

# Grain growth and superconducting properties of melt-processed (Y-Sm-Nd)-Ba-Cu-O composite oxides

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**Abstract**  $(Y_{0.5}Sm_{0.25}Nd_{0.25})Ba_2Cu_3O_y$  [(YSN)-123] high  $T_c$  composite superconductors with  $CeO_2$  addition were systematically investigated by top seeded melt growth (TSMG) process in air atmosphere. A melt textured  $NdBa_2Cu_3O_y$  (Nd-123) single crystal was used as a seed for achieving the c-axis alignment large grains perpendicular to the surface of (YSN)-123 composite oxides. The size of  $(Y_{0.5}Sm_{0.25}Nd_{0.25})_2BaCuO_5$  [(YSN)211] nonsuperconducting inclusions of the melt textured (YSN)-123 samples with  $CeO_2$  addition were remarkably reduced and uniformly distributed within the (YSN)123 superconducting matrix except in the region very close to the Nd-123 seed crystal. The sample showed a sharp superconducting transition of 91 K.

**Key words**  $(Y_{0.5}Sm_{0.25}Nd_{0.25})Ba_2Cu_3O_y$ ,  $NdBa_2Cu_3O_y$ , Single crystal,  $(Y_{0.5}Sm_{0.25}Nd_{0.25})_2BaCuO_5$

## 1. Introduction

High critical current density, even in the presence of external magnetic fields, is required for technological application of superconductors, as magnetic devices, energy storage systems, magnetic bearings, transport systems. The top-seeded melt growth (TSMG) process is widely used to grow well-oriented large single grain  $YBa_2Cu_3O_{7-y}$  (Y-123) and  $REBa_2Cu_3O_y$  (RE-123, RE: Nd, Sm, Gd, etc.) pellets [1-4] which are necessary for bulk applications. RE-123 superconducting bulks with high superconducting properties have been developed by the oxygen controlled melt growth (OCMG) process [4-7], which applies low oxygen partial pressure during heat treatment. An important drawback of these materials is, however, that the actual composition is  $RE_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ . In this system, the relatively large radii rare earths, particularly Nd, easily substitute into the Ba sites, leading to depressed  $T_c$ 's when the solidification is performed in air. Recently, several research groups have been reported on the  $(RE/Y)Ba_2Cu_3O_y$  [RE; Nd, Sm, (RE/Y)-123] composite superconductors by a melt-textured growth process in air [8-10]. For achieving better superconducting properties in (YSN)-123 system, the inclusion size of (YSN)211 should be as small as possible, as well as controlling the (YSN)211 volume and the distribution within the (YSN)123 superconducting matrix. The refinement of the (YSN)211 inclusions might explain the im-

provement of the superconducting properties. In this paper, we reported microstructure and superconducting properties of (YSN)-123 composite oxides with  $CeO_2$  addition prepared by TSMG process in air atmosphere.

## 2. Experimental Procedure

Precursor powder was prepared from raw materials of  $Y_2O_3$ ,  $Sm_2O_3$ ,  $Nd_2O_3$ ,  $BaCO_3$  and  $CuO$ . The amounts of these powders were calculated to ensure  $(Y_{0.5}Sm_{0.25}Nd_{0.25})Ba_2Cu_3O_y$ . The powders were mixed thoroughly by ball milling in acetone for 24 h and calcined consequently at  $890^\circ C$  for 24 h twice in air with intermediate grinding. The calcined powders were attrition milled for 6 h in acetone using zirconia balls with a rotation speed of 450 rpm. The attrition milled (YSN)-123 powders were dried in air and then isostatically pressed into a pellet with a diameter of 10 mm and height of 5 mm in an ethyl alcohol chamber. For TSMG processing, the (100) plane of the Nd-123 seed crystal was placed on the top of the (YSN)-123 pellet. A (001) MgO single crystal was used as the substrate. The samples were heat treated according to the heat schedule shown in Fig. 1. For oxidation, the samples were heated to  $450^\circ C$  for 100 h in an oxygen gas flow. The microstructural analysis was examined by a X-ray diffraction (XRD), scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The superconducting properties, were measured with a Quantum Design MPMS SQUID magnetometer in the zero-field cooled (ZFC) mode in an applied magnetic field of 1 mT.

