

The Effect of Wire Drawing and Cold Rolling on the Precursor 2212 Phase Texture and its Subsequent Influence on the $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ Texture In Ag-clad Tapes

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Abstract—We have studied the effect of wire drawing and cold rolling on the texture development of precursor $(\text{Bi,Pb})_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ (2212) phase and its influence on the subsequent $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ (2223) phase texture in Ag-clad tapes. A mixture of $(\text{Bi,Pb})_2\text{CuO}_4$ and SrCaCuO_3 powders having an overall 2223 composition was reacted to form the 2212 phase at different stages of deformation. X-ray diffraction peak height analysis was used to quantify the 2212 texture $F_{2212} = I(008)/[I(008)+I(115)]$ at various oxide filament thickness. Wires containing powder reacted to the 2212 phase prior to loading into a Ag tube had $F_{2212} \approx 0.3$ after drawing. This texture increased to $F_{2212} \approx 0.7$ upon rolling. Two other composites reacted to the 2212 phase immediately after drawing, and after being drawn and partially rolled, had worse initial textures, but improved during rolling to $F_{2212} \approx 0.7$. Samples reacted to 2212 after rolling was complete had $F_{2212} \approx 0.35$. The results indicate that the 2212 texture reaches a maximum value of $F_{2212} \approx 0.7$ and remains at that level upon subsequent rolling. Samples of filament thickness $\approx 80 \mu\text{m}$ were then reacted to form 2223. For the samples which contained 2212 that had received at least some mechanical deformation, the 2223 phase formed with a texture factor $F_{2223} > 0.9$, while the tape that contained 2212 formed after rolling developed a 2223 texture of only $F_{2223} \approx 0.75$. The results show that there is a substantial texture enhancement upon reaction to 2223, which could be the result of either the 2223 formation mechanism or geometrically constrained grain growth.

I. INTRODUCTION

One of the main critical current density (J_c) limiting mechanisms in BSCCO-2223 is poor connectivity between the superconducting grains. A vital aspect of overcoming this problem is to develop highly textured grains in a Ag-clad tape [1]-[6].

Silver-clad tapes are usually made by the oxide-powder-in-tube method, in which a Ag tube is packed with ceramic powder and is plugged, swaged, and drawn into a wire. The wire is then rolled into a tape. Usually the oxide powder, of overall nominal composition $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$, is reacted to form the precursor 2212 phase (and other secondary phases like $(\text{Ca,Sr})_2\text{PbO}_4$ and $(\text{Ca,Sr})_2\text{CuO}_3$) prior to loading the billet. The high T_c (110 K) 2223 phase is formed at final size by reaction of the 2212 phase and secondary phases. The reaction is believed to involve a transient liquid phase, which helps to heal cracks that are formed during the intermediate

deformation steps [7]-[11]. Two models for the 2223 formation mechanism, intercalation [1],[12],[13] and nucleation and growth [14], suggest different dependencies of the resultant 2223 grain texture on the precursor 2212 texture. Intercalation suggests that the 2212 grains serve as a grain-by-grain template for 2223 formation, and thus the grain texture before and after reaction should be similar. A nucleation and growth formation mechanism suggests that the liquid phase at least partially dissolves the 2212 grains, and that the 2223 grains are nucleated from this melt. This formation mechanism suggests a lesser dependence of the 2223 texture on the precursor 2212 texture. However, the plate-like grain morphology of both 2212 and 2223, their anisotropic grain growth, and the highly aspected geometry of the fine filament tapes also influence the 2223 texture, making interpretations of texture changes difficult. As there has been experimental evidence for the validity of both mechanisms, there is still some controversy over which model is correct, or whether both processes occur simultaneously [15]. To add to our knowledge of the texture development in 2223 tapes, we used a relatively unusual mixture of $(\text{Bi,Pb})_2\text{CuO}_4$ and SrCaCuO_3 powders as starting materials [8]. Neither of these compounds are micaceous and they are therefore expected to exhibit much less deformation-induced texturing than more conventional 2212-based powders. This powder mixture was reacted to 2212 and secondary non-superconducting phases at different points in the tape metalworking sequence. This allowed us to determine both the contributions of various metalworking processes on the precursor 2212 texture, and how the precursor 2212 texture influences the subsequent 2223 texture.

