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Neodymium Oxide Doped Melt Textured $YBa_2Cu_3O_{7-x}$ Single Crystals

Fatih Dogan, Stephen W. Sofie, William C. Hicks, Michael Strasik, Kevin E. McCrary, and Arthur C. Day

***Abstract*—Processing, microstructure and property relationships in neodymium oxide doped high temperature superconducting $YBa_2Cu_3O_{7-x}$ (Y123) were investigated. It has been observed that a small amount ($\sim$0.25–1 mol%) of Nd_2O_3 results in the formation of nanosized secondary phases which may have a significant effect on the superconducting properties of melt textured Y123 single crystals. It was further observed that addition of Nd_2O_3 greater than 1 mol% leads to multiple nucleation during solidification of Y123 and results in polycrystalline samples. Melt textured single crystals of Y123 with $<$1 mol% were successfully grown and characterized with respect to microstructural development and superconducting properties.**

***Index Terms*—Critical current, melt textured single crystals, neodymium oxide, $YBa_2Cu_3O_{7-x}$.**

I. Introduction

FOR the past several years much attention has been focused on oxygen-controlled-melt-growth (OCMG) of Nd123 and Sm123 [1], [2] as a method for obtaining higher and sharper superconducting transition temperatures (T_c) and enhanced critical current density (J_c) in high magnetic fields. Mixing different RE123 compounds and adding RE oxides to the $YBa_2Cu_3O_{7-x}$ system are two alternative methods that have been investigated in recent studies [3]–[5]. Woolf *et al.* [6] and Likodimos *et al.* [7] investigated adding of Yb123 to Y123 to achieve a lower melting temperature in the mixed composition. Matthews *et al.* [8] obtained significantly higher J_c in composite (Nd/Y)123 material as compared to Y123. Schätzle *et al.* [9] examined melt textured Nd + Y123 and Sm + Y123 composites with 25–75 mole percent of RE additions in order to achieve high J_c values. Varanasi *et al.* [10] studied Nd (4, 20 and 40 mol%)–Y123 polycrystalline samples and reported that addition of Nd led to a broad T_c and to an improved J_c. Feng *et al.* [11] investigated Er additions (20–80 mol%) and Gd additions (20–60 mol%) and reported J_c enhancements for melt textured materials.

Er211–Y123 materials prepared by Balkin and McGinn [12] and Sm211–Y123 materials by Zhang *et al.* [13] also revealed high J_c in melt textured samples. A high J_c at 2 T applied magnetic field was reported in the later work which is similar to the results reported for melt textured Sm123 at 77 K [1].

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The above studies in RE–Y123 systems have addressed the properties of sintered or melt textured polycrystalline materials. However, growth of large single crystals in these systems has not been reported. The properties of single crystals are important in applications for high flux-trap magnets. The objectives of this work are to grow large Nd-doped Y123 composite single crystals by top seeded melt texturing process and to determine the superconducting properties, T_c and J_c.

II. Experimental Procedure

Powders of Y123 with 10 mole percent Y_2BaCuO_5 (Y211) and 0.2 weight percent Pt were obtained from Praxair Specialty Ceramics, Inc., Woodinville, WA. The powder was mixed with various amount of neodymium oxide powder, homogenized in heptane by stirring and ultrasonication for 6 minutes and then dried in flowing nitrogen [14].

The powders were pressed into disc shaped samples using a stainless steel die (diameter 2.5 cm) and subsequently compacted isostatically with a pressure of 86 MPa. The compacts were sintered at 890 °C for eight hours. Sm123 seed crystals were placed on the top center of the samples which were then processed in a furnace on Y211 based setters for crystal growth. The furnace was heated to 1055 °C at a rate of 200 °C per hour and held for 0.5 hours to insure melting. The furnace was then quickly cooled to 1015 °C followed by slow cooling at 0.6 °C per hour to 960 °C to allow crystal growth. The furnace was then cooled to room temperature at a 200 °C per hour cooling rate. Oxygenation of the samples was carried out at 425 °C in flowing oxygen gas for one week.

The trapped magnetic field measurements were conducted using oxygenated samples at 77 K under a 0.5 Tesla applied field. Magnetization measurements were conducted in a magnetometer (Quantum Design Model 6000 Physical Properties Measurement System) to determine T_c and J_c values. The microstructures of the samples were studied using a Jeol JXA-8600 Superprobe Scanning Electron Microprobe (SEM) equipped with electron dispersive spectrometer (EDS).

III. Results and Discussion

Neodymium has an ion size close to that of barium, which allows substantial Nd/Ba substitution when compared to the yttrium system. Relative ion sizes can be seen in Table I.