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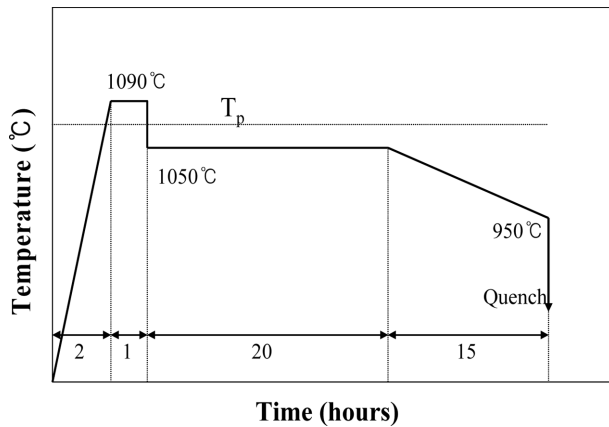


Fig. 1. Heat pattern for TSMG (YSN)-123 composite oxides. (YSN)-123 crystals were grown at 1050°C after seeding a pellet with a Nd-123 seed crystal.  $T_p$  denotes the peritectic temperature (1060°C) of the (YSN)-123 composite system.

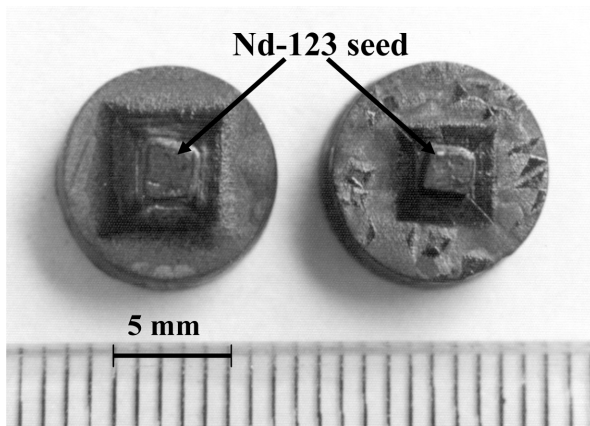


Fig. 2. The typical top view of the as grown (YSN)-123 composite crystal using melt-textured Nd-123 seed crystal.

### 3. Results and Discussion

Figure 2 shows the typical top view of the (YSN)-123 sample grown using a-b plane of a Nd-123 seed crystal. The (YSN)-123 crystal grew epitaxially from the Nd-123 seed crystal with rectangular shape in the plane of the pellet. The DTA curve for (YSN)-123 sample in air at a heating rate 10°C/min is shown in Fig. 3. The largest endothermic deflections is due to the decomposition of the superconducting phase. As can be seen in Fig. 3, the peritectic decomposition temperature ( $T_p$ ) for (YSN)-123 sample has 1060°C in air atmosphere. Figure 4 shows the XRD pattern of the cleavage surface of the melt-grown (YSN)-123 crystal. As shown in Fig. 4, there are only (00L) peaks, which means that the c-axis of the crystal is normal to the plane. Since no other phases are observed in this pattern, the grown crystal is a single

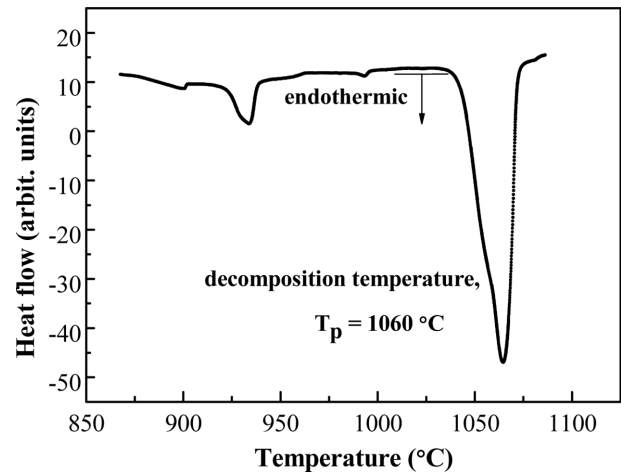


Fig. 3. DTA curve for (YSN)-123 composite oxides in air atmosphere.

