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Origin of subgrain structure within a domain in melt-processed $YBa_2Cu_3O_7$

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The microstructures of melt-processed $YBa_2Cu_3O_7$ with different cooling rates were studied by polarized light optical microscope. The present study concentrated mainly on the origin of subgrain structure within domains, where there exist controversies in the literature, as due to the polygonization process in the solid state or during the crystallization process. The present study strongly supports the view that subgrains result from the solidification instabilities during crystallization from the melt rather than due to the polygonization process in the solid state. [S0163-1829(97)05122-9]

INTRODUCTION

Melt processing of $YBa_2Cu_3O_7$(123) superconductors has emerged as a unique technique to attain high critical current densities in the material.[1–4] Although it is universally accepted that melt-processed microstructure results in high critical current densities (J_cs), there still exist controversies on the origin of some of the microstructural features, including the origin and existence of subgrain structure within domains.[5–9]

Broadly, melt processes can be divided into two: directional and nondirectional processes.[10] In the nondirectional process a 211+liquid phases mixture is slowly cooled through the peritectic formation temperature of 123. And in the directional solidification, either the sintered 123 is translated through a hot zone or the heater is moved along the length of the sample creating large temperature gradients on the sample. The essential difference in the resulting microstructures obtained by these two processes is that in the first case one sees randomly oriented domains of a few mm^2 in area, containing parallel platelets of 123 with a common orientation, while the directionally solidified samples show no multiple domain formation and the whole sample will be a single domain. The above description is the general microstructural picture reported by many groups.[11,12] If one examines the finer details of the microstructure, there exists some reports in the literature that even a single domain consists of subgrains with small misorientations generally, $<5°$.[5–9] The origin of these subgrains within a domain have been argued by Sandiumenge *et al.*[5,6] as arising from a polygonization process where the elastic stresses of a bent crystal are released. Alternatively, Diko *et al.*[7–9] have hypothesized that instead of a polygonization mechanism, subgrains result during the crystallization process itself due to some solidification instabilities. And also in another observation Sandiumenge *et al.*[5] have discussed that samples with higher 211 concentration displayed polygonization whereas samples with lower 211 concentration do not show this phenomena. As there exists controversies in the literature, we have investigated the origin of the subgrain structure within a domain in melt-processed 123.

Since in the melt-textured material, $J_c's$ of an order of magnitude less than that of thin films and single crystals are attributed for the existence of subgrains due to polygonization,[6] it is very important to know the origin of the subgrains for a better control and to further improve the J_cs of the material for practical applications. This knowledge is also very important in the context of basic studies, while modeling the microstructural evolution during melt processing of 123.

EXPERIMENTAL

The 123 powder used for melt processing is prepared by a chemical route and subsequent vacuum calcination. The powders are uniaxially pressed into pellets of size 15 mm×15 mm×7 mm under a pressure of 10 tons in a steel die. The pellets are then melt processed in a horizontal tube furnace, which has a temperature accuracy of ±0.5 °C with respect to the set temperature, in ambient atmosphere. There was no measurable temperature gradient either vertically or horizontally within the space reserved for the sample inside the furnace. The heating schedules used for melt processing is shown in Fig. 1. Samples are processed with two different cooling rates of 1 and 10 °C/h from the peritectic temperature. No subsequent oxygen annealing is given to the samples. Slices 1 mm thick were separated from the samples by

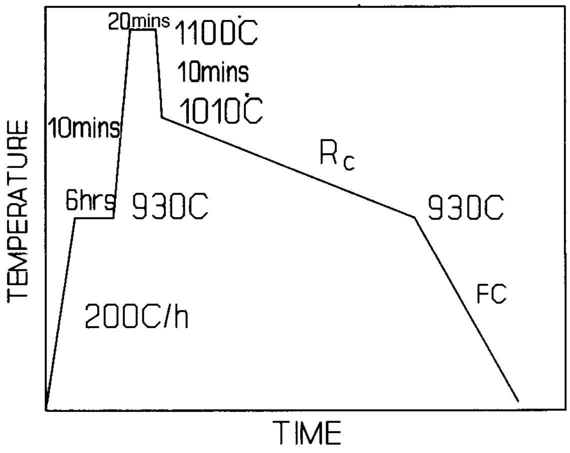

FIG. 1. The time-temperature profile used for melt processing of $YBa_2Cu_3O_7$. The first sample is processed at a cooling rate (R_c) of 1 °C/h from the peritectic temperature and the second sample at a rate of 10 °C/h.

