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Preparation of (Bi, Pb)-2223/Ag tapes by high temperature sintering and post-annealing process

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Abstract

A novel heat treatment process was developed to fabricate (Bi, Pb)-2223/Ag tapes with high critical current density (J_c). The process can be divided into two parts: reformation and post-annealing. Tapes were first heated to the maximum temperature (830–860 °C) followed by slow cooling (reformation). Then, tapes were annealed between 760 and 820 °C (post-annealing). Reformation is expected to produce a large amount of liquid phase which may heal micro-cracks, decrease porosity, and improve grain growth. However, since the sintering temperature is beyond the Bi-2223 single-phase region, much Bi-2212 and secondary phases will appear at the Bi-2223 grain boundaries. During post-annealing, these phases may convert to (Bi, Pb)-2212 and even to Bi-2223. Hence, the J_c values for these tapes are significantly improved. The microstructure and the phase composition were investigated by high-energy synchrotron XRD and SEM/EDX. Some process parameters e.g. sintering temperature, cooling rate, and post-annealing time were optimised.

Keywords: (Bi, Pb)-2223/Ag tape; Critical current density; Texture

1. Introduction

Conductors made from (Bi, Pb)$_2$Sr$_2$Ca$_2$Cu$_3$O$_x$ (Bi-2223) high temperature superconductor material have a high potential for applications in energy technology. Thus, many efforts have been made to improve their critical current density. Although the mechanisms governing the phase transformation and texture evolution in Bi-2223 superconducting tapes are not yet clear [1,2], some consensus has been reached in terms of microstructure and properties of tapes. For example, it is quite sure that the grain connectivity of the microcrystals within the ceramic core of the conductor is crucial for the current transport [3]. The grain connectivity can be improved by increasing texture degree of Bi-2223 grains, healing cracks created by mechanical deformation, and reducing secondary phases. Our previous work shows [4,5] that sintering tapes for a short time at the temperature beyond Bi-2223 single phase region may produce more liquid phase which helps to densify

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the oxide core and heal cracks. Recently, Xia et al. [6] reported that continuous cooling sintering could increase the core density. However, more secondary phases were introduced in both cases.

In this work, we proposed a novel heat treatment process which consists of two parts: sintering at high temperature followed by slow cooling (reformation) and annealing at relatively low temperature (post-annealing). Reformation is expected to induce more liquid phase during sintering, and post-annealing is suggested to minimize the secondary phase content in the tape.

2. Experimental

Bi-2223/Ag multi-filamentary tapes were fabricated by the standard powder-in-tube method. The details of the tape fabrication process have been described elsewhere [7]. The tapes were cut into pieces of 3.6 cm length for the experiment. After the first heat treatment, the tapes were uniaxially pressed before going through the reformation and post-annealing process. No mechanical deformation was applied in between later steps (see Fig. 1). A self-designed temperature gradient furnace was used to investigate the optimal reformation and post-annealing temperatures.

Since all the samples have the same dimensions, the critical current (I_c), instead of critical current density (J_c), was used to characterize the superconducting properties of tapes with different heat treatment process. The transport critical current was measured by a DC four probe method at 77 K using 1 μV/cm criterion. High-energy synchrotron X-ray diffraction (XRD) analysis was done at HASYLAB, Hamburg to examine the phase composition and the texture degree [8]. The microstructure of the fully processed tapes was investigated by SEM.

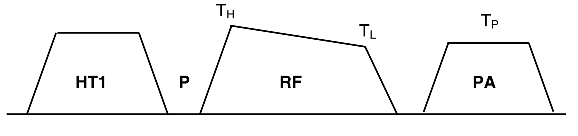

Fig. 1. Heat treatment process for Bi-2223 tapes in this experiment. P: uniaxial pressing; RF: reformation; PA: post-annealing.

3. Results and discussion

Fig. 2 shows the I_c values of tapes sintered at different temperatures. It should be pointed out that except the reformation process, all the other heat treatments were done in an isothermal tube furnace. The temperature gap for reformation ($\Delta T = T_H - T_L$) is always 60 °C, and the cooling rate is 1 °C/h. The post-annealing is 796 °C/20 h. After reformation process, the highest I_c value is around 30–34 A, which is equivalent to that of traditionally processed tape (32–33 A). After post-annealing, the highest I_c values of 38–40 A were achieved for T_H in the range 842–847 °C. SEM observations show that the tape sintered at 841 °C has dense and highly textured microstructure. While, a tape sintered at 850 °C contains a large amount of secondary phases (2201 and cuprates), and a tape sintered at 832 °C has a porous core.

