Flux Pinning Enhancement in $(Y_{0.5}Nd_{0.25}Sm_{0.25})Ba_2Cu_3O_y$ Oxides by Zone Melt Growth Process

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Abstract - Directionally melt-textured high T_c $(Y_{0.5}Nd_{0.25}Sm_{0.25})Ba_2Cu_3O_y$ [(YNS)-123] superconductor was systematically investigated by the zone melt growth process in air. A sample prepared by this method showed well-textured microstructure, and $(Y_{0.5}Nd_{0.25}Sm_{0.25})_2BaCuO_5$[(YNS)211] inclusions were uniformly dispersed in large $(Y_{0.5}Nd_{0.25}Sm_{0.25})Ba_2Cu_3O_y$ [(YNS)123] matrix. High irreversibility field and magnetization hysteresis loop of the zone melt-textured (YNS)-123 sample exhibited the enhanced flux pinning, compared with $YBa_2Cu_3O_y$ (Y-123) sample without RE(rare earth). Critical current density of (YNS)-123 sample was 2.5×10^4 A/cm^2 at 2 T and 77 K.

Keywords: $(Y_{0.5}Nd_{0.25}Sm_{0.25})Ba_2Cu_3O_y$, Zone melt growth, $(Y_{0.5}Nd_{0.25}Sm_{0.25})_2BaCuO_5$, Flux pinning, Irreversibility field.

1. Introduction

Large single-domain RE-Ba-Cu-O(RE: Nd, Sm, Eu, Gd) superconductors are promising materials for cryogenic applications since they demonstrate superior superconducting performance to Y-Ba-Cu-O bulks. Large samples of melt-grown RE-Ba-Cu-O also show superior trapped field performances as the consequence of the high J_c, making these materials as attractive candidates for applications such as quasi-permanent magnets, flywheels, magnetic separation systems and magnetron sputtering systems. In order to achieve a large critical current density in a magnetic field, it is necessary to introduce strong pinning centers into the superconductors. Recent advances in melt-growth techniques enabled the fabrication of large grain RE-Ba-Cu-O bulk superconductors with high current-carrying capability through the enhancement of flux pinning and the reduction of weak links. The J_c of RE-Ba-Cu-O bulk material is strongly dependent on the microstructure of a superconducting grain. It is well known that RE211 particles trapped in the melt-textured $REBa_2Cu_3O_y$(RE123) superconducting phase and related defects act as strong pinning centers. In the melt growth processed $REBa_2Cu_3O_y$ bulk, the pinning provided by the nonsuperconducting particles of the RE_2BaCuO_x(RE211) phase is mainly effective at low magnetic fields and high temperature above 77 K. In this case, the interfaces between RE123 and RE211 play an important role in the flux pinning, because the size of RE211 is much larger than the coherence length[1]. Muralidhar, et al. recently reported that RE-123 systems where two or three rare-earth elements $(Nd_{0.33}Eu_{0.33}Gd_{0.33})Ba_2Cu_3O_y$ (NEG123) are mixed can show very large values of critical current densities(J_c), when they are processed oxygen controlled melt growth(OCMG) technique[2,3]. A requirement for obtaining high critical current densities in bulk RE-Ba-Cu-O is to have a textured microstructure so that the high J_c directions are aligned in the direction of current travel. Zone melt growth process is a good fabrication method to eliminate the weak links and obtain large grain with good alignment so as to increase critical current density[4-7]. In the zone melt growth process, as there is no contact between the liquid and the support, these liquid losses are avoided. So, this process is an efficient way for processing in air RE-123 bars with a high and sharp superconducting transition. Three parameters are important in the zone melt growth process: the maximal temperature(T_m), the thermal gradient(G) and the growth rate(R). In this study it was decided to keep constant G and R which allow to obtain large single grains. The aim of this paper is to fabricate directionally single-grain (YNS)-123 systems with enhanced superconducting properties by the zone melt growth process in air. In this paper, we also report on the analysis of microstructure, magnetization measurement and critical current densities in the melt-textured (YNS)-123 samples prepared by this method.

