Large multiseeded domains of (Y,RE)BaCuO obtained by the MUSLE technique

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Abstract. The multiseeded seamless bulk technique (MUSLE) developed by Sawamura et al for Dy/Gd/Ho-BaCuO compositions have been adapted for Y-based superconductors. This method allows overcoming the major problem encountered with the multiseeding technique, which is the trapping of non superconducting phases between the different domains. The MUSLE process requires the choice of two compositions for the bi-layer system. The criterion is a sufficient difference in the peritectic decomposition temperatures. In this condition, the higher peritectic temperature layer serves as nucleating site for the final YRE123 domain. Differential thermal analyses have been performed to select suitable compositions of as the upper layer and YYb123 as the lower layer. The influence of (Y, RE)2BaCuO5 (YRE211) composition on the crystal growth rate and on the superconducting properties was also investigated. Finally, compositions and thermal cycle have been defined to texture 2 cm multiseeded bi-layers samples. Electrical properties, coupled with SEM observations reveal the sample quality.

1. Introduction
The top seeding method is used to texture large samples of REBa2Cu3Oy (RE = rare earth, REBCO). It is known that the sample size is limited with this technique. Indeed, the texture quality and the material homogeneity depend on the position from the seed location. Domains larger than 4-5 cm often present large inhomogeneities between the centre and the edges [1].

To overcome this problem, several techniques have been developed. Among them, welding has given good and promising results, although it was only applied to a few numbers of samples or small samples. This technique is based on the difference in peritectic decomposition temperatures between the welding agent and the samples to weld. Generally, rare earth combinations or silver addition are used for this purpose.

The multi seeding method presents the major advantage of significantly shorten the processing time with the number of used seeds. REBCO crystal growth being really slow, this parameter is of great importance. However, growth fronts always push agglomerations of non superconducting phases. When the domains issued from the several seeds encounter, non superconducting phases are trapped in between. No large single domain samples have hence been obtained with this technique.

Recently, Sawamura et al [1] have presented a new method called multi seeded seamless bulk (MUSLE) technique. It combines the two previously mentioned techniques: difference in decomposition temperatures and multiseeding. It consists in using two layers of different compositions. The criterion is a sufficient difference in recrystallisation temperature of the two layers, that allows fully texturing the thin upper layer from several well orientated seeds before beginning the growth of the lower layer. In this configuration, the upper layer can be considered as several very large seeds joining each other. The growth of the lower layer only proceeds first by epitaxial growth and then c-axis growth, avoiding the trapping of secondary phases between the domains grown from the different seeds.

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Sawamura et al have applied this method to Dy,Gd and Ho based REBCO compounds. The aim of this work is to adapt this process to more known Y based compositions. Hence, a study of the suitable two layers composition was performed. Multiseeded samples were successfully obtained and characterised by microscopy and transport measurements.

2. Experimental details
First of all, a composition study was performed to choose the composition for the two layers. These compositions should have the melting temperatures of interest but also, correct superconducting properties for what concerns the lower layer. Their characteristics are investigated in the first part. Then the MUSLE technique was applied using the results.

2.1. Composition study
Different \((Y_{0.75}RE_{0.25})Ba_2Cu_3O_y\) (YRE123) compositions were investigated for the choice of the upper and lower layer compositions. Stoichiometric proportion of \(Y_2O_3\), \(RE_2O_3\), \(BaCO_3\) and \(CuO\) with \(RE=Sm, Eu, Gd, Dy, Yb\) were mixed and calcined 3 times at 920°C with intermediate grinding. Phases’ purity was checked by X ray diffraction. Differential thermal analyses were performed to determine the melting temperature (Labsys, Setaram, 2°/min).

\((Y_{0.75}RE_{0.25})BaCuO_5\) (YRE211) with \(RE = Eu\) and \(Yb\) were obtained by mixing of the oxides and carbonates and calcination of 12 hours at 950°C. Attritions of these powders as well as of \(Y_2BaCuO_5\) (Y211) commercial powder were then performed during 2 hours in alcohol and in Yttria stabilized Zirconia balls for reducing the particles size.

\(YRE123+0.25\) mol \(YRE211\) or \(Y211+0.5wt\%CeO_2\) were mixed by dry ball milling. Recrystallisation temperatures as well as growth rates in defined temperature windows were evaluated by quenching experiments. Samples were therefore heated at 1045°C during 3 hours and cooled at 1°C/h to the quenching temperature.

For characterization of the superconducting properties at 77K, cleaved samples were annealed during 150 hours at 430°C. These properties were measured using DC SQUID magnetometer and critical current density \(J_c\) was calculated with the modified Bean model [2].

2.2. MUSLE textured samples
Using the first part characterisations, the thermal cycle was defined and 2-seeded samples were obtained. For preparation of the samples, the two layers are pressed separately to avoid problems linked with different shrinkage rate between the two compositions. The upper layer (UL) thickness is in the range 1.5 to 2 mm. The two Sm123 seeds were placed with 100/100 orientation on the UL. The samples were characterised through optical microscopy (Olympus) and SEM observations coupled with EDS analyses (Philips FEG XL30). Transport measurements were also realized in continuous increasing current (10 A/s) with a criterion of 10 µV/cm for evaluating the \(J_c\) through the whole sample at 77K and in self field.