Three different cooling rates (1, 0.8, and 0.5 °C/h) followed by the post-annealing at 796 °C/20 h were tested. The tape with T_H of 846 °C and cooling rate of 0.8 °C/h shows the best transport properties. Fig. 3 shows the dependence of texture degree and critical current for the tape cooled at 0.8 °C/h from the reformation temperature (T_H). It is clear that with increasing of T_H, the FWHM value decreases. Meanwhile, the I_c value increases greatly. However, it was also found that when T_H is 850 °C the I_c value was lower than at 846 °C even though the texture degree is the highest. One

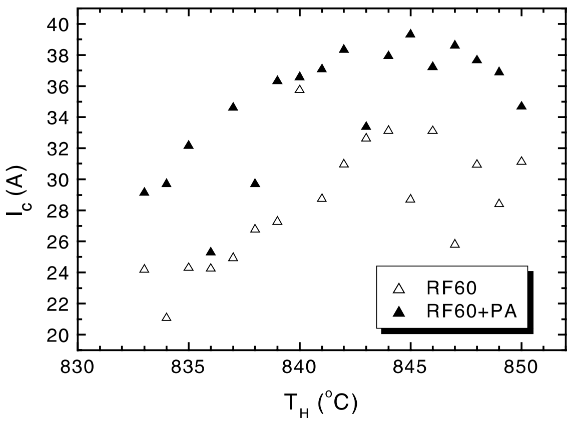

Fig. 2. Dependence of I_c values on the reformation temperature (T_H).

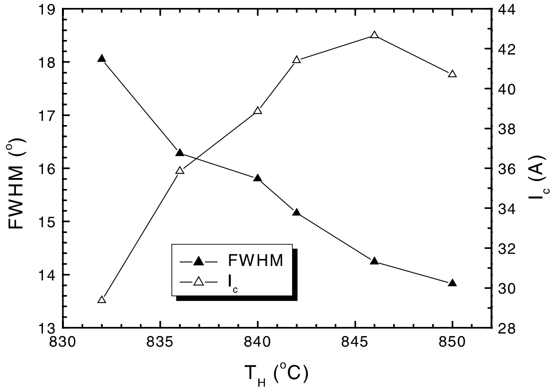

Fig. 3. Texture degree and critical current for tapes sintered at different T_H.

possible reason is that the increased amount of secondary phases limits the current transport paths. The relative percentage of 2212 in the samples was determined by using the $(1\,1\,5)_{2212}$ and $(1\,1\,5)_{2223}$ peak intensities in the XRD patterns. Fig. 4 quantitatively shows the relationship between the relative 2212 amount and the reformation temperature. For comparison, the I_c values of the same samples are also plotted in this figure. It is seen that, the 2212 content first decreases with increasing T_H, and reaches the lowest level when T_H is at 846 °C. Then the 2212 amount increases with the increase of T_H. This phenomenon is consistent with the evolution of I_c, which implies that the secondary phases may play an important role in this case.

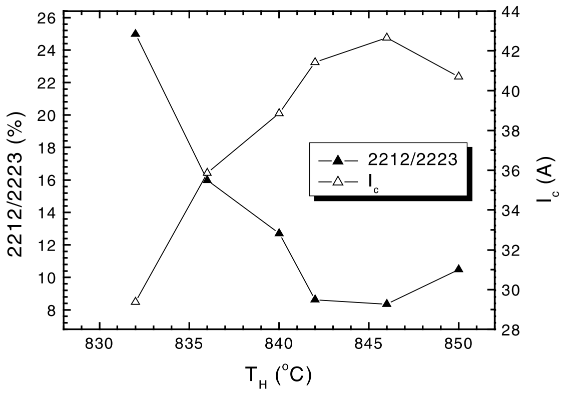

Fig. 4. 2212 phase content and critical current for tapes sintered at different T_H.

Since the reformation process is carried out over a large temperature range, the phase transformation is very complex. Possible reactions could be like this: during the slow cooling from T_H to T_L, the liquid phase coming from the melted Pb-rich phase and 2212 phase decomposition precipitates and forms 2223 grains. The 2223 grains grow up and become texturized 2223 colony. Meanwhile, 2201 phase and some cuprates are also formed because of the slow 2223 formation rate. The 2201 phase and the cuprates may react and form 2212 and 2223 phases at relatively low temperature e.g. 800 °C. This could be the reason why post-annealing can greatly improve the current transport properties. The residual content of 2201 and cuprates due to too high T_H and/or a high cooling rate will exist in the oxide core as barriers for current transport.

4. Summary

The Bi-2223 tapes were made by using reformation and post-annealing method. The current transport properties of these tapes were improved. Microstructure analysis showed that these tapes had better texture and denser superconducting core than traditionally processed tapes. By tuning the cooling rate and the sintering temperature in the reformation process, the secondary phases such as 2201, 2212 and cuprates could be minimized. Hence the critical current density could be improved even further.

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