2. Experimental

High purity commercial powders of Y_2O_3, Nd_2O_3, Sm_2O_3, $BaCO_3$, and CuO powders were weighed and mixed in appropriate amounts to produce a nominal

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composition of $(Y_{0.5}Nd_{0.25}Sm_{0.25})Ba_2Cu_3O_y$[(YNS)-123]. Precursor powders were thoroughly ground and calcined at 900°C for 30 h twice in air with intermediate grinding. It is known that in Y-123 and RE-123 systems, the flux pinning properties are improved by the presence of small Y_2BaCuO_5(Y211) or RE_2BaCuO_5(RE211) particles dispersed in the superconducting matrix[1,8,9]. In analogy, in the whole sample 30 mol% of $(Y_{0.25}Nd_{0.25}Sm_{0.25})_2BaCuO_5$ [(YNS)211] phase was added. It was reported that CeO_2 is effective for increasing the viscosity of the Ba-Cu-O melts and can act as a growth inhibitor for RE211 particles[10,11]. For these reasons, 1 wt% CeO_2 was added. All these powders[(YNS)-123+(YNS)211+CeO_2] were mixed and milled with attrition for 6 h in acetone using zirconia balls with a rotation speed of 450 rpm. The (YNS)-123 powders milled with attrition were dried in air, and then isostatically pressed at 200 MPa into a small cylindrical type(2 mm in inner diameter, 6 mm in outer diameter and 100 mm in length) using rubber mould. The pre-heated(930°C, 5 h) (YNS)-123 rod samples were processed in a vertical zone melting furnace, applying a thermal gradient(G) close to 200°C/cm. An unidirectional solidification was obtained by moving the sample upward through a hot zone(1100°C) at a rate of about 3 mm/h in air. The zone melt growth apparatus used in this study was shown in Fig. 1. The post-heat treatment was performed at 450°C oxygen gas atmosphere for 150 h in a separate tube furnace. To analyze the effect of RE substitution, another zone melt-textured $YBaBa_2Cu_3O_y$ (Y-123) sample, added 30 mol% of Y211 phase was also fabricated by the same procedure. Each directionally grown sample of (YNS)-123 and Y-123 was cleaved into smaller sections (2~3 mm long) for characterization. The microstructure observations were performed on well polished surfaces of the samples by means of scanning electron microscopy(SEM), transmission electron microscopy(TEM), and high-resolution transmission electron microscopy(HR-TEM). Chemical composition of the matrix and secondary phase was determined by energy dispersive x-ray(EDX) analysis. For analyzing the flux pinning effect, magnetization hysteresis of the samples were measured for H//c-axis in a Quantum Design SQUID magnetometer at various temperature from 10 K to 85 K. The critical current density(J_c) values were estimated based on the extended Bean's model[12] using the following equation $J_c = 20(\Delta M)/[a^2d(b-a/3)]$, where a and b are the cross sectional dimensions(a < b), d is the sample thickness and ΔM is the difference of magnetic moments during increasing and decreasing field processes in the M-H loop.

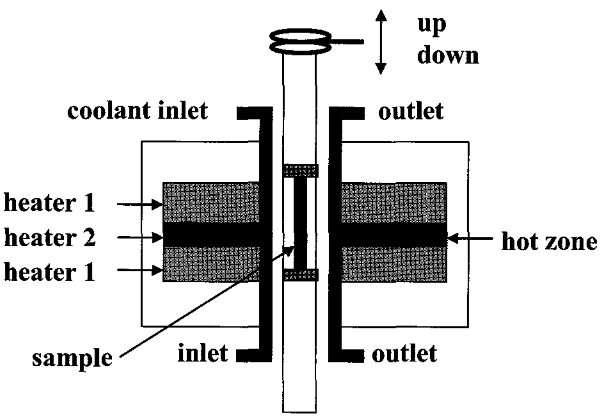

Fig. 1 Schematic illustrations of the experimental apparatus for zone melt growth method