II. EXPERIMENTAL PROCEDURE

Two distinct types of starting monofilament wires were fabricated by hand-packing $(\text{Bi,Pb})_2\text{CuO}_4$ and SrCaCuO_3 powders having an overall composition of $\text{Bi}_{1.7}\text{Pb}_{0.3}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ into 99.9% silver tubes with an initial outer diameter of 1.35 cm (I.D. ~ 1.0 cm). The difference between these two starting wires was that one contained powder that was reacted to 2212 by heating the $(\text{Bi,Pb})_2\text{CuO}_4$ and SrCaCuO_3 mixture in air for 10 minutes at 850°C and 12 hours at 860°C before being packed into the silver tube. This wire/tape will be referred to as Tape A. The other wire contained just the blended cuprates. These composites were then plugged, swaged, and cold drawn to a diameter of ~ 1.2 mm, (true strain ≈ 2.5) using a 10% reduction in area per

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TABLE I
NAMES AND DESCRIPTIONS FOR THE FOUR TAPES IN THIS STUDY

Tape Name	Description (when 2212 was formed in metalworking sequence)	Filament thickness when 2212 was formed (μm)
A	Pre-reacted	Before tube packing
B	Reacted after drawing	700
C	Partially rolled then reacted	220
D	Reacted after rolling	80

pass. The composite which contained the blended cuprates was then sectioned into three parts, thereafter differentiated by when the 2212 phase was formed in the rolling sequence. Table I lists the names and descriptions for these sections. One section (Tape B) was reacted immediately after drawing to a diameter of ~ 1.2 mm. A second (Tape C) was reacted after being rolled to an overall thickness of ~ 380 μm (filament thickness ~ 220 μm), and the third (Tape D) was reacted to 2212 after rolling to a final overall thickness of ~ 150 μm (filament thickness ~ 80 μm). The $(\text{Bi,Pb})_2\text{CuO}_4$ and SrCaCuO_3 powder mixtures in the Ag wires were converted into 2212 (and secondary phases) by heat treating for 10 and then 6 hours at 600°C and 825°C , respectively, in a flowing 7.5% O_2 /balance N_2 atmosphere. This treatment led to the formation of 2212, but no 2223 phase, as determined by x-ray diffraction (XRD). A schematic flow chart of the experimental design is shown in Figure 1.

All of the round wires were rolled into tapes with 3.81 cm

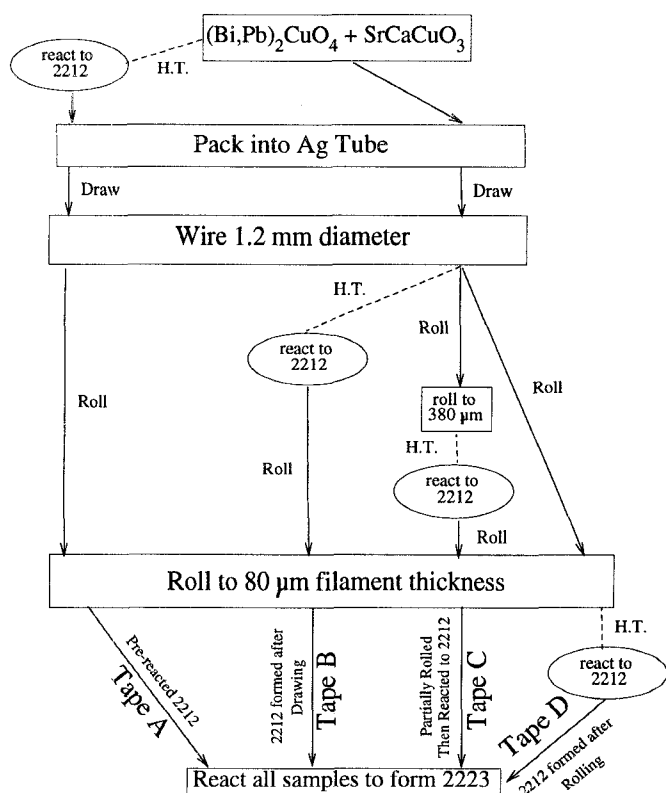


Fig. 1. A schematic flow chart of the experimental design which was used to create monofilament tapes with different 2212 precursor textures.

