

Melting of $\text{SmBa}_2\text{Cu}_3\text{O}_{7-y}$ -seeds during preparation of YBCO Bulk Superconductors by Infiltration Growth Method

L Vojtkova, P Diko and D Volochová

Institute of Experimental Physics, Slovak Academy of Sciences, Watsonova 47,
04001 Košice, Slovakia

E-mail: vojtkova@saske.sk , dikos@saske.sk , volochova@saske.sk

Abstract. The conditions for single-grain growth of YBCO bulk superconductors by top seeded infiltration growth were tested. It is shown that the interaction of melt formed from $\text{BaCuO}_2 + \text{CuO} + \text{Y}_2\text{O}_3$ precursor with the Sm123 seed causes dissolution of the seed at maximum melting temperature 1045 °C. Experiments with low weight Y211 pellet confirmed that the low concentration of Y in the infiltration melt is responsible for this effect. The most effective way suppressing the seed dissolution was shown to be the insertion of Y123 + Y211 buffer layer between the seed and the Y211 pellet. This buffer layer possesses the melt which is saturated with yttrium what prevents dissolution of the seed.

1. Introduction

Single grain YBCO bulk superconductors are perspective and extensive studied materials for practical applications. There are several ways how to prepare YBCO bulk superconductors [1][2][3][4]. Top seeded infiltration growth method gives good assumptions for production of bulk single crystals with fine and uniformly distributed Y211 particles in Y123 matrix, low porosity and with big dimensions and also with complicated shapes [5]. Some authors [6-10] describe the various ways to prepare YBCO bulk single crystal by infiltration method. In general process consists of heating to melting temperature, infiltration of liquid source to Y211 pellet, cooling to peritectic transformation, slow cooling through interval of peritectic transformation and furnace cooling to room temperature. Temperature and time of individual steps as well as mass of particular components are very important for obtaining resultant bulk single crystal without parasitic ones. Problems associated with transport of atoms between liquid phase and the seed are described by Jee [11]. Some authors solve various problems connected with diffusion between the seed and liquid phase using buffer layer at crystal growth by TSMG process [12-17] and TSiG process [18].

In this work influence of maximum infiltration temperature and configuration of individual layers of experiment were studied.

2. Experimental materials and methods

2.1. Experimental material

As initial material $\text{Y}_2\text{Ba}_1\text{Cu}_1\text{O}_5$ (Y211) powder (Solvay Fluor GmbH, Germany) with a maximum particle size $d_{\text{max.}} = 5 \mu\text{m}$ was used. The 12 g of the powder was compacted by uni-axial pressing to Y211 pellet with diameter 20 mm. Liquid source pellet (LSP) was prepared as a mix of Y_2O_3 (Heraeus



Feinchemikalien und Forschungsbedarf GmbH), CuO (Huizhoutian Yi Rare Material co., Ltd) and BaCuO₂ (prepared by calcination) powders in ratio 1:6:10 respectively in accordance to reaction 1.



Powders were mixed in grinding mill with ZrO₂ bowl for 20 minutes. Mixed powders were uniaxially pressed into 20 gram pellet with diameter 30 mm. The diameter of liquid source is bigger than Y211 pellet for ensuring of complete infiltration and better support for final YBCO sample.

2.2. Growth experiments

Growth experiments of samples were inspired by work of Guo-Zheng Li and others [10]. The samples were heated to melting temperature $T_m = 1045^\circ\text{C}$, heating rate 100°C/h , holding time at T_m 3 hours. Then the samples were cooled to a temperature $T_p = 998^\circ\text{C}$ by rate 60°C/h and from 998°C to 982°C were slowly cooled for 64 hours (rate 0.25°C/h).

The time-temperature regime is presented in Figure 1. In order to find optimal regime of YBCO single crystal growth from the seed, temperatures of solidification growth (T_p and T_o from Figure 1) were varied approximately 5°C up and down.