3. Results and Discussion

Microstructure control of high temperature superconductors is the key parameter for successful industrial applications of these materials. In the REBCO bulk system, a large number of crystal defects are present in the RE123 matrix that can act as effective pinning centers. In addition to the crystal defects, fine dispersion of nonsuperconducting RE211 secondary phase particles is known to be effective in enhancing flux pinning at 77 K and large applied fields. The smaller size of these particles, the more effective flux pinning site is on them[2,13]. Fig. 2 shows a SEM micrograph of the fracture cross section transverse to the long axis of the melt-textured (YNS)-123 crystal. In Fig. 2, we can see that the (YNS)211 inclusions are trapped within the (YNS)123 matrix, which have a layered structure(platelets). In order to obtain more detailed

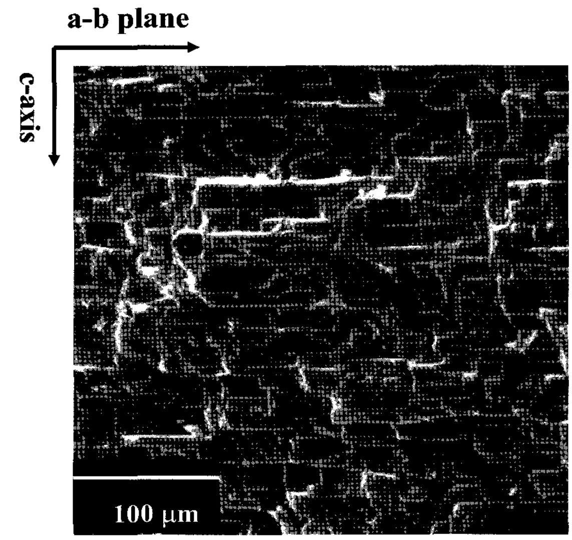

Fig. 2 SEM micrograph of the cross sectional view of the zone melt-textured (YNS)-123 sample. Note dispersed (YSN)211 inclusions are trapped into the (YNS)123 matrix with platelet crystals

information on the (YNS)211 inclusion size and distri-a lot of {110} twins have been observed from the [001] direction of the platelet crystal. As shown in Fig. 3, (YNS)123 matrix and (YNS)211 inclusions are well crystallized, respectively. From the analysis of TEM-EDX, only homogeneous (YNS)123 and (YNS)211 are observed, and impurity phase of RE is not found. bution, we performed transmission electron microscopy (TEM) on the same sample. TEM was performed on the sample annealed at 450°C for 100 h. Fig. 3 shows TEM bright field image around the (YNS)211 inclusion of the (YNS)123 matrix viewed from a [001] direction. In Fig. 3, The 123 and 211 interface is predicted to provide effective flux pinning when the interface is sharp. In Fig. 4, high resolution TEM image really showed clean interfaces between the (YNS)123 and (YNS)211 without any impurity phase. At such a sharp interface, the order parameter rapidly varies over the distance of the coherence length, leading to a large flux pinning force. To increase pinning effect, total area of the interface should be important ; smaller Y211 inclusions are more efficient in enhancing flux pinning[8]. It is reported that the melt-textured Y-123 and RE-123 bulk systems, fabricated by melt-textured growth method, exhibit enhancing flux pinning effect because 211 inclusions dispersed in 123 matrix work as pinning centers[2,9]. The SEM photograph of (YNS)-123 sample in Fig. 2 shows that (YNS)211 inclusions of about ~10 μm^2 are distributed in a large (YNS)123 matrix. Fig. 4 exhibits that the interface between (YNS)211 inclusion and (YNS)123 matrix is sharp. The flux pinning effect in OCMG-processed REBCO bulk sample is larger than that in YBCO, indicating that the superconducting (YNS)123 phase in our (YNS)-123 sample may have enhanced pining effect as well. The sharp interface between the (YNS)123 and (YNS)211 as well as the substitution of RE for the half of Y in (YNS)-123 compound may provide more effective pinning centers than in Y-123 samples.

