Studies on structural and magnetic properties of melt textured growth YBa$_2$Cu$_3$O$_{7-x}$ superconducting bulk materials

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Abstract. In this study, production methods and the structural and magnetic properties of melt-textured growth of YBa$_2$Cu$_3$O$_{7-x}$ bulk superconductors are discussed. During the research; various temperatures for melt texturing are tried for the optimization of the production of bulk YBCO. With the help of AC magnetic susceptibility measurements, some insights for the produced samples are handed out including critical phase transition temperatures and the effects of crystal growth inside the YBCO samples. XRD measurements showed bigger and greater number of crystal growth in a correlation with the increasing temperature and the microphotographs of the samples verified successive growth of YBCO crystals inside the samples. Sharper phase transition is reached with the increasing texturing temperature. Top seeding for the selected crystal orientation has also been tried at the 1070°C for the texturing temperature, but the resulting melt textured sample was not different with the only-textured sample in the means of magnetic properties. Optimization over the texturing temperature found out to be between 1070°C and 1080°C while this result only depends on magnetic and structural properties.

Introduction

Developments on the high temperature superconductivity have taken a great advance over the last 20 years. The discovery of the high-Tc oxide superconductor, named as Y-Ba-Cu-O system [1] brought a great deal on the area of superconductivity. These new type of superconductors have many new possibilities which are useful to design them in desired shapes and contain useful mechanical and electromagnetic properties. Producing this type of superconductors and optimization of the fabrication conditions have been a great concern and been overcome by several approaches. Flywheel energy storage systems [2-3], benefiting magnetic levitation properties [4] and a good value of critical current density are all among the required properties of the bulk samples. However, the quality of the bulks depend on many factors; including grain size, crystal orientation and the other properties such as secondary phases, homogeneity of the sample and microcracks have critical effects. In this case; obtaining good quality YBCO bulk superconductors is at most importance. One of the important and useful approaches to fabricate bulk YBCO is the melt-textured growth [5-6].

In this study production method of melt-textured growth has been experimented for obtaining bulk YBCO superconductors. Magnetic properties and structural properties, together with x-ray diffraction and microphotography have been applied to get characterization and optimization of the samples. Peritectic decomposition temperature has been changed to get the difference of crystal growth among the samples while other parameters kept fixed.
Experimental details

Throughout this study the method of Melt-Textured-Growth [7] of YBCO bulks has been experimented. Experimentation consisted of some predefined temperature steps which are crucial to crystallization in the means of peritectic decomposition temperature. These temperatures are selected with the help of previous studies [8-9], so that resulting YBCO bulks expected to differ in the means of crystal sizes, crystal orientation and samples’ magnetic properties. These samples differed among their decomposition temperature followed as in this order: 1030 °C, 1050 °C, 1070 °C, 1080 °C, 1090 °C. Other variables; such as size, mass and the homogeneity of the samples, thermal cycle with further oxygenation at which melt-textured growth has taken place are tried to keep fixed while the decomposition temperature is changed.

All samples are prepared according to the method of solid state reaction by required chemicals which are weighted by a 4-digit weightier in stoichiometric proportions and mixed for a suitable time for getting homogenous mix. This mixture then subjected to calcination process which is required to vaporize CO₂ from the mixture. After the calcination, mixture is grounded, mixed homogenously and pelleted to get disks of 8mm in diameter and 1mm in thickness which will be used in sintering process. Sintering process is performed at 920°C under air atmosphere for all samples for a suitable time period. Sintering process is required for getting a good melted and homogenous mixture. These disks then called as ‘sintered samples’, are then ready for Melt-Textured-Growth. For all the thermal cycles, a Euroterm horizontal tubular furnace is used. Sintered samples are subjected to a thermal gradient via entering them into the hot zone of the furnace with a speed of 3cm/min at the peritectic decomposition temperature. At this temperature two phases of Y211 and Y123 are melted and mixed so that a well mixed liquid phase is reached. After this temperature plateau, samples are subjected to a slow cooling rate at which the first Y123 crystal seeding starts. This cooling ends at the temperature of 980°C and samples are subjected to this temperature for a long period so that the stabilization of the growth crystals and further crystal formation are enhanced. After this dwell, samples are cooled to the room temperature. Further oxygenation at 600 °C is needed in case of getting orthorhombic structure which supplies property of superconductivity.

Results and Discussion
Numerous microphotography and XRD measurements are performed to engage into the inner structure of the samples. Due to the superconductive properties of the YBCO crystal form, a predefined and good growth of layers should be provided during the crystal growth. This layer formation consists of a growth along the c-axis and also a periodicity along the a-b layer. Also the texturing of the samples which is the main goal of this study should be significant with the peaks along the c-axis.

XRD measurements are performed with an X’pert Pro Cu-Kα X-ray diffractometer and results are tabulated as a graph of intensity versus 2θ. Intensity peaks are expected to show well developed Y123 crystal and Y211 phase signals along the a-b plane with c-axis between the angles of 20° to 60°. The first sample is shown at Fig. 1 which has crystal growth temperature of 1030 °C shows numerous intensities from all degrees which are close to each other and many peaks are mixed in the means of signal competence. There are some peaks for the growth along the (00l) but none of them reflect strong peaks for the c-axis growth. Also the other peaks of a-b plane are also mixed and no strong signal is recorded. There are also (040)’ and (151)’ peaks which may result from Y211 phase. This situation shows a bad crystal growth or no crystal growth at all. On the other hand, the sample having crystal growth temperature of 1050 °C shows many strong and clear peaks for the c-axis growth as seen in Fig. 2. Characteristic orthorhombic structure peaks [10] of (103), (013), (213), and (116) are also seen in the diffraction pattern. Growth along the a-b plane can also be seen providing the periodic crystal structure. There is also a low melting point phase signal as (002)’ peak of CuO. The sample with crystal growth temperature of 1070 °C as seen in Fig. 3 shows some (00l) peaks along the c-axis. Intensity data is similar to the previous one in the means of peak positions and coherence of characteristic orthorhombic structure peaks. Intensities are clean and strong so that there has been a good crystal growth in c-axis.
Figure 1. XRD pattern of the sample with crystal growth temperature of 1030 °C