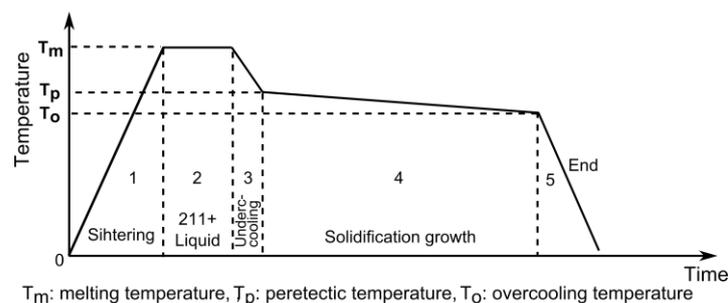


Figure 1. Time-temperature regime of single crystal Y123 production by infiltration growth method

Macrostructure observation and microstructure analysis were performed on polished specimens of the processed samples using an optical microscope equipped with a polarizer. EDX analysis was also performed.

3. Results and discussion

Some experiments were performed with samples composed of layers with arrangement as shown in Figure 2. Bottom layer was formed of MgO single crystal. On the MgO single crystal Yb₂O₃ pellet was placed. These bottom layers was used in order to avoiding reaction of liquid source pellet with Al₂O₃ base. The upper part consisted of Y211 pellet and Sm123 single crystal, prepared by TSMG process, as a seed placed on that.

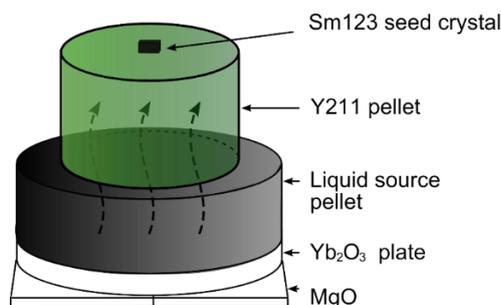


Figure 2. Schematic arrangement of individual layers of YBCO single crystal production by infiltration growth method

We have found that the seed melting occurs at heating to melting temperature $T_m = 1045^\circ\text{C}$. In Figure 3 microstructures of three samples sectioned along the seed in the c-axis direction are presented. All three samples were identically heat treated, but seeds with different height were used. In Figure 3a the microstructure of sample with biggest seed is shown. It can be seen that lower part of seed was melted and subsequently crystallized at cooling as polycrystalline material. The upper part of seed remained unchanged in single crystal form. On the other hand in Figure 3c the sample with smallest seed is presented. In this case the seed was completely melted and mixed with other parts of the sample. The middle sample (Figure 3b) was treated by using the seed with intermediate height. The whole seed was melted and crystallized in polycrystalline form at cooling.

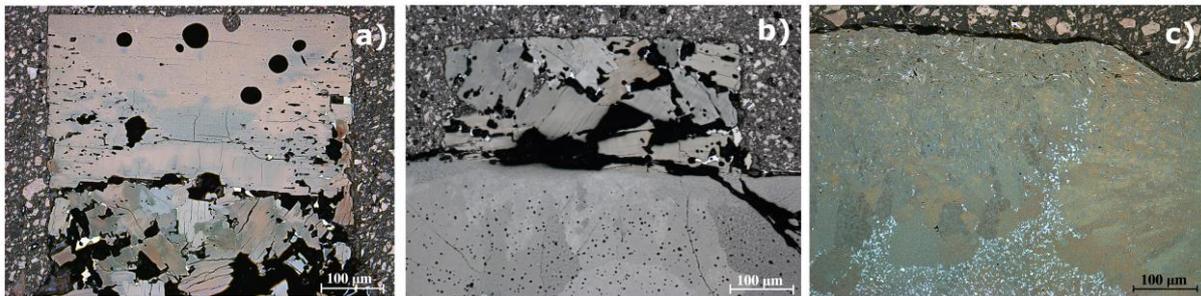


Figure 3. Microscopic analysis of samples sectioned through seed in axis c.

