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# Potential for improvement of pinning properties for REBCO melt-textured bulks by high energy electron irradiation



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#### 1. Introduction

RE-Ba-Cu-O (REBCO; RE = rare earth) melt-textured bulks which consist of  $REBa_2Cu_3O_{\gamma}$  (RE123) superconducting matrix with high critical temperature ( $T_c > 90 \text{ K}$ ), RE<sub>2</sub>BaCuO<sub>5</sub> (RE211) precipitates and other impurities are one of the promising materials for strong superconducting magnet systems. Trapped fields of such single grain bulk superconductors are basically determined by critical current density J<sub>c</sub>, sizes and shapes. Therefore, controlling size, density and distribution of effective pinning centers, such as nonsuperconducting nanoparticles and weak superconducting regions, is crucially important for enhancing J<sub>c</sub> and developing bulk magnets with high field-trapping properties. For REBCO melt-textured bulks, small regions where RE ions substitute for Ba sites in RE123 crystal are one of the pinning centers acting in magnetic fields [1,2]. However, excessive RE/Ba substitution leads to a serious reduction of  $T_c$  and deterioration of pinning properties [3]. It is well known that the level of RE/Ba substitution increases with an increase in ionic radii of RE<sup>3+</sup>. Our previous study on REBCO melttextured bulks revealed that this level can be controlled by mixing of RE elements [4]. In addition, it was found that the bulks with

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#### ABSTRACT

In order to investigate the potential for the increase in pinning properties for REBCO melt-textured bulks, high-energy electron irradiation (35 MeV) was performed to sample pieces with various RE/Ba substitution levels. Critical current density was drastically enhanced especially in low magnetic fields by the irradiation. It was indicated that the increase in pinning force is larger for samples with lower RE/Ba substitution level possibly due to their larger superconducting condensation energy of the RE123 crystal. © 2017 Published by Elsevier B.V.

moderately low level of RE/Ba substitution maintain the strong superconductivity of RE123 crystal up to high magnetic fields [4].

On the other hand, artificial pinning centers can be also introduced to REBCO materials by irradiation of heavy ions, neutrons, protons and electrons. Columnar, cascade or point-like defects are formed in the RE123 crystal by irradiation depending on the type of particles and their energy. For example, irradiation of heavy ion, such as Sn and Pb, introduces columnar defects parallel to the irradiation direction which can act as strong pinning sites especially when magnet fields are applied parallel to the linear defects [5]. Irradiations of neutrons and protons are also effective methods to improve field-trapping properties of REBCO melt-textured bulks [6,7]. As for electron irradiation, it is considered that point-like defects and/or their small clusters which were hardly detected by high-resolution microstructural observation were introduced [8]. The first order melting transition of the vortex system in clean untwined YBCO crystals was reported to be suppressed by electron irradiation [9]. In addition, Giapintzakis et al. revealed that a factor-of-2 increase in Jc was shown at 10 K in 1 T for YBCO single crystals after 1 MeV electron irradiation [8]. They indicated that the displacement of Cu from the CuO<sub>2</sub> planes due to 1 MeV electron irradiation corresponds to the generation of effective pinning sites [8].

In this paper, potential for improvement of pinning properties for REBCO melt-textured bulks with various level of RE/Ba

Table 1			
Electron irradiation	doses and	orthorhombicity	for five specimens.

Sample name	Orthorhombicity $\gamma_0$ / –	Electron irradiation doses / $10^{17}$		0 <sup>17</sup> cm <sup>-2</sup>	
		#0	#1	#2	#3
(Dy,Er)BCO	8.3 <sub>0</sub>		5.2	9.6	15.5
(Dy,Dy)BCO	8.4 <sub>7</sub>		4.4	10.3	-
(Y,Dy)BCO	8.5 <sub>3</sub>	pristine	5.2	9.6	15.5
(Y,Er)BCO	8.8 <sub>0</sub>		4.4	10.3	-
(Y,Y)BCO	8.9 <sub>5</sub>		4.4	10.3	-

substitution is discussed through a series of studies on the effects of high-energy (35 MeV) electron irradiation on enhancement of  $J_c$  under magnetic fields.

#### 2. Experimental

Rectangular sample pieces ( $\sim 2 \text{ mm} \times 1 \text{ mm} \times 1 \text{ mm}^{1/c}$ ) for electron irradiation were cut from the same positions, 2 mm below seed crystals, of the REBCO bulks, (Dy,Dy)BCO, (Dy,Er)BCO, (Y,Dy)BCO, (Y,Y)BCO and (Y,Er)BCO, prepared in our previous study [4]. Note that (RE',RE")BCO represents the REBCO melt-textured bulk fabricated from RE'123 and RE"211 starting powders. All the specimens showed sharp superconducting transitions with  $T_{\rm c} \sim 92 \,\rm K$  before electron irradiation [4]. Our previous study revealed that RE' and RE" are homogeneously distributed in RE123 matrix in each bulk, and that the levels of RE/Ba substitution varied depending on the combination of RE' and RE" [4]. Orthorhombicity  $\gamma_0$  (=1000 (*b*-*a*)/(*b*+*a*)) of the RE123 phase, calculated from lattice constants of a- and b-axis (b > a)), was used as an index of RE/Ba substitution level. Small  $\gamma_0$  corresponds to high-level substitution of RE for the Ba sites, because oxygen atoms easily occupy the oxygen vacancy sites between the Cu-O chains when RE<sup>3+</sup> ions substitute for Ba<sup>2+</sup> sites, which decrease the orthorhombic distortion of RE123 crystal [3].

