Melt Growth and Superconducting Properties of Textured Ag-$YBa_2Cu_3O_7$ Conductors

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Abstract- **Textured Ag foils 200 μm thick have been obtained after cold rolling and recrystallization at 800 °C with a reduction ratio of up to 95 %. Subsequently, a thick layer of $YBa_2Cu_3O_7$ (20 μm) has been deposited by screen printing. A directional solidification process under a temperature gradient with displacement velocity between 1 mm/h and 10 mm/h has been performed to induce a biaxial texture to the superconducting layer. This last treatment has been performed at low PO_2 atmosphere and at maximum temperatures below 945°C. The texture of the layers has been studied by x-ray diffraction, rocking curves and pole figures, whilst the critical currents have been measured inductively by SQUID magnetometry. The relationship between the in-plane crystalline orientation and the observed critical currents has been discussed.**

I-INTRODUCTION

The development of high critical current superconducting tapes is a topic that has been recently strongly stimulated by the discovery of successful thin film deposition techniques on metallic substrates. Impressive high critical current at 77K and smooth field dependencies have been demonstrated in $YBa_2Cu_3O_7$ (YBaCuO) coatings on buffered Ni or Hastelloy tapes when a biaxial deposition is achieved, either through the IBAD technique [1] or the RABiT methodology [2]. In the first case, a biaxial YSZ buffer layer is deposited by IBAD and later on YBCO is evaporated by pulsed laser deposition. In the second case, rolling-assisted-biaxially textured substrates of Ni were achieved and later on a combination of several buffer layers were vapor-deposited. The succesful development of a second generation of conductors may launch high temperature superconductors for power applications provided that materials processing becomes cost-effective in the km range [3,4].

The need to achieve low cost biaxial YBCO conductors has launched new efforts on the use of thick film processing techniques which were earlier investigated and were dropped because low critical currents were achieved due to the persistence of high angle grain boundaries.

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Melt processing of YBCO on metallic substrates was first reported by Jin et al [5] but it became immediately apparent that only Ag-based alloys displayed low chemical reactivity to YBaCuO . Triggered by this limitation several authors have recently investigated the preparation of YBaCuO biaxial texturing over Ag single crystals or textured foils.

Texturing of Ag foils can be easily achieved through cold working and further annealing at high temperatures. The resulting texture is {110}<1 $\bar{1}$0>, even if minor contamination of other grain orientation can also be found.

After the investigation of Budai et al [6] of the epitaxial growth of vapor deposited (PLD) YBaCuO on different Ag surfaces, we know that a single in-plane orientation is only achieved in the (110) case. When (001) or (111) Ag surfaces are used several discrete orientations of YBaCuO a-b plane were observed and this result was justified on the basis of lattice matching considerations (near coincident lattice model) .

It seemed then promising to further investigate if biaxial texturing of YBaCuO could be achieved over {110} textured silver tapes. Biaxial melt texturing of bulk YBaCuO has been achieved through controlled grain nucleation, either by using seeds or by directional solidification under temperature gradients [7]. Particularly, a vertical Bridgman geometry has allowed to grow long single domains of YBaCuO 123/211 composites ($l \approx$ 100mm) [8,9].

To proceed with this technique in Ag-YBaCuO thick films, however, we require to perform the peritectic reaction under reduced PO_2 atmosphere in order to process at temperatures below the melting point of silver. Several authors have investigated the Ag-YBaCuO phase diagram and they have shown that melt texturing of YBaCuO on solid Ag substrates may be carried out in a narrow T-PO_2 window [10-11].

In this work we investigate the use of melt texturing as a technique to obtain biaxial deposition of YBaCuO thick films on textured Ag foils prepared by cold rolling.

II-EXPERIMENTAL

Textured silver substrates were prepared after successive cold rolling of polycrystalline, high purity (>99.9 %), Ag bars. The final thickness was 200μm and the total reduction was 95%. A primary recrystallization process of the foils was achieved afterwards through thermal anneal at 800°C in air.

The thick film deposition process used in this work was screen printing and the typical final thickness after the melt texturing process was 20±5μm. Commercial YBaCuO powders were used to prepare the slurry which was deposited on Ag tapes of typical dimensions 4x20 mm^2. The organic binder was eliminate through a heating protocol in air which consisted mainly of a controlled warming up step and a final isothermal anneal at 800°C during about 15 hours. The directional solidification process was performed in an horizontal furnace with controlled atmosphere. After heating the sample in a homogeneous region, the tape is displaced through a region having a temperature gradient G≈30°C/cm. The typical characteristics chosen for the growth process are T^{max}≈ 920°C , PO_2 ≈10^{-3} tor and R≈1.5-10mm/h. Under these conditions the peritectic reaction Y_2BaCuO_5(s)+ [$3BaCuO_2$+2CuO](l) →$2YBa_2Cu_3O_x$ (s) is complete without Y_2BaCuO_5 particles remaining trapped, no evidences for liquid losses are observed and the silver foil remains solid. Overall, the processing time did not exceed 48 hours. Afterwards the duration of the oxygenation process was optimized to about 2 hours at 450°C in flowing O_2. The purity of the final samples was examined with X-Ray powder diffraction patterns, also evidencing the crystallographic texture. Quantitative estimation of the texture developed in the cold rolling process of the substrate and after melt texturing the YBaCuO coating, was investigated from X-ray pole figures. Optical and SEM microscopy was used to observe the crystallites morphology after the growth process. The superconducting properties of the samples were investigated by using a SQUID magnetometer. Low field dc susceptibility measurements were used to determine the transition temperature. Temperature dependent critical currents in self-field were deduced from the observed temperature dependence of the magnetic remanence. Finally, isothermal magnetic hysteresis loops, in fields up to 5T, were recorded to asses the field dependence of the critical currents. In all the cases the generalized critical state model with the whole dimensions of the samples were used to calculate the critical currents[12].