Figure 2. XRD pattern of the sample with crystal growth temperature of 1050 °C
Samples are polished by using diamond paste and microphotographs are taken in a few different magnifications with a polarized light microscope. As stated by previous studies [11], melt-textured growth induces non-uniform heating. Sample’s surfaces mostly reach maximum temperature during the thermal cycle and also the interior may not reach the surface temperature above the peritectic decomposition. The microstructure and phase distribution may be effected with this non-homogenous heating and resulting structure may contain many cracks. Corollary, all samples in this study regardless of their crystal growth temperature show highly porous structure with cracks and many miss aligned plate-like structures. In figure 4, the sample with crystal growth temperature of 1070 °C can be seen in high magnification. There seems to be 3 different planar regions; the region at the right bottom corner of the image and the others are the upper top corner of the image and the regions of white color. These color difference may be resulted by the reflection of the light with different angles from the a-b plane periodicity of Y123 phase but also there seems some low melting phase regions which have reflections in bluish color. Regions which are between 50 µm to 100 µm show many cracks in various directions. They may be resulted with the further oxygen annealing and with the temperature differences [11]. Figure 5 show the image of the sample with crystal growth temperature of 1030 °C. Porous structure is visible and also no sign periodicity in a-b plane. Figure 6 shows the image of the sample with crystal growth temperature of 1050 °C. There seems to be different planes again with pores and cracks. Sample may have a-b plane growth for the periodicity in different regions and these regions seem to be at around 500 µm in size.
Figure 4. Microphotograph of the sample with crystal growth temperature of 1070 °C

Figure 5. Microphotograph of the sample with crystal growth temperature of 1030 °C
AC magnetic susceptibility measurements are mostly used for qualifying samples for the superconductivity. Since the material expresses perfect diamagnetism property of shielding the entire applied magnetic field and exhibiting the susceptibility of exact -1 in SI units, this property can be used to determine the onset temperature of the critical transition and its temperature [12]. AC magnetic susceptibility consists of two parts of real and imaginary. Real part of AC magnetic susceptibility is in phase with the applied field and represents superconductivity transition from normal conductivity. This part also contains the information of inter and intra granular interactions of the crystalline structure of the sample [13-14]. The first peak like bending part occurs at a temperature slightly below the critical transition temperature indicating the first response of the sample to the applied field as vortices [15]. This response is called as inter-granular interaction at which the superconductive grains of the crystalline structure behave as small cylinders and start to trap the applied field. The second response of the sample occurs at a temperature lower than the first interaction, and called as intra-granular interaction. At this stage, the motion of trapped magnetic flux creates a Josephson coupling [16] and shows itself as a bend-like formation in the real part of the AC susceptibility. On the other hand, the imaginary part of the AC susceptibility reflects the magnetic loss of from the applied field to establish the ordered state of the superconductivity.

AC magnetic susceptibility measurements of the samples are performed by a lock-in magnetic field supplier of SR530 Lock-in Amplifier under vacuum atmosphere with Keithley 224 Current source and Keithley 196 System DMM nanovoltmeter. Results can bee seen at Fig. 3. These measurements are performed by using liquid nitrogen as a cooler and samples are subjected to AC magnetic field of three different magnetic field values.
means of temperature as expected. Also at the corresponding imaginary parts, a decrease in the temperature of the peak and a general widening of the peaks are indicated which are predicted by the bean critical state model [17]. In figure 8, it's clearly seen that intra-inter granular interactions took place for the sample of 1030 °C and this interaction can also be seen at the figure 9 for the sample with crystal growth temperature of 1090 °C. But these measurements are not sufficient to correctly interpret the magnetic response of the samples in the means of flux dynamics. On the other hand, measurements show that at all the applied field values, samples with crystal growth temperature of 1070 °C and 1080 °C have the higher onset transition temperature and also narrow transition range, which may state that the optimization may took place between 1070 °C and 1080 °C. Figure 9, which is the measurement with the highest applied field value, this optimization is clearly indicated. Real part of the magnetic response of all the samples except 1030 °C took place between the response of the 1070 °C and 1080 °C indicating that optimization may be in that range.

Figure 7. AC Magnetic Susceptibility Measurement of all the samples at applied magnetic field of $H_a = 18.5$ A/m.
Figure 8. AC Magnetic Susceptibility Measurement of all the samples at applied magnetic field of $H_a = 185$ A/m.

Figure 9. AC Magnetic Susceptibility Measurement of all the samples at applied magnetic field of $H_a = 4000$ A/m.
4. Conclusion

Through this study, an optimisation over the peritectic decomposition temperature of the bulk YBCO has been experimented by using melt-textured growth method. XRD results and microphotography of the samples show a linear correlation with higher peritectic temperature as increasing $a$-$b$ plane periodicity and $c$-axis alignment. AC magnetic susceptibility measurements reflected a narrow superconductive transition temperature window for the samples of higher peritectic temperature. These results show the importance of the slow cooling rate part of the MTG cycle. Higher the peritectic temperature resulted with more time for the crystal formation with fixed cooling rate. A more homogenic mixture of the YBCO may supply the possibility of less porous structure and also less temperature differences during the cycles may result with fewer cracks in the samples. However, MTG cycle resulted with crystal formation in various samples respectively.

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6. References

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