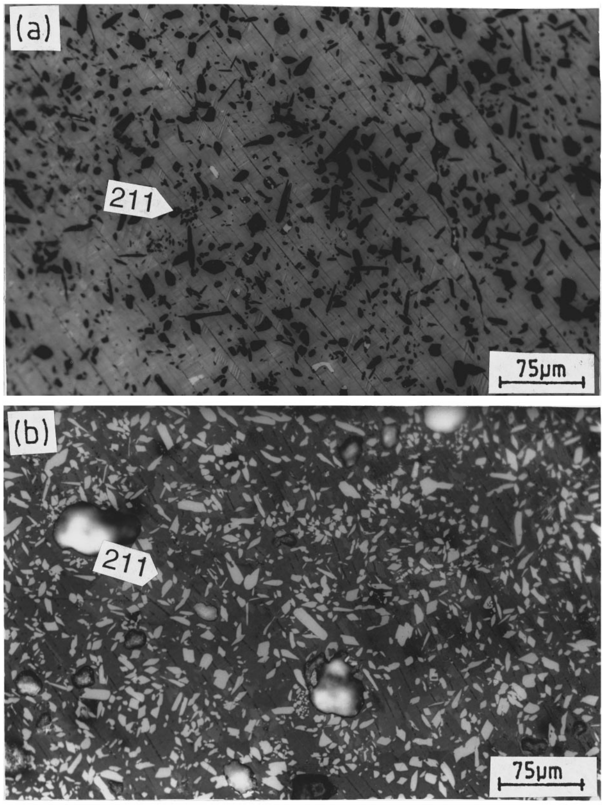

FIG. 2. Optical microstructures taken under polarized light, of melt-processed $YBa_2Cu_3O_7$ samples cooled at a rate of (a) 1 °C/h and (b) 10 °C/h from the peritectic temperature. The pictures are representative regions within a domain of respective samples. Mosaiclike structure (small regions with different shading, which are the regions with slight misorientations) and the bending of the gaps represents the presence of subgrain structure in the case of the 10 °C/h cooled sample. In the 1 °C/h cooled sample, the absence of mosaic structure and the perfect parallel gaps between the platelets running from one end to the other of the microstructure confirms the absence of subgrain structure.

a low speed diamond saw and used for microstructural studies. The specimens for microstructural studies were mounted in a cold setting resin and polished on a fine cloth mounted on the rotating disk of a polishing machine with different grades of diamond paste as polishing media. A Leitz optical microscope equipped with a polarizer was used to investigate the microstructural features of the polished surfaces. No etching was done to the polished surfaces.

RESULTS AND DISCUSSION

As the samples are processed in the absence of a temperature gradient, the resulting microstructure shows differently oriented domains in which platelets of 123 are observed. Figure 2(a) shows the microstructure of a sample processed at a rate of 1 °C/h through the peritectic temperature. The region shown is a portion within a domain and is representative of the microstructure of the sample. It can be seen that no subgrains exists within the domain. The subgrains which are misoriented at small angles generally around 5° are visible at some orientation of the polarizer as a mosaic structure with different shades. The typical size of subgrains reported is around 25–40 μm.[5,7–9] Another way of visualizing the subgrains is to check the alignment of platelets which are separated by small gaps, within a domain. The absolutely parallel gaps without any deflections from one end to the other of the microstructure can be seen in Fig. 2(a). As the typical size of subgrains is around $25\times 40\ \mu\mathrm{m}^2$, if at all any subgrains exists in this sample, an area of around $500\times 350\ \mu\mathrm{m}^2$ should show at least some of them. As there is no indication of any mosaic structure or change in color pattern under polarized light or bending of gaps, we can convincingly conclude that the present sample is free of subgrains.

Figure 2(b) shows the microstructure of 123 melt processed at a rate of 10 °C/h from the peritectic temperature. The microstructure represents a region within a domain and is taken using a polarized light. The slight variations in the shades, i.e., mosaiclike pattern, represents the existence of subgrain structure. And also the bending of gaps clearly represents the existence of slightly misoriented subgrains within a domain in this case. From the observations discussed above, we conclude that cooling rates of 1 °C/h resulted in the absence of subgrain microstructure whereas a cooling rate of 10 °C/h resulted in subgrains within a domain.

Another parameter of interest is that the composition of the starting precursors is the same in both cases and also the average size and volume percentage of 211 after melt processing is the same. There is no ambiguity in the present case regarding the difference in the volume percent of 211 in the resulting material, whereas Sandiumenge *et al.*[5] have reported that the composition with a lower percent of 211 does not show polygonization whereas samples with a larger 211 content exhibit polygonization.

The existence of subgrain microstructure so far reported in the literature[5–9] has been seen either in directionally solidified samples or in the case of samples processed in the presence of magnetic fields. In the directionally solidified processes, where the processing conditions are complicated, including high temperature gradients, and the movement of samples through these gradients (which are important in determining the morphology and the orientation of the growth front of 123), there is a chance for some perturbations in the processing conditions leading to the solidification instabilities. Processing in the presence of magnetic fields, which makes the processing parameters even more complicated, may also induce instabilities to the solidification front.

In the present study, in the case of very low cooling rates of 1 °C/h, which represents good equilibrium conditions, one can assume that there exist no solidification instabilities and hence results in the absence of subgrain structure. Increasing the processing speeds to 10 °C/h results in solidification instabilities during the crystallization process from the melt and hence results in the subgrain microstructure, as observed by others[5–9] where the processing conditions favored solidification instabilities. Hence we conclude that the existence of subgrain structure is not an intrinsic property of the melt-processed microstructure and exists in a material where the processing conditions favor the solidification instabilities and are not a result of the polygonization mechanism in the solid state.

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