Studies have shown that Nd123 crystals and heavily Nd-doped Y123 crystals yield decreased critical temperatures and current densities as a result of neodymium substitution with barium. Substitution of Nd into Y sites results in the formation of a new superconducting cell with very similar properties, which may not create new flux pinning sites. The barium site substitution of Nd, however, has been shown to have a negative effect on

TABLE I
RELATIVE ION SIZES FOR RE123 SYSTEMS

Ion Type	**Ion Radius (nm)**
Yttrium (Y^{3+})	0.102
Barium (Ba^{2+})	0.128
Copper (Cu^{+})	0.078
Neodymium (Nd^{3+})	0.112

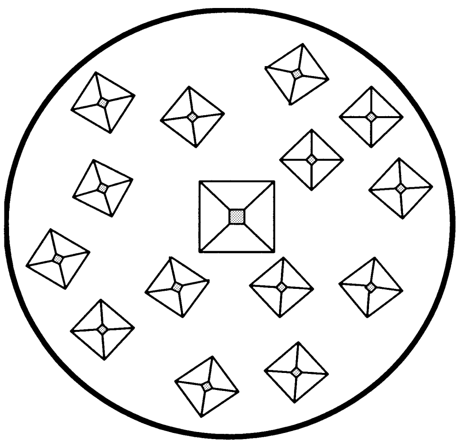

Fig. 1. Schematic illustration of Nd123 cluster/seed formation resulting in polycrystalline Y123.

Fig. 2. Melt textured Y123 processed with 2.0 mol% neodymium oxide.

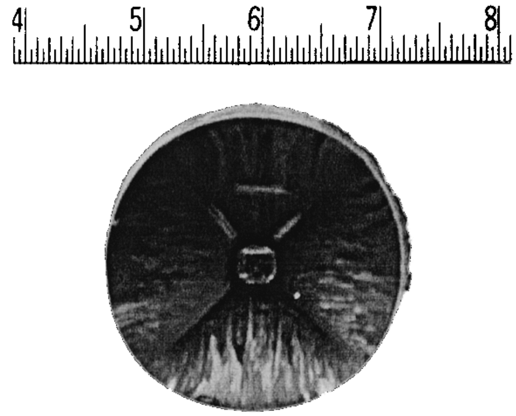

Fig. 3. Bulk single crystal grown with 0.25 mol% neodymium oxide.

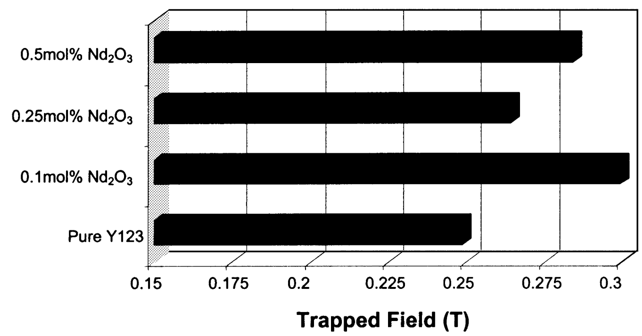

Fig. 4. Maximum trapped field of 2.5 cm diameter undoped and Nd-doped Y123 crystals magnetized under 0.5 T.

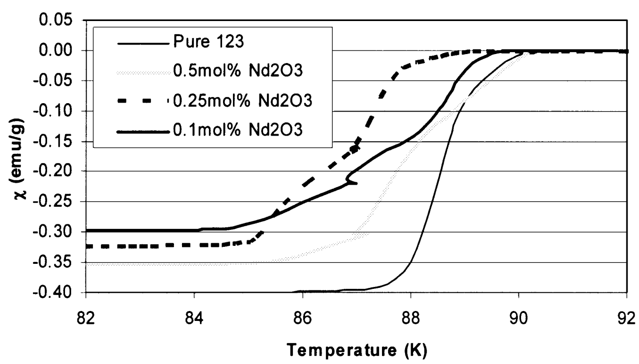

Fig. 5. Critical temperature T_c of undoped and neodyminm oxide doped single crystals.

the superconducting properties of Nd123, which, in turn may provide a method of introducing extremely small, yet strong flux pinning sites into Y123 if doped with a proper amount. Preliminary studies on Nd–Y123 proved unsuccessful using dopant quantities ranging from 1–5 mol%. The resultant crystals were polycrystalline, where the number of grains increased with increasing neodymium concentrations. This difficulty is linked to in-situ formation of Nd123 clusters large enough to act as seeds for nucleation. As the Y123 solidifies, the Nd123 clusters (melting temperature approx. 65 °C higher than that of Y123) result in burst of nucleation and growth of multiple crystals in random orientations as shown in Figs. 1 and 2.

The solubility limit of neodymium in Y123 appears to be approximately 0.5 mol% at which point Nd exchanges significantly for the Y sites as well as the Ba sites. Given the establishment of a Nd_2O_3 solubility limit in Y123, 25–30 grams ($\sim$2.5 cm diameter) melt textured single crystals have been grown successfully with 0.1, 0.25, and 0.5 mol% neodymium oxide additions as seen in Fig. 3.

