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## Site depending impurity effect on the properties of melt-processed bulk superconductors

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#### Abstract

The effect of doping impurities to Gd-based bulk superconductors prepared by melt processing was studied. The trapped field at liquid nitrogen temperature increased with ZnO content up to 0.1 wt%, with an enhancement of 37% compared to the non-doped sample. The trapped field decreased, however, when the content of ZnO was increased beyond this optimum level. When  $Co_3O_4$  was added, on the other hand, the trapped field increased monotonically with the  $Co_3O_4$  content up to 0.2 wt% and an enhancement of 32% was observed. We also measured the local superconducting properties on small samples that were taken from the bulk superconductors, which revealed that the difference of how the impurities affect the trapped field can be primarily attributed to the different influence on the critical temperature depending on the substituting site.

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Keywords: Melt processing; Bulk superconductor; Impurity effect; Trapped field; Critical current density

### 1. Introduction

Since the report by Krabbes et al. that adding a small amount of ZnO to Y-Ba-Cu-O prepared by melt processing resulted in appearance of a peak effect in the magnetic field dependence of the critical current density  $(J_c)$  and an enhancement in the trapped field  $(B_T)$  [1], many studies have been devoted to improve the properties of bulk superconductors through Zn doping [2–8]. The enhancement of  $J_c$ and  $B_{\rm T}$  can be attributed to pinning sites which are created by locally suppressing superconductivity when Zn substitutes for Cu in the  $CuO_2$  plane. A similar effect can be expected for other impurities as well, and an enhancement of the superconducting properties have been reported for Ni and Li doping, which are considered to substitute also for Cu in the CuO<sub>2</sub> plane [9,10]. There are also impurities which substitute to the other Cu site, the CuO chain site, and affect the superconductivity. Because of the different role the two Cu sites are playing in high temperature superconductivity, it is of great interest to compare how the properties of melt-processed bulk superconductors are affected with substitution of impurities to these two Cu sites.

In the present study, we compare the effect of doping Zn and Co to melt-processed Gd-Ba-Cu-O. The Gd-system was chosen to explore the effect of Zn doping in a different material, as we have already studied the Dy- and Sm-based materials [5-8]. Whereas Zn ions substitute for Cu in the CuO<sub>2</sub> plane, Co ions are considered to substitute mainly for the CuO chain site [11]. The superconducting transition temperature ( $T_c$ ) of polycrystalline YBa<sub>2</sub>(Cu<sub>1-x</sub>Co<sub>x</sub>)<sub>3</sub>O<sub>y</sub> was reported to decrease only slightly [12], or to show even a little increase [13], when the Co content x was small. This is in a sharp contrast with Zn doping, which strongly depresses  $T_{\rm c}$ . Nevertheless, our study indicates that the enhancement of  $B_{\rm T}$  is similar for Zn and Co doping. This result contrasts with that of a quite recent paper which reports only a very small enhancement of  $B_{\rm T}$  when Co was doped to Y-Ba-Cu-O [14]. The dependence of  $B_{\rm T}$ on the impurity content was different between our Znand Co-doped samples, however, and we studied the local

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properties to discuss the difference of the effect of Zn and Co on the superconducting properties.

#### 2. Experimental

The Gd-Ba-Cu-O bulk superconductors were prepared by a melt process. Commercially available powders of the GdBa<sub>2</sub>Cu<sub>3</sub>O<sub>v</sub> (Gd123) and Gd<sub>2</sub>BaCuO<sub>5</sub> (Gd211) phases were mixed in a molar ratio of Gd123:Gd211 = 5:2, and 15 wt% Ag<sub>2</sub>O was added to improve the mechanical strength of the sample, 0.5 wt% Pt to refine the Gd211 precipitates. Zn- or Co-doped samples were prepared by adding various amounts of ZnO or Co<sub>3</sub>O<sub>4</sub> up to 0.2 wt% to this starting powder. The content of ZnO or Co<sub>3</sub>O<sub>4</sub> is described as a percentage of the weight sum of the Gd123 and Gd211 powders throughout this paper. This is because we cannot expect that all Zn or Co atoms will be taken into the Gd123 phase and the actual atomic composition of the superconducting phase would be different from the nominal one. To be able to compare our results with the data published by other groups however, it would be worthwhile to point out that if all Zn atoms would substitute for Cu of the Gd123 phase, adding 0.1 wt% of ZnO corresponds to replacing 0.398% of Cu ions to Zn, and 0.1 wt% Co<sub>3</sub>O<sub>4</sub> to 0.404% of Cu ions to Co.

Melt processing was performed in a 1% O<sub>2</sub>/99% Ar atmosphere. After the melt processing was finished, the sample was heated to 400 °C, and then slow cooled to 300 °C in 350 h under flowing pure oxygen gas. The finished bulk superconductors were 30 mm in diameter. Mapping of the magnetic flux density trapped by the samples was performed by scanning a Hall sensor, as described elsewhere [15]. To study the local superconducting properties, we measured magnetization of small specimens that were cut from the bulk samples using a SQUID magnetometer (Quantum Design, MPMS-7) by applying the external field parallel to the *c*-axis. The irreversibility field ( $B_{irr}$ ) was determined from the field dependence of  $J_c$  with a criterion of  $J_c = 50 \text{ A/cm}^2$ .

#### 3. Results and discussion

Fig. 1 shows the trapped field measured at liquid nitrogen temperature of the Gd-based bulk superconductors prepared with various amounts of ZnO and Co<sub>3</sub>O<sub>4</sub>. The field distribution of all samples plotted in Fig. 1 had a single peak, indicating that the samples are free from severe macro-cracks or weak links that deteriorate their capability of trapping magnetic flux. Fig. 1 shows that  $B_T$  increased both for Zn and Co doping.  $B_T$  showed a peak at ZnO content of 0.1 wt%, with an enhancement of 37% compared to the non-doped sample.  $B_T$  of the Co<sub>3</sub>O<sub>4</sub> added sample also increased with the impurity content, and an enhancement of 32% was observed at the upper limit of the Co<sub>3</sub>O<sub>4</sub> content of the present study. This is in contrast with the results of Zhou et al. who reported quite recently that Y-based bulk superconductor did not show a significant increase



Fig. 1. The trapped magnetic field at liquid nitrogen temperature of meltprocessed Gd–Ba–Cu–O as a function of ZnO and  $Co_3O_4$  content. The trapped field was measured at the peak position of the field distribution map.

in  $B_{\rm T}$  when Co was doped [14]. The reason of the discrepancy is not clear, but the carrier concentration of the samples in the two studies may be slightly different because of a different annealing treatment or the effect of Co on the superconducting properties may be different for the Y- and Gd-based systems, especially because the latter forms a Gd/Ba solid solution.

Fig. 1 shows that  $B_T$  increased steeply with ZnO content, but then dropped sharply when the impurity content was increased beyond 0.1 wt%. The increase of  $B_T$  was more gradual when Co<sub>3</sub>O<sub>4</sub> was added, and  $B_T$  continued to increase up to at least 0.2 wt%. To elucidate the reason of this difference in the impurity content dependence, we measured the local superconducting properties on small samples that were cut from the bulk superconductors.



Fig. 2. (a) A schematic drawing of the locations from where the small specimens for magnetization measurements were taken from the bulk superconductor. (b) The position dependence of  $T_c$  of the Gd–Ba–Cu–O bulk superconductors prepared with different amounts of ZnO and Co<sub>3</sub>O<sub>4</sub>.

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