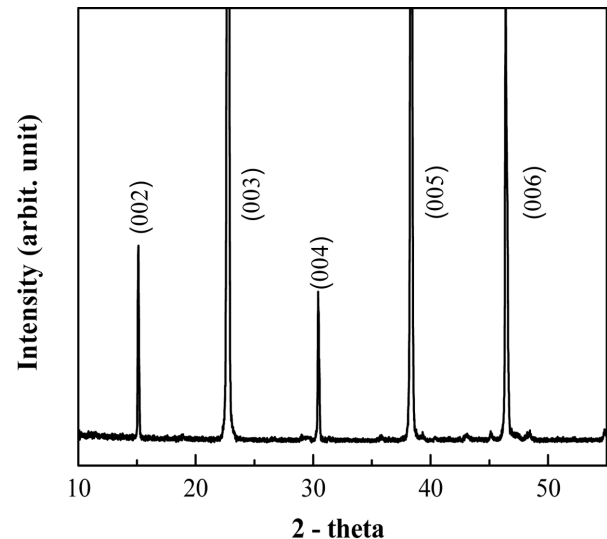


Fig. 4. The X-ray diffraction pattern of the TSMG (YSN)-123 single crystal, showing the near perfect c-axis orientation.

phase of (YSN)123. Figure 5 shows the SEM photograph of melt-grown (YSN)-123 composite superconductors with 1 wt% CeO<sub>2</sub> addition. It can be seen in Fig. 5 that the uniform (YSN)211 inclusions are trapped within the (YSN)123 superconducting matrix. In the Y-Ba-Cu-O system, it has been reported that the Y211 shape is influenced by various fabrication parameters such as the type of additive [1, 2, 11]. In CeO<sub>2</sub> addition sample, the (YSN)211 inclusion size was remarkably reduced and the inclusions finely dispersed within the (YSN)123 matrix [1, 11]. The detailed microstructural analysis has been carried out by transmission electron microscopy (TEM) on the same samples. TEM was performed on a sample annealed at 450°C for 100 h. After oxygen annealing, many defects are observed in the orthorhomb-

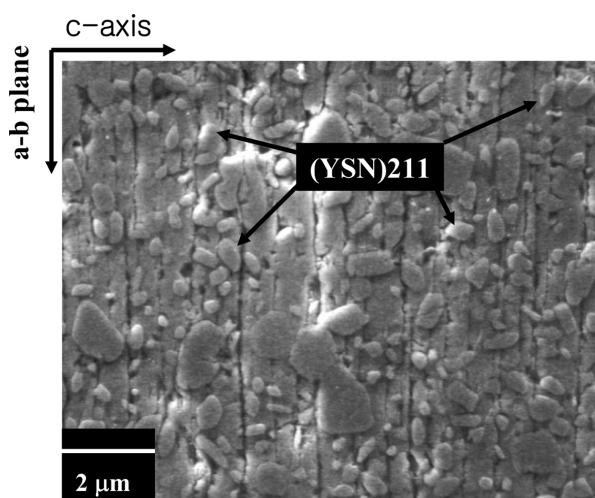


Fig. 5. SEM photograph of the TSMG (YSN)-123 composite crystals with 1 wt%  $\text{CeO}_2$  addition. Note finely dispersed the (YSN)211 inclusions are trapped into the (YSN)123 matrix.

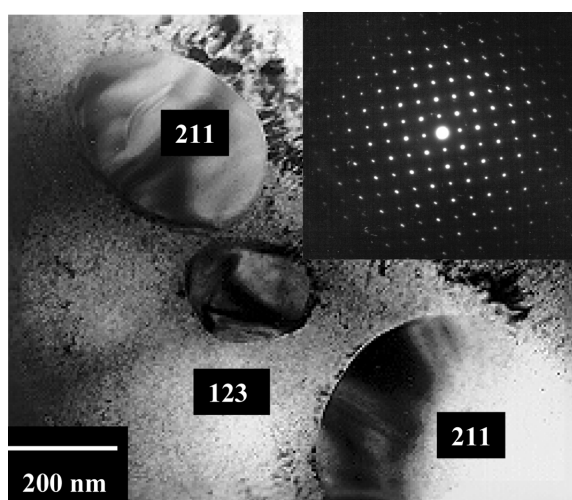


Fig. 6. TEM bright field image and selected area electron diffraction (SAED) pattern of the TSMG (YSN)-123 composite crystal with 1 wt%  $\text{CeO}_2$  addition.