In order to find out how the sample consists of superconducting phase, magnetic susceptibility of the zone melt-textured (YNS)-123 sample was measured. The field of 1 mT was applied parallel to the c-axis from 10 to 100 K in zero field cooled(ZFC). As shown in Fig. 5, the (YNS)-123 sample shows a sharp superconducting transition at 91 K, indicating that the sample consists of a homogeneous superconducting phase.

For the analysis of flux pinning, magnetization hysteresis loops of (YNS)-123 and Y-123 samples were measured from 10 to 85 K as a function of the magnetic field(//c-axis) up to 5 T. Among the hysteresis results, a representative hysteresis loop at 50 K is shown in Fig. 6. The (YNS)-123 sample shows about 2.5 times larger magne-tization hysteresis loop than that of zone melt-textured Y-123 sample at 2 tesla.

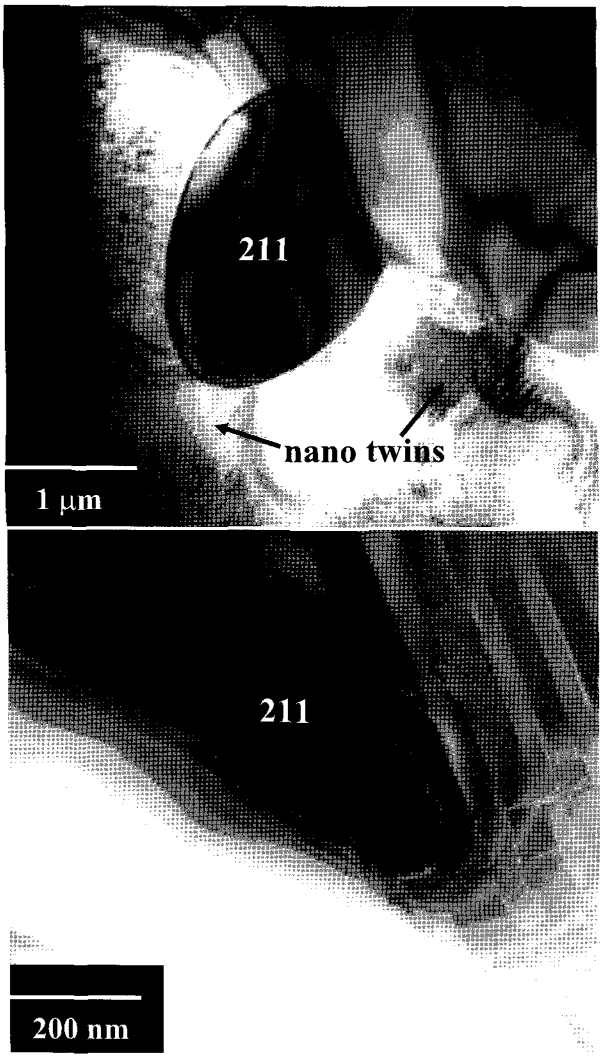

Fig. 3 TEM micrographs of the zone melt-textured (YNS)-123 sample. Sub-micron (YNS)211 inclusions and dense twin patterns are seen in the image

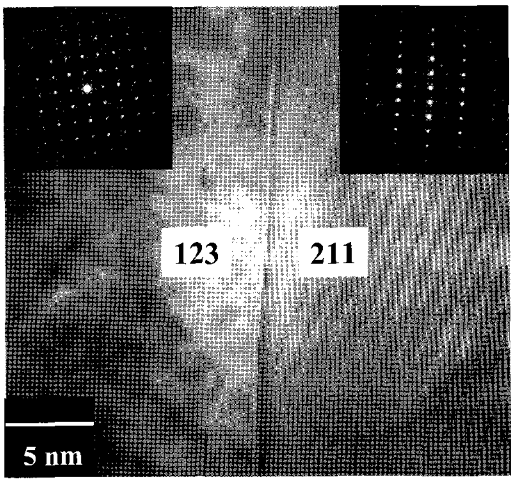

Fig. 4 High resolution TEM image of the interface between a (YNS)211 inclusion and (YNS)123 matrix showing its shape and structure

