Preparation of $Y_x Yb_{1-x} Ba_2 Cu_3 O_{7.5}$ bulks for superconducting joint between REBCO-coated conductors

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Abstract. In this study we prepared $Y_x Yb_{1-x} Ba_2 Cu_3 O_{7.5}$ ($x = 0.1, 0.2, 0.3, 0.4, 0.5$) powders and bulks to use as intermediate for superconducting joint between REBCO-coated conductors. Superconducting joints are essential in the development of high-field REBCO persistent magnets. All the powders and bulks were sintered at 850 °C for 10 hours in air. Bulks were prepared by pressing at 70 MPa. Calcination of the bulks was performed under the same conditions as the powders. Two joint samples between GdBa$_2$Cu$_3$O$_{7.5}$-coated conductors were prepared by using the bulk samples. For $x = 0.1$, a superconducting joint was successfully obtained.

1. Introduction

REBa$_2$Cu$_3$O$_{7.5}$ (REBCO or RE123)-coated conductors are promising because of their high $I_c$ properties in high magnetic fields. Kimura et al. developed REBCO-coated conductors that exhibit high critical current in a high field ($I_{c, \text{min}} > 50$ A/cm$^2$ in 3T) [1]. For magnet application, superconducting joints are essential to extend the length of REBCO-coated conductors. Park et al. reported a joining method by diffusion. The complicated joining process includes fabrication of 20 μm micro holes, chemical etching of Ag stabilizer on GdBa$_2$Cu$_3$O$_{7.5}$ (Gd123)-coated conductor, heat treatment of joint in vacuum and annealing in oxygen for 350 hours [2].

Previously, Jin et al. demonstrated a new joining method called crystalline joint by a melted bulk (CJMB) [3]. A superconducting joint was developed between two Gd123-coated conductors using a YBa$_2$Cu$_3$O$_{7.5}$ (Y123) bulk. The core concept of this method is (a) to induce incongruent melting in the low melting temperature Y123 bulk while keeping the high melting temperature Gd123 in the coated conductors incongruent during heat treatment of the joint, and (b) allowing Y123 to melt and regrow into crystal during the cooling process. Therefore, the criteria of a good bulk intermediate would be low melting temperature and crystal growth. In 2019, Zhang et al. adopted this method and as a first step, prepared ErBa$_2$Cu$_3$O$_{7.5}$ (Er123) powder to use as intermediate. The feasible joint temperature is estimated to be in the range of 965–1013 °C after doping with Ag [4].

YbBa$_2$Cu$_3$O$_{7.5}$ (Yb123) has a low melting temperature among RE123 superconductors, making it ideal for use as a bulk intermediate. However, there is a known difficulty of inducing crystal growth in Yb123 crystals. On the other hand, it is possible to grow Y123 crystals via various methods, yet the melting temperature of Y123 (1000 °C) is higher than that of Yb123 (930 °C) in air. High heat treatment temperature accelerates the degradation of REBCO-coated conductors and as a result, the
critical current of the joint is decreased. Hence, it is preferable to use RE123 bulk with the lowest possible melting temperature.

As a solution, we have synthesized $Y_xYb_{1-x}Ba_2Cu_3O_{7-\delta}$ ($x = 0.1, 0.2, 0.3, 0.4, 0.5$) bulks to make low melting temperature bulks with an addition of Y element that may promote crystal growth.

2. Experimental Procedure

Bulk samples were prepared by solid state reaction using $Y_2O_3$ (99.999 %), $Yb_2O_3$ (99.95 %), $BaCO_3$ (99.9 %) and $CuO$ (99.9 %) powders in the stoichiometric ratio $Y:Yb:Ba:Cu = x: (1-x): 2: 3$ ($x = 0.1, 0.2, 0.3, 0.4, 0.5$). The powders were sintered at 850 °C for 10 hours in air. After that, the sintered powder was grinded and pressed into bulks at 700 MPa. These bulks were then calcined under the same conditions as the sintering process illustrated in figure 1. Firstly, the temperature was increased to 700 °C in 3 hours before being held constant for 2 hours. Then, the temperature was increased to 850 °C at 2.5 °C/min. After sintering or calcination at 850 °C for 10 hours, the temperature was slowly cooled to 100 °C over 8 hours. The total duration of the sintering was 1 day; the process was conducted in air.

In this experiment, two joint samples were fabricated using Gd123-coated conductors manufactured by Sumitomo Electric Industries, Ltd. The coated conductor is copper stabilized (20 μm thick on one side) and 4 mm wide. The thickness of Gd123 and buffer layers are 3-4 μm and 0.4 μm, respectively. Substrate layers consist of 2 μm thick Ni, 17 μm thick Cu, and 100 μm thick stainless steel. The protective Ag layer on the Gd123 layer has a thickness of 7-9 μm.

A sample was first prepared by cutting two coated conductor strips of equal lengths. Then, Cu and Ag layers on both sides were partially delaminated with a cutter. These two layers were left intact on the sample ends to attach current leads and voltage taps. The coated conductors were stacked such that the exposed Gd123 layers were facing each other. Following this step, a thin YYb123 bulk was inserted between the copper-free ends. The bulk was grounded beforehand with 2000 grit sandpaper to reduce its thickness of 1 mm to 50-80 μm. The schematic diagram of a joint sample is presented in figure 2. The joint samples were thermally treated at 930 °C for 1 min in $O_2$ atmosphere.

The first sample was shortened, and the bulk end was preserved for XRD measurements. Critical current properties were evaluated on the second sample by standard four-probe method. In order to keep the electrodes apart, an insulating layer such as polyimide tape is inserted between them.