3. Results

3.1. Composition study
Table 1 gives the results of the DTA analyses. The difference in melting temperatures between the YSm123/YEu123 and the YYb123 is 24-27°C, which should be highly sufficient according to [2]. These compositions can hence be chosen as respectively upper and lower layers’ ones (UL and LL). In order to avoid a reaction of the UL with the seed, and since the YSm123 and YEu123 melting temperatures are really close to one another, the YEu123 composition is preferred to YSm123.

| Composition | YSm123     | YEu123     | YGd123     | YDy123     | YYb123     |
|-------------|-------------|-------------|-------------|-------------|-------------|
| Melting temperature | 1035°C      | 1032°C      | 1026°C      | 1013°C      | 1008°C      |

Table 1 : Melting temperatures of several YRE123 compositions determined by DTA.
In fact, what is really important is not so much the difference in melting temperatures between the UL and LL compositions but in recrystallisation temperatures. The solidification of REBCO is characterized by two temperatures, which are the temperature of nucleation under the seed and the temperature of spontaneous nucleation. These temperatures define the solidification window. For the UL to act as a seed for the LL, the UL must be completely textured before the beginning of the recrystallisation of the LL. Hence, the best way is to have well shifted solidification windows for the two compositions. In this sense, diffusion of Yb in the UL and Eu in the LL must also be controlled. It should be noticed that the addition of silver only to the LL to shift down its solidification window is not effective. Silver diffuses in large quantities in the UL so that there is no more sufficient difference in solidification windows.

The solidification windows of YEu123+YEu211+CeO2 (YEu), YYb123+YYb211+CeO2 (YYb) and YYb123+Y211+CeO2 (YYbY) were evaluated. To take into account the eventual diffusion, the quenching experiments were always carried out with the bi-layers configuration (i.e. YEu on YYb or YYbY). Results are given in table 2. It could be seen that solidification windows of the UL and LL compositions are really separated. Average growth rates were evaluated in these windows. As the UL is large but thin, the important parameter is a or b direction growth rate. For the LL, as the growth proceeds from the full UL surface, the c direction growth rate is the interesting parameter.

| Composition                  | Solidification window | Average growth rates               |
|------------------------------|-----------------------|-----------------------------------|
| YEu123+YEu211+CeO2 (YEu)    | 990-985°C             | 0.2-0.3 mm/h along a or b direction |
| YYb123+YYb211+CeO2 (YYb)    | 961-959°C             | 0.7 mm/h along c direction         |
| YYb123+Y211+CeO2 (YYbY)     | 962-958°C             | 0.8 mm/h along c direction         |

Table 2: Solidification windows and growth rates of the three selected compositions.

It should be noticed that YEu growth rates are really low, despite attrition of YEu211.

Figure 1 illustrates typical microstructure of the YYb and YYbY compositions. Y211 particles appear smaller than YYb211 particles. Moreover, BSE images show different contrasts in some of the Y211 particles of the YYbY sample: the core is darker than the edges (figure 1c). In fact, EDS analyses performed on these zones show that brighter regions correspond to Yb richer areas and darker ones to rich Y even pure regions. This means that during the peritectic decomposition of YYb123 or during the high temperature dwell, Y211 have grown with an YYb solid solution. This contrast is also observed, although less pronounced on the YYb211 particles of the YYb sample. In this case, the core of the particles is brighter. Whatever the case, EDS analyses show that the Y/Yb ratio is not homogeneous from one RE211 particle to another.

Finally superconducting properties of YYb and YYbY compositions were evaluated by magnetic measurements (figure 2). Both compositions exhibit an onset critical temperature of 91K with transition width of 1-1.5K. Critical current density is higher for the YYbY composition, as can be awaited from the observations on RE211 size. The value obtained in this case is similar to what is obtained classically for pure Y123 with cerium doping that is to say 50 kA/cm² in self field and at 77K [3].

![Figure 1: BSE-SEM microstructures of (a) YYb, (b) and (c) YYbY. (a) and (b) have the same scale.](image-url)
Therefore, the compositions chosen for testing the MUSLE technique with Y based compounds are YEu for UL and YYbY for LL.

![Graph showing magnetic field vs Jc](image)

Figure 2: Superconducting properties of textured YYb and YYbY compounds.

3.3 MUSLE textured samples.

Figure 3a shows an optical photograph of the cross section at the limit between 2 domains issued from 2 seeds. In the UL, a crack separates the domains, as usually seen in multiseeded samples. However, it is clear that this crack continues on only 200 µm in the LL. Only one single domain can be observed under this point. Figure 3b shows a BSE image of the limit between the UL and LL. The 200 µm thick area, located just under the UL, corresponds to the thickness of diffusion of Eu in the LL. No Yb has been detected in the UL.

Transport measurements were performed on bars extracted from the LL parallel to the ab plane and crossing the two domains. They reveal a critical current density of 12 kA/cm$^2$ in self field at 77K. Several measurements on the same sample have not broken it.

![Image showing cross section](image)

Figure 3 : (a) optical photograph of the cross section at the limit between two domains and (b) BSE image of the cross section.

4. Conclusion

Y based compositions have been studied for MUSLE technique. (Y$_{0.75}$Eu$_{0.25}$)Ba$_2$Cu$_3$O$_y$ and (Y$_{0.75}$Yb$_{0.25}$)Ba$_2$Cu$_3$O$_y$ have been chosen for respectively the upper layer and the lower layer. It is shown that this method is efficient in obtaining one single grain from 2 seeds located on the top of the upper layer. Transport measurement has shown that 12kA/cm$^2$ can flow through the sample, illustrating the quality of the single domain.

References

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