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Properties of Mg-doped Nd-Ba-Cu-O generic seed crystals for the top seeded melt growth of (RE)-Ba-Cu-O bulk superconductors

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Abstract: We have recently developed a new generic seed crystal that has been used successfully to fabricate any oriented, single grain (RE)-Ba-Cu-O bulk superconductor by a cold seeding technique. In this paper we report the chemical, structural and microstructural properties of these seed crystals, including the variation of melting point, crystallographic parameters and volume fraction of Mg-rich inclusions in the $Nd_{1+x}Ba_{2-x}(Cu_{1-y}Mg_y)_3O_z$ matrix as a function of externally added MgO content. The influence of Mg-doping on the superconducting transition temperatures of YBCO grains fabricated using these seeds is investigated. Finally, an optimum MgO content of the generic seed that effectively controls the orientation of the seeded grain without compromising its superconducting properties is suggested from the many seed crystals fabricated with a wide range of Mg-rich addition.

1 Introduction

Melt processed (LRE)-Ba-Cu-O superconductors [(LRE)BCO], where LRE is a light rare earth element such as Nd, Sm, Eu and Gd, are known to exhibit higher J_c's and higher irreversibility fields than YBCO [1-3]. In the absence of a suitable seed, however, it has been difficult to fabricate large grain (LRE)BCO superconductors using a practical cold seeding process. We have reported recently that Mg-doped NdBCO exhibits generally a similar crystal structure to that of the $(RE)Ba_2Cu_3O_{7-\delta}$ (RE-123) family of compounds but with a significantly higher melting temperature. As a result, we have demonstrated successfully that Mg-doped NdBCO melt-processed crystals can be used as generic seeds for the growth of large grain (LRE)BCO superconductors [4], and that the new seed crystal simplifies significantly the fabrication process of light rare earth (LRE)-Ba-Cu-O single grain superconductors grown by top seeded melt growth (TSMG) [5]. In this paper, we report the fabrication and properties of a wide range of Mg-containing NdBCO generic seed crystals. The influence of Mg-doping on the superconducting transition temperatures on large YBCO grains fabricated using these seeds is investigated. Finally, the optimum composition of MgO-doped generic seed that enables effective control of the orientation of large seeded grains without degrading their superconducting properties is identified.

2 Experimental

Mg-doped generic seed crystals with varying MgO content were fabricated by solid state reaction and conventional melt processing. $Nd_1Ba_2Cu_3O_{7-\delta}$ (Nd-123) and $Nd_4Ba_2Cu_2O_{10}$ (Nd-422) phase powders were mixed thoroughly with starting compositions Nd-123 + 12 mol% Nd-422 + z wt% MgO, where $0 < z < 20$. Differential thermal analysis (DTA) in flowing air was carried out on each precursor. The powders were then pressed uniaxially into pellets and melt processed under air using the thermal profile described in Ref. 4. The crystal structure of each calcined powder was identified by XRD and T_c of the fully-processed single grains was measured using a SQUID magnetometer.

3 Results and Discussion

3.1 Seed Crystal Characteristics

Melting point: DTA traces for precursor powders with compositions Nd-123 + 12 mol% Nd-422 + 1 wt% MgO, YBCO, GdBCO and NdBCO are shown in Figure 1. The melting point of the MgO doped NdBCO powder is observed to be at least 15-20 °C higher than that of other (RE)BCO compositions, and is the highest decomposition temperature observed to date of all the (RE)BCO systems.

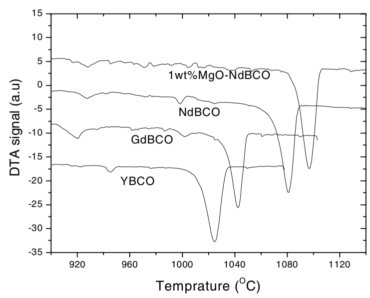

Figure 1. DTA traces of YBCO, GdBCO, NdBCO and NdBCO containing 1 wt% MgO

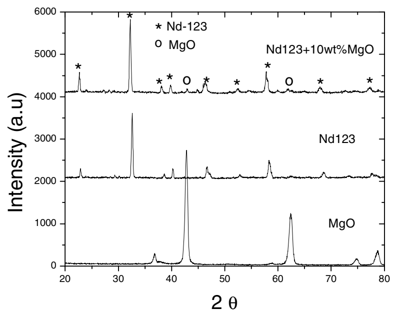

Figure 2. XRD patterns for Nd-123, MgO and for a powdered melt processed sample with starting composition Nd-123 + 10 wt% MgO

***Crystal structure*:** Figure 2 shows that all the XRD peaks can be indexed to the Nd-123 (but with shifted peak positions) and MgO phases. The peak positions corresponding to Mg-doped Nd-123 are similar to those of undoped Nd-123, suggesting that the basic crystal structure is not affected by Mg-doping. The difference in the peak positions for the Mg-doped Nd-123 and un-doped Nd-123 phases observed in the XRD data correspond to a mismatch in the *a/b*-axes of the samples of < 0.7%.