III-RESULTS AND DISCUSSION

Extensive cold rolling and recrystallization of Ag foils lead immediately to oriented {110} planes normal to the rolling direction ($1\bar{1}0$) as evidenced by θ-2θ X-ray patterns and pole figures (Figure 1(a) and 2), however, a single in-plane orientation was more difficult to achieve. In Figure 1(a), for instance, the rolling direction is found to lie at about 30° and 45° away from <$1.\bar{1}0$> axes in two main grains having respective weight of 80% and 20%. In θ−2θ geometry, however, only the (220) reflection was systematically observed in the silver foils. In Figure 2 a typical powder diffraction pattern of a sample after the melt texturing process is displayed. As it may be observed only the $YBa_2Cu_3O_7$ and Ag reflections are observed. In samples localized at the final part of the tape in the directional solidification process additional reflections arising from remnant liquid phase are also detected. This evidences the main advantage of the directional solidification process, i.e. the insulating impurities associated to an uncompleted peritectic reaction are pushed by the growth front and so clean grain boundaries should remain in the remaining part of the tape. As it may also be observed in Fig.2, the (00l) reflections from YBaCuO are dominating. However, the (103)/(013) reflections from $YBa_2Cu_3O_7$ and the (311) reflection from Ag are also sometimes observed, thus indicating that some degree of out-of-plane texture was lost during the high temperature growth. This is indicative of the relatively low stability of the {110} texture of silver [11,13], which as it is shown here, it may be partially destroyed when a YBaCuO coating is deposited it, probably due to the stresses induced from a differential thermal expansion. We estimated that the twin texture {311} of silver never exceeded 4% of the total volume before melt processing while this value may increase to 10% in the solidified sample.

As a consequence of the partial loss of the {110} texture of the silver foils the pole figure of YBaCuO-coated tapes has poor crystallite orientation (Figure 1(b)). As it may be observed in this Figure the {103} peaks are still dominating but their width in the ϕ−scan achieve values around 20° . This value is somehow larger than that

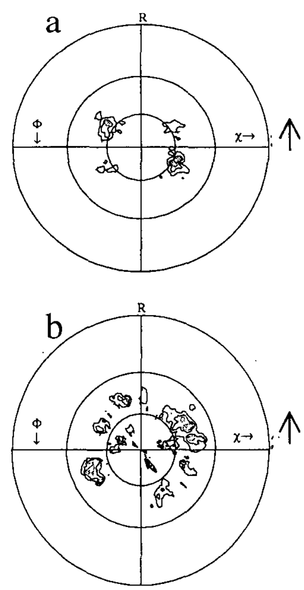

Fig.1 (a) {111} pole figure of cold rolled Ag foil after primary recrystallization . (b) {103} pole figure of a melt processed $YBa_2Cu_3O_7$ tape.

reported in samples deposited on Ag foils prepared by hot-rolling and recrystallization [12] which leads to a sharper definition of the substrate texture. It should be expected then that an improved in-plane texture for the YBaCuO coating developed here would be achieved with substrates prepared by hot rolling and recrystallization.

It is, however, remarkable that a complete peritectic reaction could be achieved under our present conditions even if very high crystallization rates were used (up to 10mm/h were tested). This means that the limiting growth rate actually arises from the transverse direction and so the crystal nucleation occurs at the Ag interface. Consequently, with this observation the grain size (up to 500 μm) of the YBaCuO crystallites along the rolling direction of the tape decreases when the processing rate is increased.

A typical superconducting transition may be observed in Figure 3 , where it may be seen that even if the transition remains relatively sharp, they never reach values higher than $T_c^{on} \approx 85K$, thus indicating that probably some carbon impurities remain in the YBaCuO matrix after the heating process of the organic binder used to carry out the screen printing of the powders. A thorough analysis of this problem is currently being performance since it generates a strong degradation of the high temperature properties of the superconducting tapes.

In Figures 4(a) and 4(b) we display the temperature dependences of Jc in two samples without embedded flux and with some remnant flux, respectively. As it is clearly seen the intergranular critical currents are reduced by a factor 4 when the grain boundaries are contaminated by the insulating phases, this demonstrates the validity of the directional solidification approach.