bic (YSN)123 matrix, especially around the trapped (YSN)211 inclusions. Figure 6 show the [001] TEM bright field image and selected area electron diffraction (SAED) pattern of the melt-grown (YSN)-123 crystal with  $\text{CeO}_2$  addition. It can be seen that fine (YSN)211 inclusions, spherical in shape, are observed in the (YSN)123 matrix. A DC magnetization measurement was carried out using a quantum design SQUID magnetometer. Figure 7 shows the temperature dependence of magnetization of (YSN)-123 composite single crystals with  $\text{CeO}_2$  addition. The field of 1 mT was applied parallel to the c-axis from 10 K to 110 K. In melt-grown (YSN)-123 sample with  $\text{CeO}_2$  addition an onset  $T_c > 91$  K and sharp transition was observed. This result is attributed to, in part,

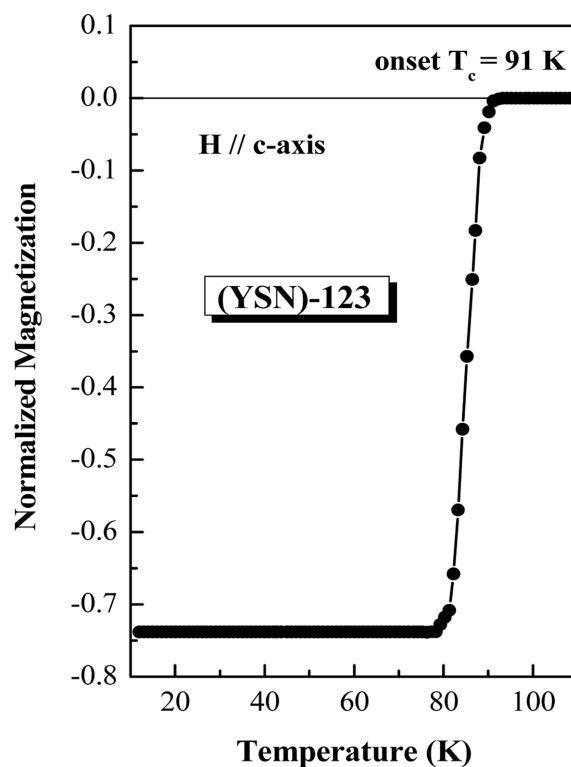


Fig. 7. Temperature dependence of dc magnetization of the TSMG (YSN)-123 composite crystals with 1 wt%  $\text{CeO}_2$  addition. The measurements were performed with zero field cooled (ZFC) warming procedure with an applied field of 1 mT and for  $H // c$ -axis direction.

the finely dispersive (YSN)211 inclusions which apparently act as effective pinning centers. It seems that the flux pinning mechanism by the (YSN)211 addition within the (YSN)123 matrix is more effective in improving superconducting properties ( $T_c$ ,  $J_c$ ) of the (YSN)123 phase than the coarse (YSN)211 dispersion.

#### 4. Conclusions

In summary, we have succeeded in the syntheses of c-axis oriented (YSN)-123 composite superconductors with  $\text{CeO}_2$  addition by the TSMG process in air atmosphere using Nd-123 as seed crystal. Well-textured (YSN)-123 composite crystal grew epitaxially from the Nd-123 seed crystal with rectangular shape in the plane of pellet. The size of (YSN)211 nonsuperconducting inclusions of the melt textured (YSN)-123 samples with  $\text{CeO}_2$  addition were remarkably reduced and uniformly distributed within the (YSN)123 matrix. The melt-textured (YSN)-123 composite superconductors with  $\text{CeO}_2$  addition, showed an onset  $T_c$  91 K and sharp superconducting transition.

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