The Nd_2O_3 doped samples showed an increase in trapped field over pure Y123 shown in Fig. 4. Although the measurements on multiple samples and averaging the data would have generated more accurate results, a clear improvement of flux pinning capacity has been observed in Nd–Y123 single crystals as compared to that of the undoped Y123. Further work will be required to verify these data.

The T_c data in Fig. 5 show a broad transition ranging from 85–90 K. Considering the very small differences in Nd_2O_3 concentration, combined with the solid state mixing it is difficult to achieve the level of homogeneity which would allow good cross comparison between Nd_2O_3 dopant concentrations. While there

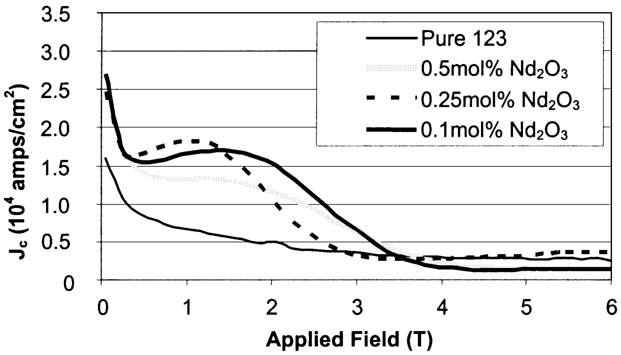

Fig. 6. Critical current density as a function of applied field.

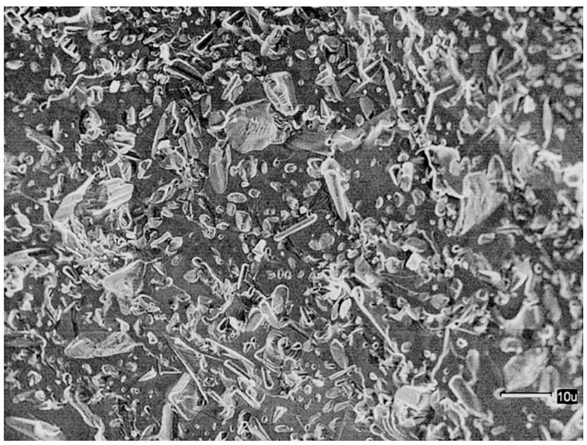

Fig. 7. Melt textured microstructure of 0.1 mol% neodymium-doped Y123 (bar $= 10\ \mu$m).

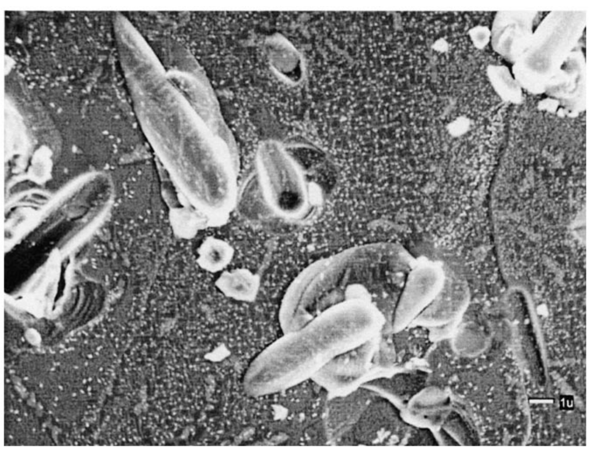

Fig. 8. Melt textured microstructure of 0.25 mol% Nd-doped Y123 showing large Y211 particles and uniformly distributed nanosized inclusions (bar $= 1\ \mu$m).

is a significant T_c suppression with the Nd_2O_3 dopant, the J_c behavior of these samples in Fig. 6 show strong fishtail effects in the 1–2 Tesla region. Undoped Y123 does not exhibit a fishtail effect of J_c and any T_c suppression as shown previously [15].

A typical microstructure of Y123 with needle-like Y211 particles distributed within the matrix is depicted in Fig. 7. Given that the Nd forms substitutional defects in Y123 system, the presence of secondary phases was not expected at low Nd-concentrations. While this is the case in the 0.1 mol% neodymia-doped samples, formation of light contrast phases as nanosized inclusions are readily evident in Fig. 8 as the Nd concentration increases to 0.25 mol%. Moreover, very low Nd_2O_3 dopant levels should not result in appreciable stoichiometry offsets in the system. The defects associated with the Nd/Ba substitution and the presence of secondary phases as nanosized inclusions can be attributed to formation of additional flux pinning centers. Further microstructural studies are planned to identify the composition of nanosized secondary phases.

IV. Conclusions

Melt textured Y123 single crystals were successfully grown with addition of less than 1 mol% neodymium oxide. Higher amount of additives lead to multiple nucleation during solidification and polycrystalline. Improvement of superconducting properties in melt textured Y123 single crystals doped with <1 mol$\%$ were attributed to the formation of substituitional defects and nanosized secondary phases.

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