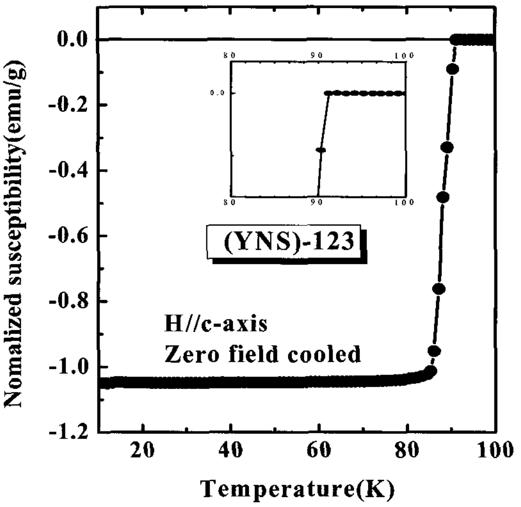

Fig. 5 Temperature dependence of the normalized magnetic susceptibility for the zone melt-textured (YNS)-123 sample

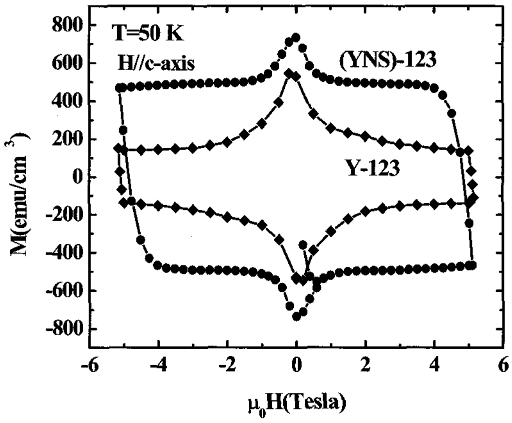

Fig. 6 Magnetization hysteresis loops measured 50 K with magnetic field applied parallel to the c-axis of the zone melt-textured (YNS)-123 and Y-123 sample in air

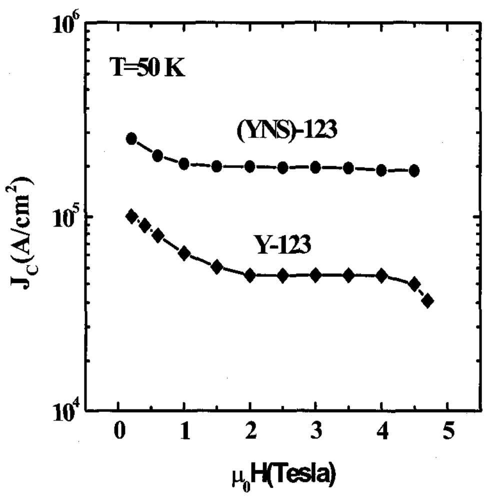

Fig. 7 Field dependence of critical current densities of the zone melt-textured (YNS)-123 sample at 50 K

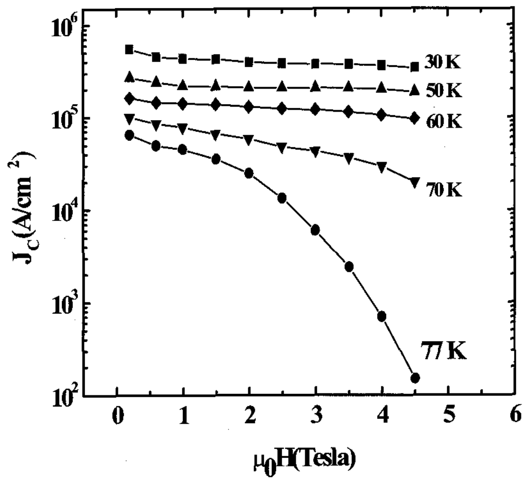

Fig. 8 Field dependence of critical current densities of the zone melt-textured (YNS)-123 sample at various temperatures

The critical current densities of the (YNS)-123 sample derived from the magnetization hysteresis loops at 50 K and various temperatures are shown in Fig. 7 and Fig. 8, respectively. As illustrated in Fig. 8, the field dependence of critical current densities rapidly decreases with increasing of applied fields at 77 K, while it does not change that much at 60 K regions. The critical current density J_c of our sample is calculated at 2 T and 77 K. The value of J_c is 2.5×10^4 A/cm^2, which is higher than that of Y-123 sample.