It is reasonable to suppose that the reason for the seed melting is diffusion of Sm from the seed to the sample melt which starts at the seed/sample interface. This process leads to decomposition of Sm123 phase according to reaction (S-solid, L-liquid, G-gas):



The kinetic of this reaction is controlling destruction of the seed. It is faster at higher temperatures due to faster Sm diffusion in the sample melt as well as due to higher solubility of Sm in the melt at higher temperatures [19]. The seed is in fact composite of Sm123 crystal and Sm211 particles, therefore it keeps its original shape even if the Sm123 phase is decomposed according to reaction (2). Then, during cooling the self nucleation of Sm123 crystals leads to polycrystalline form (Figure 3).

In order to avoid the seed dissolution we had to decrease maximum temperature of infiltration process below 1045°C . An effective maximum temperature of infiltration process was determined to be 1025°C where the single-grain YBCO sample grows from the seed (Figure 4).

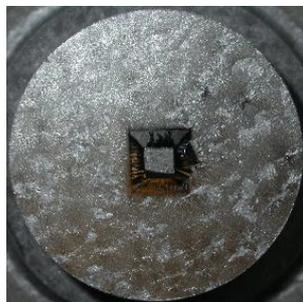


Figure 4. Sample prepared with $T_{\text{max}} = 1025^\circ\text{C}$

Higher melting temperature of the process influences infiltration of the melt to Y211 pellet and homogeneity of the melt [20, 21]. If melting temperature of the melt of infiltration process is low, some clusters with structure similar to solid form of Y123 can exist in the melt. These clusters can serve as

seeds of Y123 crystals at cooling. If infiltration temperature is higher there are less clusters and clusters have smaller dimensions. Thus, for the growth of single crystal without parasitic crystals, higher infiltration temperature should be used. However, this leads to dissolution of Sm123 type seeds. One possibility how to eliminate degradation of the seed is to saturate RE concentration in the melt. The solubility limit of Sm in the sample melt is about 2 wt. % [19]. This possibility was tested with the small Y211 sample, as presented in Figure 5, (diameter 8 mm, weight 0,5g). The weight of the seed was enough to saturate RE concentration (Sm + Y) in the sample melt by partial dissolution of it in the melt. The sample was identically heat treated with the same holding time at maximum melting temperature 1045°C of the process. As a result of using small sample the perfect Y123 single crystal was grown from the seed.

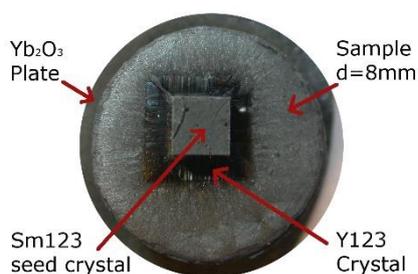


Figure 5. The change of ratio of seed size versus sample size in favour of seed

Conducted experiments and their results point on very low concentration of Y in the infiltrated melt which is obviously caused by slow dissolution of Y211 particles in the melt at the infiltration process. This is in contrast with the melt which is formed from Y123 phase at TSMG process. During peritectic decomposition of Y123 phase according to reaction (S-solid, L-liquid, G-gas):



the melt L is saturated with Y and the Sm123 type seed is not destructed by reaction (3). From this consideration an idea comes to put an interlayer containing Y123 phase between the Sm123 seed and the Y211 pellet. This arrangement should provide Y reach melt near the seed and to prevent its melting.

For this experiment the same powders and pellets (Y211 and liquid source pellet) as in previous experiment were used. The main difference was in using of buffer layer between Sm123 seed and Y211 pellet. Buffer layer was prepared as a mixture of Y123 and Y211 (weight ratio 7:3) powders with addition of 1 wt. %, CeO₂ [22]. The buffer layer powders were also mixed in grinding mill and uniaxially pressed into the 0.15 gram pellet with diameter 5 mm. The arrangement of individual layers is presented in Figure 6 and consists of MgO single crystal, Yb₂O₃ plate, liquid source pellet, Y211 pellet, buffer layer (composed of Y123 and Y211) and Sm123 seed. All were placed on Al₂O₃ base.