![Temperature profile(setting) for sintering or calcination process](image-url)
3. Results and Discussion

X-ray diffraction (XRD) measurements were carried out with CuKα1 beam line by Rigaku Miniflex 600. The XRD patterns of \( x = 0.1, 0.2, 0.3, 0.4, 0.5 \) powders are shown in figure 3. 123 phase is mainly observed in all the powders. 211 phase is present due to peritectic decomposition of YYb123 phase: YYb123(s) \( \rightarrow \) YYb211(s) + Liquid. Figure 4 shows the XRD patterns for bulk samples. The bulk patterns are still seen to be primarily composed of 123 phase, but there is a decrease in peak intensity compared to powder samples. In the case of \( x = 0.1 \) bulk, the peak intensity of 123 has been reduced to be almost same as that of 211 phase. This may be attributed to the change in composition of Y and Yb during heat treatment. 211 phase in the powder or bulk can act as effective pinning centres for vortices in the superconducting state and thus may improve superconducting properties. BaCuO as liquid during joint process is used for bonding of the coated conductors with high mechanical strength. Therefore, BaCuO and 211 phase are necessary in joint technology. This is a common characteristic of superconducting joint between REBCO-coated conductors.

Among the fabricated bulks, \( Y_{0.1}Yb_{0.9}BCO \) bulk is considered to possess the lowest melting temperature due to highest composition ratio of Yb. This is in line with our interest to join using the lowest melting temperature bulk. To determine an appropriate joining temperature, a \( Y_{0.1}Yb_{0.9}BCO \) bulk was heat treated at different temperatures for 10 min in air. Figure 5 shows XRD patterns of \( Y_{0.1}Yb_{0.9}BCO \) after each heat treatment. 123 phase peaks including the highest peak at \( 2\theta = 32° \) are observed to be largely reduced at 950 °C, indicating that the bulk had almost melted. This result suggests that the melting point of \( Y_{0.1}Yb_{0.9}BCO \) bulk is about 930–950 °C. Given that CJMB method requires a temperature between the melting temperatures of Gd123 and YYb123 phases, a suitable joint temperature may be in the range of 930–950 °C. Previously, a superconducting joint was successfully fabricated with a YbBCO bulk at 930 °C for 1 min [5]. The small difference in composition ratio between YbBCO and \( Y_{0.1}Yb_{0.9}BCO \) suggests that a joint using \( Y_{0.1}Yb_{0.9}BCO \) bulk could also be fabricated at the same temperature of 930 °C. Thus as a first step in this study, 930 °C was chosen to prepare the joint sample.

![Figure 2. Schematic diagram of joint sample](image)

Figure 2. Schematic diagram of joint sample
To investigate the crystal growth of YYb123 on the coated conductor, a thin YYb123 bulk is used to prepare a joint. Figure 6 shows the XRD patterns for powder, coated conductor and joint boundary. In the top pattern for the joint boundary, (00l) peaks like YYb123 single crystal were observed near the (00l) peaks of Gd123 in coated conductor. This indicates that crystal growth of YYb123 phase on the thin bulk has been induced. This result may be useful in joints between coated conductors with high quality of growth in future study.

Figure 7 shows the voltage versus current characteristics of a prepared joint. The joint using a YYb123 (x = 0.1) bulk is measured at 77 K. The patterns show superconducting behavior clearly. The criterion to determine the critical current is 1 μV. The critical current of the joint is 6.5 mA. This critical current is too small for application and should be increased. However, as a first step of this study, the achievement of a superconducting joint is considered to be important in the future development of joint technology.

Figure 5. XRD patterns of $Y_{0.1}Y_{0.9}BCO$ bulk heat-treated at different temperatures for 10 min in air.

Figure 6. XRD patterns of the joint boundary on thin bulk (top), the Gd123-coated conductor (middle) and $Y_{0.1}Y_{0.9}BCO$ powder (bottom).
4. Conclusion
We have synthesized $Y_xYb_{1-x}Ba_2Cu_3O_{7-\delta}$ ($x = 0.1, 0.2, 0.3, 0.4, 0.5$) bulks to be used in conjunction with the CJMB method. The melting temperature of $x = 0.1$ bulk was about $940 \degree C$ in air. Crystal growth of YYb123 is observed at the joint boundary, indicating the possibility of a joint with high critical current in the future. The critical current of the joint using $x = 0.1$ bulk is $6.5 \text{ mA}$. Given the small value, there is a need for improvement in the critical current for actual use in magnets. This can be achieved by optimizing the joining process. Further investigations are needed to determine the parameters of controls including heat treatment temperatures and annealing oxygen concentration. As a first step of future research, a superconducting joint was successfully developed.

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References
[1] Kimura K et al. 2015 IEEE Transactions on Applied Superconductivity 25 6604204
[2] Park Y, Lee M, Ann H, Choi Y, Lee H 2014 NPG Asia Mater 6 e98
[3] Jin X, Yanagisawa Y, Maeda H and Takano Y 2015 Supercond. Sci. Technol. 28 075010
[4] Zhang Z, Wang L, Liu J, Wang Q 2019 Crystals 9 492
[5] Jin X, Yanagisawa Y, Maeda H 2018 IEEE Transactions on Applied Superconductivity 28 4602604

Figure 7. Voltage versus current plot for a joint using $x = 0.1$ bulk on Gd123-coated conductor. The critical current of the joint is $6.5 \text{ mA}$. 