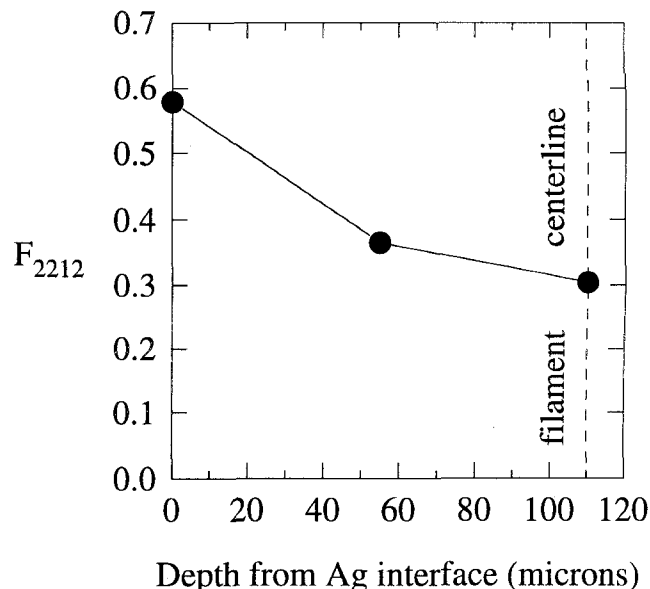


Fig. 2. Texture profile from the Ag interface to the midpoint of a 220 μm thick filament sample from Tape C. Texture decreases away from the Ag.

diameter work rolls, using a nominal 5% thickness reduction per pass. All tapes were rolled through a total true strain of ~ 6.0 , to a filament thickness of ~ 80 μm . Samples were taken periodically during the rolling sequence for texture analysis of the 2212 phase. Samples of overall thickness ~ 150 μm (filament thickness ~ 80 μm) from each of the four different tapes were then converted from 2212 to 2223 by heat treating for 250 hours at 825°C in a flowing 7.5% O_2 /balance N_2 atmosphere, without any intermediate deformation step. The texture of the 2212 and 2223 grains was studied by XRD analysis with $\text{Cu-K}\alpha$ radiation, using a version of the Lötgering method [16]. The cold worked 2212 samples were heat treated for 1 hour at 780°C to get well defined x-ray peaks [6],[17]. To prepare samples of tapes thinner than 250 μm overall thickness, the Ag tape sides were cut and peeled apart. To measure thicker samples, the Ag was removed from one side by masking one side with varnish and etching the other with a mixture of NH_4OH and H_2O_2 [11],[18]. Because it was found that the region immediately adjacent to the Ag had a higher texture than the central portion of the tape (Figure 2), samples with cores thicker than 120 μm were measured before and after being fine ground to their midpoint using SiC paper. The texture value that is given for the etched samples is the texture after grinding, and thus is representative of the bulk, rather than the interface texture.

III. RESULTS

To quantify the grain texture by XRD, we used texture parameters, F_{2212} and F_{2223} , defined as

$$F_{2212} = \frac{I(008)}{I(008) + I(115)} \quad F_{2223} = \frac{I(0010)}{I(0010) + I(115)}$$

where $I(008)$, $I(0010)$, and $I(115)$ are the heights of the (008), (0010), and (115) diffraction peaks of the corresponding 2212 or 2223 phase.

Figure 3 shows how the 2212 texture evolved in the rolling sequence. Tape A had a texture of $F_{2212} \approx 0.3$ at a filament thickness of $\sim 700 \mu\text{m}$, which increased to $F_{2212} \approx 0.7$ at $\sim 260 \mu\text{m}$, and remained at that level while rolling to $80 \mu\text{m}$. Tape B had a texture of $F_{2212} \approx 0.2$ at a filament thickness of $\sim 700 \mu\text{m}$, which increased to $F_{2212} \approx 0.7$ at a final filament thickness of $\sim 80 \mu\text{m}$. Tape C had an initial texture of $F_{2212} \approx 0.3$ at a thickness of $\sim 220 \mu\text{m}$, which increased to $F_{2212} \approx 0.7$ at a filament thickness of $\sim 80 \mu\text{m}$. Finally, the sample that was reacted to 2212 after rolling (Tape D) had a texture of $F_{2212} \approx 0.35$ at a filament thickness of $\sim 80 \mu\text{m}$. Thus a limiting texture of $F_{2212} \approx 0.7$ was achieved in all 3 composites which were deformed when the 2212 phase was present (A-C) but little texture was evident in the composite deformed while the powders were $(\text{Bi,Pb})_2\text{CuO}_4$ and SrCaCuO_3 (Tape D).