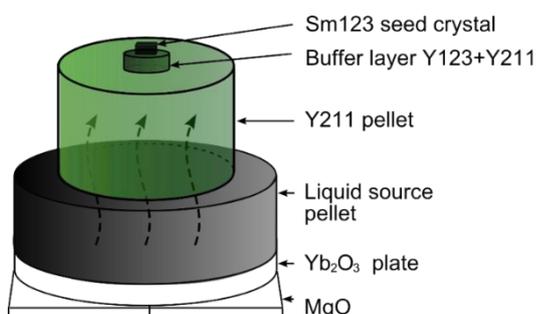


Figure 6. Schematic arrangement of individual layers of YBCO single crystal production by infiltration growth method with buffer layer

In Figure 7 the sample from experiment with used buffer layer is presented. Maximum temperature of infiltration process was 1045°C. Seed remained undissolved and grown single crystal can be seen (Figure 7a). In Figure 7b microstructure of the sample sectioned through seed and buffer layer along the a/c-plane is presented. The growth sector boundary (GSB) of Y123 single crystal progressing from seed through buffer layer to Y211 pellet can be seen.

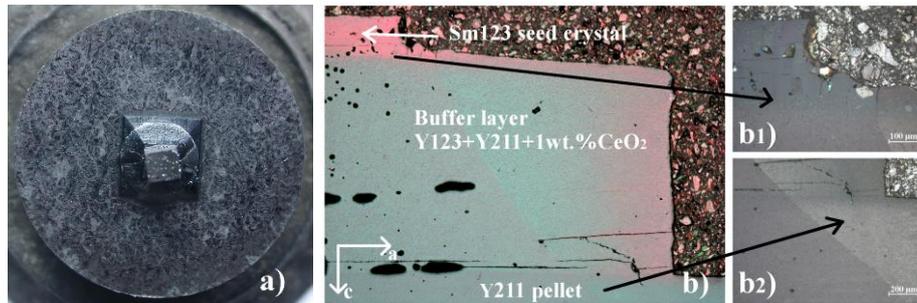


Figure 7. Sample prepared using buffer layer at $T_{\max} = 1045^{\circ}\text{C}$, a) top view of the sample, b) part of the sample cut along the a/c-plane, b₁) detail of the GSB in the buffer layer, b₂) detail of the GSB in the buffer layer and in the Y123 single crystal.

4. Conclusions

The dissolution of the Sm123 type seed at 1045 °C melting temperature was observed at top seeded infiltration growth of YBCO bulk single-grain samples. We found three possibilities how to protect the seed. The first is to lower the melting temperature. The maximum melting temperature of infiltration process was determined to 1025°C. The second possibility is to decrease the weight of the Y211 pellet. The experiments with smaller Y211 bulk pellet showed that the main factor of dissolution of the seed is saturation of the melt with the rare earth element. If the Y211 pellet of 0.5 gram in weight was used, the single-grain sample has been grown from the Sm123 seed also when maximum melting temperature 1045 °C was used. The insertion of Y123+Y211 buffer layer between the seed and Y211 pellet has been shown to be the third solution of the problem. The buffer layer possesses the melt which is saturated with yttrium what prevents dissolution of Sm from the seed in this melt even at melting temperature 1045 °C.

Acknowledgements

This work was realized within the framework of the projects: ITMS 26220120019, ITMS 26220120035, ITMS 26220220061, ITMS 26220220041, SAS Centre of Excellence: CFNT MVEP, PhysNet ITMS 26110230097, NANOKOP ITMS 26110230061, APVV No. 0330-12, VEGA No. 2/0121/16 and Stefanik Project SK-FR-2013-0025.

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