Chemical stability: The stability of the seed with the Ba-Cu-O liquid phase in the peritectically molten state was investigated by performing DTA on mixed powders of small concentrations of Ba-Cu-O + Nd_2O_3 + seed crystal. The DTA trace obtained showed no noticeable phase transformation between 950 °C and 1080 °C, which is the temperature range within which the peritectic reaction occurs in most (RE)-123 compounds.

As demonstrated by the above results, Mg-NdBCO seeds have a phase composition that is chemically compatible with the (LRE)BCO system, but with a significantly higher melting point. Significantly, the lattice parameters of the generic seed are similar to the undoped Nd-123 phase, which, in turn, has similar lattice parameters to the $(LRE)Ba_2Cu_3O_{7-\delta}$ [(LRE)-123] superconducting phase.

3.2 Properties of seeds containing various amounts of MgO

***Melting point*:** The melting point, T_m, of the seed crystals of composition Nd-123 + 12 mol% Nd-422 + z wt% MgO with $0.1 < z < 10$ as a function of MgO content in the precursor powder is shown in Figure 3(a). The melting point of the seed crystals increases by at least 15-20 °C as MgO content increases by up to 0.6 wt%; it then remains constant for higher concentrations of MgO. This suggests that the samples containing ≥ 0.6 wt% MgO are suitable for use as generic seed crystals for melt processing all (RE)BCO systems.

***Microstructure*:** Image analysis reveals that the area fraction of MgO particles is negligible for an MgO content of less than 1wt%, as shown in Figure 3(b), which suggests strongly that the entire Mg content in the precursor powder substitutes into the Nd-123 matrix during melt processing. The area fraction of MgO inclusions is observed to increase linearly when the MgO content of the precursor powder is increased above 1 wt%. This linear behavior indicates that the Mg substitution level in the matrix saturates and the excess Mg becomes trapped in the solidifying $Nd_{1+x}Ba_{2-x}Cu_{3-y}Mg_yO_{7-\delta}$ matrix.

Mg solubility: The measured Mg substitution level, *y*, in the $Nd_{1+x}Ba_{2-x}Cu_{3-y}Mg_yO_{7-\delta}$ phase matrix of melt processed grains was measured by electron probe microscopy (EPMA) and is shown as a function of MgO content in the (Nd-123 + 12 mol% Nd-422) precursor pellet in Figure 3(c). The amount of Mg substitution increases rapidly from 0 to 0.17 when the MgO content in the precursor powder is increased from 0 to 1 wt%, and saturates for concentrations between 1 and 10 wt%.

***T_c degradation*:** The onset T_c of superconductivity as a function of MgO content in the NdBCO precursor powder for the melt processed samples is shown in Figure 3(d). T_c is observed to decrease rapidly from 92.5 K to about 65 K by increasing the MgO content from 0 to 1 wt%. Further increase in MgO concentration is observed to suppress the superconductivity so much that samples containing above 2 wt% MgO are not superconducting in the measured temperature range above 10 K. Error bar represents variation from sample to sample due to the variation of Nd/Ba solid solution. Suppress in T_c due to Mg doping in Cu site has been also observed in YBCO system [6].

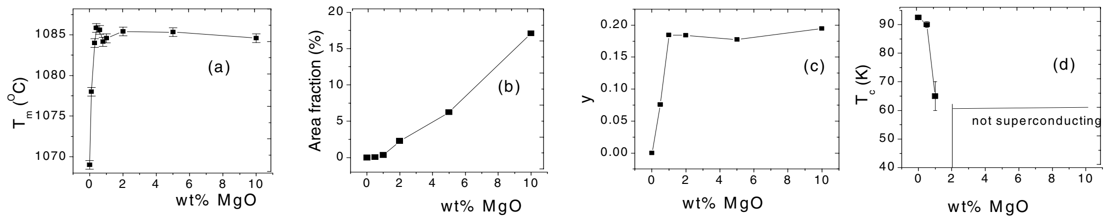

Figure 3. Properties of seed crystals melt processed from precursor powders with starting composition Nd-123 + 12 mol% Nd-422 + z wt% MgO. (a). The melting point (b) The area fraction of MgO inclusions in Nd-Ba-Cu-Mg-O matrix (c) Mg concentration measured from EPMA and (d) the transition temperatures as a function of MgO content in the precursor powder.