35 MeV electron beam with a fluence of  $4.4\sim 5.9\times 10^{17}\,cm^{-2}$ was irradiated in air to the sample pieces by a linear accelerator. The dose was estimated by the electron beam current which corresponds to  ${\sim}5 \times 10^{12}\,e^-\,cm^{-2}\,s^{-1}$  and the irradiation time ~28 h The diameter of the electron beam was 4 mm. (Dy,Dy)BCO, (Y,Er)BCO and (Y,Y)BCO were irradiated twice in total, while (Dy,Er)BCO and (Y,Dy)BCO were irradiated for three times. Magnetic properties were measured by a SQUID magnetometer (Quantum Design: MPMS-XL5s) up to 5 T at 40, 60 and 77 K before and after each irradiation. Note that the magnetic measurements were performed several months after each irradiation, because the specimens were strongly radioactivated by 35 MeV electron irradiation.  $J_{\rm c}$  values were deduced from the width of magnetization hysteresis loops based on the extended Bean model [10]. Hereafter, the specimens irradiated once, twice and three times are expressed as #1, #2 and #3, respectively, while the pristine ones are shown as #0. The total doses of irradiated electron and  $\gamma_0$  of the RE123 phase of samples are summarized in Table 1.

#### 3. Results and discussion

#### 3.1. Enhancement of critical current density by addition of irradiation

The insets of Fig. 1(a)–(e) indicate temperature dependences of magnetization for (Dy,Er)BCO, (Dy,Dy)BCO, (Y,Dy)BCO, (Y,Er)BCO and (Y,Y)BCO, respectively. With an increase in the doses of irradiated electrons,  $T_c$  was decreased by ~1 K and the superconducting transition width in ZFC magnetization curves became slightly broader. It can be considered that these changes are partly due to inhomogeneous displacement of oxygen in RE123. Similar phenom-

Table	2
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 $J_c$  at 40 K in 0.5 T for #0, #1, #2 and #3 of the five specimens.

Sample name	$J_{\rm c}~(40~{\rm K},~0.5~{\rm T})/10^5~{\rm A~cm^{-2}}$			
	#0	#1	#2	#3
(Dy,Er)BCO	4.2	6.3	7.2	7.6
(Dy,Dy)BCO	2.8	4.4	5.4	-
(Y,Dy)BCO	3.3	5.2	6.4	7.0
(Y,Er)BCO	3.3	7.2	7.8	-
(Y,Y)BCO	2.5	5.5	5.6	-

ena were also confirmed in the study by Rangel et al. on 1.3 MeV electron irradiation to Ag-doped YBCO thick polycrystalline films [11]. It should be noted that temperature of the sample during irradiation was lower than 55 °C, where oxygen ion is hardly released from Cu-O chains.

On the other hand, J<sub>c</sub> was drastically enhanced especially in low magnetic fields for all specimens as clearly seen in Fig. 1(a)-(e), which show  $I_c$ -B curves at 40, 60 and 77 K for (Dy,Er)BCO, (Dy,Dy)BCO, (Y,Dy)BCO, (Y,Er)BCO and (Y,Y)BCO, respectively. For example,  $J_c$  values at 40 K in 0.5 T were increased from  $3.3 \times 10^5$ to  $5.2 \times 10^5$  and  $6.4 \times 10^5 \,\text{A}\,\text{cm}^{-2}$  after the first and the second irradiation, respectively, for (Y,Dy)BCO as shown in Fig. 1(c). This result suggests that the density of irradiation defects was increased by each irradiation and that those defects acted as effective flux pinning centers in low fields. This tendency was also confirmed for the other specimens, while the magnitude of  $J_c$  enhancement was different between them. For instance, the  $J_c$  values at 40 K in 0.5 T for #1 of (Y,Dy)BCO and (Y,Er)BCO were  $\sim\!5.2\times10^5$  and  ${\sim}7.2\times10^5\,A\,cm^{-2},$  respectively, while their pristine samples exhibited similar  $J_c \sim 3.3 \times 10^5 \, A \, cm^{-2}$  at the same measurement conditions. Jc values at 40 K in 0.5 T of the examined five specimens before and after electron irradiation are summarized in Table 2. Fig. 2 shows  $J_c^{\#1} / J_c^{\#0}$  at 40 K as a function of magnetic fields for the five specimens. It is clearly seen that the sample pieces with higher  $\gamma_0$  *i.e.* lower level of RE/Ba substitution, such as (Y,Y)BCO and (Y,Er)BCO, exhibited larger enhancement of J<sub>c</sub> after the irradiation. Fig. 3 shows the  $J_c$  normalized by that for pristine samples at 40 K in 0.5 T as a function of irradiation dose. The enhancement of  $J_c$  for the specimens with lower  $\gamma_0$  was apparently suppressed. This tendency can be also confirmed in data at the other temperatures above 40 K. Fig. 4 shows temperature dependences of  $J_c$  in 0.5 T for #0 and #1. Jc values at 60 and 77 K were also largely enhanced by irradiation especially for (Y,Y)BCO and (Y,Er)BCO.

### 3.2. Change in critical current properties due to the difference in RE123 matrix

Difference in magnitude of increased J<sub>c</sub> implies that pinning force of introduced defects by electron irradiation depends on the RE/Ba substitution level of RE123 phase. As one of the possible origins, difference in superconducting condensation energy before irradiation can be pointed out. It can be presumed that pinning force of defects is substantially suppressed in RE123 crystal with a higher level of RE/Ba substitution because of lowered superconducting condensation energy. Therefore, introduction of additional defects by electron irradiation can be more effective for RE-BCO melt-textured bulks with strong superconductivity of RE123 phases, such as (Y,Y)BCO and (Y,Er)BCO. Kinjo et al. investigated the effect of 200 MeV proton irradiation on field-trapping properties of DyBCO melt-textured bulks and reported that samples with lower field-trapping properties showed larger improvement by the irradiation [7]. That tendency seems to be similar to that observed in this study.

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