Samples of Tapes A-D at a filament thickness of $\sim 80 \mu\text{m}$ were reacted to form the 2223 phase. Table II lists the grain textures before and after reaction to 2223. Tapes A-C, all with precursor 2212 texture of $F_{2212} \approx 0.7$, developed 2223 grains with $F_{2223} \approx 0.95$ after reaction. An example of this behavior is shown in Figure 4, which shows XRD patterns of a $80 \mu\text{m}$ thick sample from Tape B both before and then after reaction to 2223. Note from Table II that all of the samples which contained mechanically-deformed 2212 powders

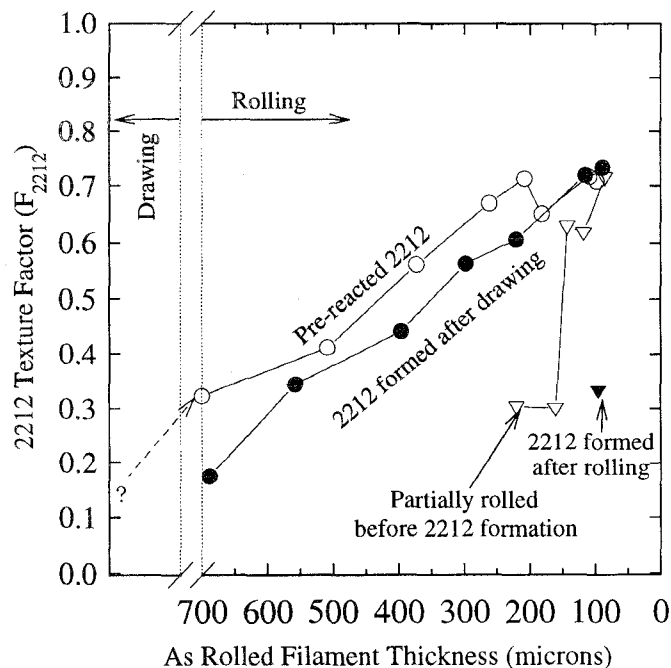


Fig. 3. 2212 texture evolution during the rolling sequence. The 2212 texture in rolled samples quickly reaches a maximum of $F_{2212} \approx 0.7$.

Tape Name	F_{2212} (2212 texture before 2223 formation)	F_{2223} (2223 texture after reaction)	f_{2223} (% 2223 after 250h at 825°C)
A	0.71	0.97	96
B	0.73	0.97	96
C	0.72	0.94	98
D	0.35	0.75	83

$$F_{2212} = I(008)/[I(008)+I(115)]; F_{2223} = I(0010)/[I(0010)+I(115)]$$

$$f_{2223} = I(0010)_{2223}/[I(008)_{2212} + I(0010)_{2223}]$$

showed almost 100% conversion to 2223. The tape that was rolled to an $80 \mu\text{m}$ filament thickness before 2212 formation (Tape D) had an initial texture of $F_{2212} \approx 0.35$, which increased to $F_{2223} \approx 0.75$ upon reaction to 2223. In this sample, the reaction to 2223 was only 83% complete after 250 hours at 825°C .

IV. DISCUSSION

The results presented here show three main points: 1) tapes containing 2212 reach a maximum texture of about $F_{2212} \approx 0.7$ during rolling, 2) the 2223 texture is substantially better than the prior 2212 texture, and 3) better-textured 2212 produces better-textured 2223. For example, the pre-reacted composite (Tape A) had a texture of $F_{2212} \approx 0.3$ after drawing, which then increased to $F_{2212} \approx 0.7$ at a rolled filament thickness of $\sim 260 \mu\text{m}$ and remained at that level for the remainder of the rolling sequence. The texture increased to $F_{2223} \approx 0.95$ upon reaction to 2223. This is consistent with the