3.3 Fabrication of (LRE)-Ba-Cu-O single grains with $Nd_{1+x}Ba_{2-x}Cu_{3-y}Mg_yO_{7-\delta}$ generic seeds

Fig. 4 shows a variety of single grains grown using Mg-doped NdBCO seeds. Table 1 shows a list of (RE)BCO single grains grown under different processing atmospheres using the generic seed crystal. The superconducting properties of these single grains are comparable to that of grains fabricated with other seeding processes such as hot seeding and seeding with MgO seed crystals. The influence of the seed crystal composition on the superconducting properties of the seeded grains is discussed in next section. This is the first time that a wide range of single grain (RE)BCO superconductors has been fabricated successfully under both air and reduced oxygen partial pressures with a single, generic seed crystal.

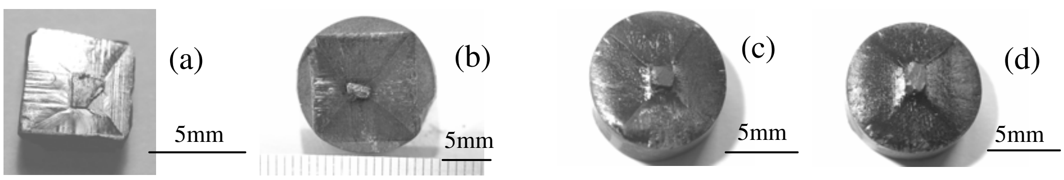

Figure 4 Photographs of top seeded melt grown single grains with starting compositions (RE)-123 + 30 wt% RE-211 + 0.1 wt% Pt. (a) RE= Nd, (b) RE = Sm, (c) RE= Gd, and (d) RE = Y.

Table1. List of (RE)BCO single grains fabricated with generic seed crystal.

(RE)BCO systems	Atmosphere	(RE)BCO systems	Atmosphere
NdBCO	Air,	GdBCO	Air
NdBCO	1 % O_2 + 99 % N_2	(Y,Nd)BCO	Air
(Nd, Eu, Gd)BCO	1 % O_2 + 99 % N_2	(Y,Nd)BCO	1 % O_2 + 99 % N_2
SmBCO	Air, 1 % O_2 + 99 % N_2	YBCO	Air

3.4 The effects of the Mg-doped seed on T_c of YBCO

The use of Mg-doped NdBCO as a seed crystal may have serious implications for suppressing the superconducting properties of seeded grains, since Mg doping affects severely T_c of MgO-NdBCO. The superconducting properties of YBCO single grains melt processed using $Nd_{1+x}Ba_{2-x}Cu_{3-y}Mg_yO_{7-\delta}$ seed crystals containing various concentrations of MgO were measured to investigate the effect of Mg contamination. The results show that T_c is the same as that of YBCO seeded by undoped NdBCO and is constant for 0.5 and 1wt% concentrations of MgO. T_c decreases slightly (up to 1 K) and ΔT_c increases up to 3 K for samples seeded by MgO-NdBCO containing 5 and 10 wt% MgO. The results of this study suggest that an optimum seed crystal of starting composition Nd-123 + 0.6 - 1.0 wt% MgO yields the best superconducting properties in bulk melt processed (RE)BCO grains.

4 Conclusions

It has been shown that the addition of Mg to $NdBa_2Cu_3O_{7-\delta}$ increases the melting point of the parent superconducting compound whilst retaining the crystal structure of the undoped material. A careful investigation has revealed that Mg substitutes into the Nd-123 lattice and saturates at a substitution level of y = 0.17 in $Nd_{1+x}Ba_{2-x}Cu_{3-y}Mg_yO_{7-\delta}$. The melting point of the Mg-containing compound is also observed to saturate at this level of doping. Melt processed Mg doped NdBCO crystals have been used successfully to seed the growth of various (RE)BCO systems under both air and reduced O_2 partial pressure atmospheres. T_c of YBCO prepared by TSMG is not affected by the use of a Nd-Ba-Cu-Mg-O seed containing a maximum of 1 wt% MgO. As a result, seeds with an MgO content of up to 0.6-1 wt% are recommended for TSMG processing of (RE)BCO single grains.

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