Nevertheless, we must note that the observed critical currents at 5K remain still a factor 20 below those observed in bulk melt textured YBaCuO [14]. The field dependence, on the other hand, appears to be weak at low temperature and strongly enhanced when approaching T_c.
This suggests that J_c is weak-link-limited at high temperatures.

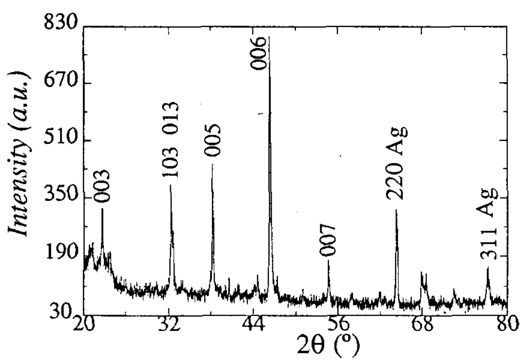

Fig.2 Typical X-ray powder diffraction pattern of a melt processed $YBa_2Cu_3O_7$/Ag tape.

We must note also that the observed critical currents are much lower than those which should be expected on the basis of our distribution of the crystallites orientation within the plane [4] as determined by φ-scan (fig. 1(b)). It is more likely then that additional disorder associated to the remaining twin texture of silver generate this additional decrease in J_c.

It must be stressed, finally, that even if the observed critical currents are the highest observed so far in $YBa_2Cu_3O_7$

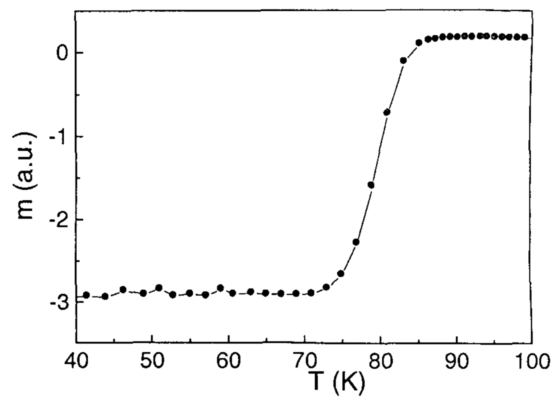

Fig. 3 Superconducting transition measured with a SQUID magnetometer at H=10Oe with H//c.

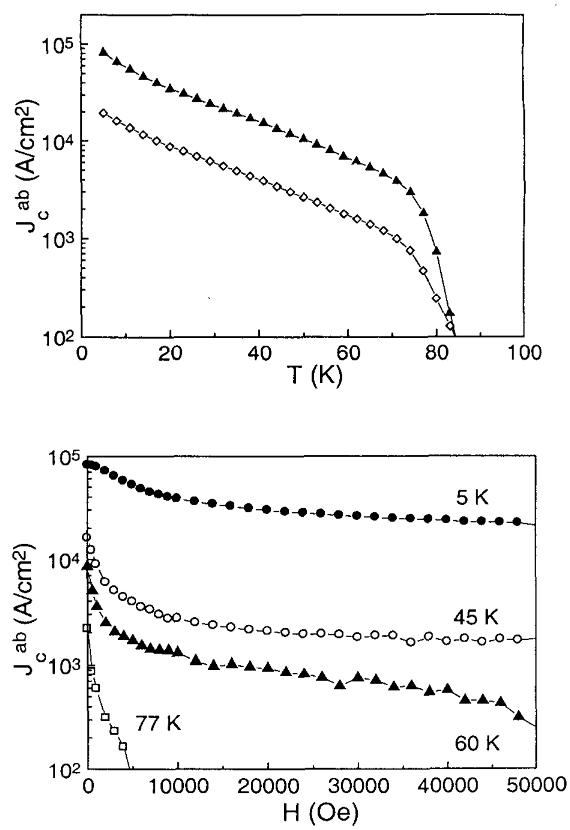

Fig. 4 (a) Temperature dependence of the inductive critical current density of a tape without remnant liquid (▲) and with remnant liquid.(◊). (b) Field dependence of the critical currents in the sample without remnant liquid.

tapes prepared by melt processing, there is still a considerable room for improving them through further enhancement of the stabilization of the {110} texture or by using buffered layers chemically compatible with melt processing

IV-CONCLUSIONS

In conclusion, we have shown that the combination of a two step process to prepare biaxially textured YBaCuO tapes by means of the thick film technology is a promising low price technology which could compete with more powerful methodologies such as IBAD deposition. Our approach differs from that reported previously in the use of directional solidification conditions. This new approach has, as its main advantage, that remnant flux may be completely avoided leading to a less disturbed nucleation and growth of the YBCO grains and to cleaner grain boundaries. The main issue which should be solved is to find how to stabilize and define more deeply the {110}<1 $\bar{1}$0> texture of silver. Another alternative could be to develop buffer layers on more robust surfaces, such as the cubic texture of Ag {100}<100>, and which should remain benign from the point of view of the electrical properties of the YBaCuO phase. A similar approach has been followed recently in thick films grown by LPE [15].

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