used. We suppose that Y211 particles were created by reaction (1) in the sample B and

they are smaller than particles in sample A. As the result, the crystal growth in sample B was more intense just due to bigger reaction surface between Y211 and liquid at peritectic reaction

$$Y_2O_3 + BaCuO_2 \rightarrow 910 \,^{\circ}C, 20 h \rightarrow Y_2Ba_1Cu_1O_5.$$
 (1)

## 3.2. Analysis of trapped field

Figure 3 shows the trapped field profiles of samples A and B of diameters 20 mm. We can see that sample A has a much lower magnetic trapped field than sample B. In spite of the very low trapped field  $B_{\text{max}} = 0.013$  T, the sample exhibits superconductivity. The lower value of  $B_{\text{max}}$  could be caused by worse oxygenation. This has also been confirmed by microstructure analysis, as discussed later (Fig. 4).



Fig. 3. Trapped magnetic field profiles for the top surface of samples A and B.

The sample B exhibits typical single-peak trapped field profile on the main part of the sample. Another small peak corresponds to parasitic crystal. The maximum measured trapped field for sample B was  $B_{\rm max} = 0.169$  T.

### 3.3. Structure observation of studied samples

The macro structures of samples A and B after oxygenation process on the sections parallel to (100) plane are presented in Fig. 4 (left). In both cases the boundaries between a- and c-growth fronts (growth sector boundaries GSB [8]) can be seen. The macrostructure of samples is quite different. The structure of sample A is typical for infiltration growth process with small portion of pores (about 13 vol.%), which are elongated in horizontal direction. The smaller portion of pores in sample A caused deficient oxygenation. Tetragonal nonsuperconducting Y123 phase visible as horizontal dark areas (Fig. 4, sample A) is present in a high portion.



Fig. 4. Cross-sections of samples A and B in polarised light (left). Y211 particles and twins in the sample A and B (right). Polarised light micrographs taken in the a-growth sector 2 mm from the seed.

The porosity of sample B is higher and reaches 22 vol.%. This higher porosity is not typical for samples prepared by infiltration process, but rather for classic top seeded melt growth process (TSMG). In the case of TSMG, it is supposed that the pores are formed from  $O_2$  as a result of peritectic reaction at sample melting

2YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6</sub> $\leftrightarrow$ Y<sub>2</sub>BaCuO<sub>5</sub>+Ba<sub>3</sub>Cu<sub>5</sub>O<sub>5.5</sub>+0.75O<sub>2</sub>.(2) Probably, the similar effect was realised in the case of sample B. In the preparation process of Y211 precursor, the Y123 phase could be formed due to excess of BaCuO<sub>2</sub>, and subsequently at melting by peritectic reaction, O<sub>2</sub> pores appear. The pores are helpful in the process of oxygenation of bulk crystal, as it is seen in Fig. 4 (left, sample B). Only a very small part of the sample in the upper right corner is not oxygenized (Fig. 4, left).

In Fig. 4 (right), the microstructure of Y123 bulk crystal with  $CeO_2$  addition (sample A) and the microstructure of Y123 bulk crystal produced by new method without  $CeO_2$  addition (sample B) are shown. The particle sizes were evaluated by image processing software. The fine and spherical Y211 particles are uniformly distributed in the Y123 matrix in both samples. The mean size of most particles in sample A is in the range of 1.5 to



Fig. 5. The size distribution of Y211 particles in the samples A and B.

5  $\mu$ m. There are several large particles in sample B, but mean size of the most particles is in the range of 0.5 to 2  $\mu$ m. The size distribution of the Y211 particles in samples is presented in Fig. 5. The smaller Y211 particles in Y123 matrix are more effective in pinning of magnetic flux lines, which positively influences amount of trapped magnetic field.

# 4. Conclusions

In this work, two different methods for Y123 single crystal production by TSIG process were tested. One of them was classic method with Y211 solid phase pellet, in the second method the solid phase was formed from the mixture of  $Y_2O_3$  and BaCuO<sub>2</sub>. We have found that the second method produces YBCO bulks with higher porosity, which significantly improves oxygenation of the sample. Besides that, the second method causes refinement of Y211 particles without addition of cerium oxide. Higher trapped magnetic field measured for the sample prepared by second method was caused by by its much better oxygenation.

#### Acknowledgments

This work was realized within the framework of the projects: ITMS 26220120035, ITMS 26220220061, ITMS 26220220041, APVV No. 0330-12, VEGA No. 2/0121/16 and Stefanik Project SK-FR-2013-0025, SAS Centre of Excellence: CFNT MVEP.

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