For a comparison of the pinning strength between different high T_c superconductors, a simultaneous location of their irreversibility lines in the field-temperature(H-T) has been useful. Irreversibility line defines a boundary in the H-T phase diagram above which J_c is immeasurably small or zero. Zero resistance state is only achieved in pinned or irreversible high T_c superconductors, in which flux motion is prevented by pinning centers against the electromagnetic force.

Since high T_c superconductors cannot be used beyond this line, it is desirable that the materials have high enough irreversibility field at a temperature where they are used. It was found that a high irreversibility field B_{irr} over 14 T at 77.3 K was obtained for the NEG123 bulk system with 5 mol% NEG211 inclusions. In this work, the nano-lamellar structure was found in the high B_{irr} NEG123 sample and is the new effective pining center for the improvement of the irreversibility line[2,14]. Therefore, an investigation of the detailed flux pinning characteristics is necessary in order to understand the relation of those microstructures to the critical current densities and the irreversibility field. According to the collective flux creep theory, the irreversibility field B_{irr} is given by $B_{irr} \propto [1-(T/T_c)^m]^n$. A fitting formula has been tried for various exponents m and n. This scaling behavior of $B_{irr} \propto [1-(T/T_c)^2]^{1.5}$ for melt-textured YBCO superconductors was also reported[15]. Several techniques are used in determining the irreversibility field. In this paper, we have adopted the DC magnetization method by SQUID magnetometer.

Fig. 9 shows the irreversibility field(B_{irr}) versus reduced temperature (T/T_c) for the (YNS)-123 sample. In Fig. 8, the irreversibility fields(B_{irr}) of (YNS)-123 sample shows larger value than that of Y-123 sample, indicating that the (YNS)-123 sample may have enhanced flux pining effect as well.

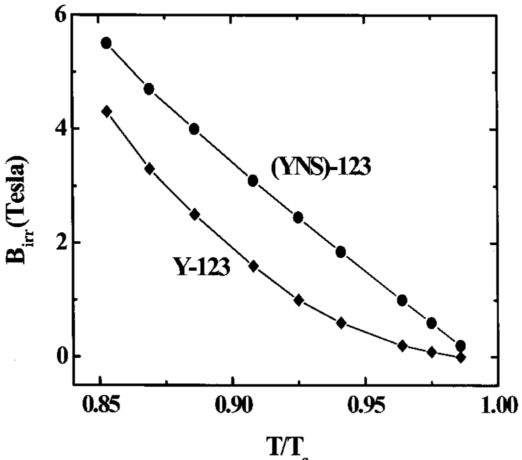

Fig. 9 Irreversibility field with reduced temperature of the zone melt-textured (YNS)-123 sample

4. Conclusions

In this paper, we have succeeded in enhancing pinning effect of directionally melt-textured (YNS)123 superconductor, where (YNS)211 inclusions are uniformly dispersed within (YNS)123 matrix by the zone melt growth process in air. The zone melt- textured (YNS)-123 sample as a sharp transition temperature of 91 K consists of homogeneous superconducting phase. The sample shows a larger hysteresis loop than other Y-123 zone melt-textured superconductors, indicating that flux pinning is effectively increased. The enhancement of flux pinning may be associated with the well-defined interface between (YNS)211 inclusions and (YNS)123 matrix as well as the substitution of RE for the half of Y site in (YNS)-123 compound. This result would be useful for the application fields of the high T_c bulk superconductors that require large critical current even at high magnetic field.

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