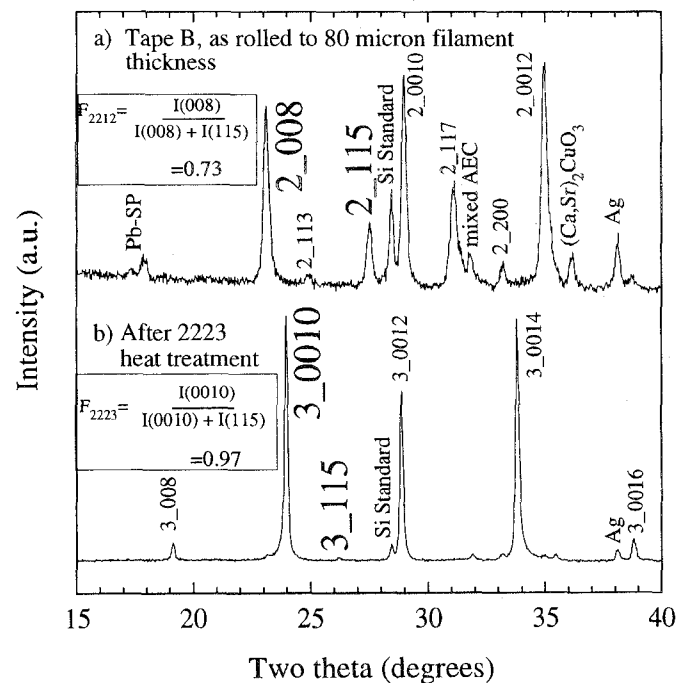


Fig. 4. XRD patterns of an $80 \mu\text{m}$ thick filament Tape B sample showing a) the 2212 texture before heat treatment, and b) the subsequent 2223 texture. The texture increased upon conversion to the 2223 phase.

results of Grasso, Perin, and Flükiger [6], who reported that a maximum 2212 texture is reached once a tape has been rolled to a filament thickness of $\sim 50 \mu\text{m}$.

The results give some insight into the contributions of drawing and rolling to the texture. At a filament thickness of $\sim 700 \mu\text{m}$, the pre-reacted (Tape A) samples had a 2212 texture of $F_{2212} \approx 0.3$ while the reacted after drawing (Tape B) sample had a texture of $F_{2212} \approx 0.2$. The pre-reacted (Tape A), reacted-after-drawing (Tape B), and partially-rolled-then-reacted (Tape C) samples all reached a maximum 2212 texture of $F_{2212} \approx 0.7$ (Figure 3). This indicates that: 1) rolling is more effective at increasing the texture than drawing, 2) the maximum 2212 texturing benefit from rolling can be obtained relatively quickly, and 3) it is not necessary to react the starting powders to 2212 before deformation to obtain the highest texture. Thus, a tape needs only to be rolled through a true strain of greater than ~ 1.5 after reaction to 2212 in order to assure that the 2212 phase develops maximum texture. This information may be useful for making wires with unsausaged fine filaments using powders with more isotropic properties in place of the anisotropic 2212.

From Table II it is clear that the texture of the 2223 phase is strongly influenced by the texture of the precursor 2212 phase. The effect on the texture can be partially attributed to the 2223 formation mechanism. As discussed earlier, if 2223 forms purely by intercalation, the grains should inherit the same crystallographic orientation as the 2212 phase. By contrast, if the 2223 phase forms by a nucleation and growth process from the liquid, one might expect a decrease in texture, because the 2223 grains should nucleate and grow with a more random orientation [14]. However, because the 2223 grains are deposited epitaxially on the 2212 precursor [19], it is probable that the 2223 grains would have some preferential orientation with respect to the 2212 grains, whichever of the above cases is valid. Our results do not provide direct evidence for either hypothesis, as 2223 formed with greater texture than 2212. The explanation may be provided by Wang et al. [13], who recently reported that 2212 is textured by a liquid phase during the 2223 reaction. However, it should be also noted that the increase in texture could be the result of constrained growth texturing [2],[3],[6], by which grains become textured during reaction to 2223 because they are geometrically constrained by the silver sheath or other aligned grains.

As a final note, we found that poor texture results in less conversion to 2223, as evidenced by the Tape D composite, which was only $\sim 83\%$ converted to 2223 after 250 hours of reaction. In a poorly textured and thus less dense tape, there is a lower contact area between grains. Because the grains are not in intimate contact, diffusion is slowed, thereby impeding conversion to 2223 [20].

V. CONCLUSIONS

The effect of wire drawing and cold rolling on the development of the precursor 2212 phase and subsequent 2223 texture in Ag-clad tapes was studied using x-ray diffraction. It was found that the 2212 texture increases to a maximum of $F_{2212} \approx 0.7$ during cold rolling through a true strain of about 1.5 after the reaction to 2212. We found that rolling has a much greater effect on the texture than does wire drawing. Samples reacted to 2212 before the end of the mechanical deformation sequence all had a texture of $F_{2212} \approx 0.7$ at a filament thickness of $\sim 80 \mu\text{m}$, which increased to $F_{2223} \approx 0.95$ after reaction to the 2223 phase. Samples reacted to 2212 after rolling had a texture of $F_{2212} \approx 0.35$, which lead to a 2223 texture of only $F_{2223} \approx 0.75$. These results show that the 2223 texture is determined by both the precursor 2212 texture and reaction-induced texturing, and is consistent with a model whereby 2223 grains form by intercalation of 2212 grains that have had their texture improved early in the reaction by rotation in a liquid phase, and also undergo some reaction-induced (constrained growth) texturing.

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