Effect of addition of soft magnetic alloy particles on the flux trapping in Gd123 bulk superconductors

Y Xu, K Tsuzuki, Y Zhang, Y Kimura, M Izumi

1 Department of Electronics and Mechanical Engineering, Tokyo University of Marine Science and Technology, 2-1-6, Etchujima, Koto-ku, Tokyo 135-8533, Japan
2 Department of Mathematics and Physics, Shanghai Institute of Electric Power, 28 Xuehai Road, New Pudong District, Shanghai 201300, P. R. China

E-mail: d062030@kaiyodai.ac.jp

Abstract. Pinning stability and the introduction of magnetic flux pinning is an essential problem in applications of high-\(T_c\) superconductors. Study on the role of addition of a variety of metal oxides into GdBa\(_2\)Cu\(_3\)O\(_{\delta}\) (Gd123) bulk superconductors was carried out. We found that the addition of 0.05 wt. % of soft magnetic alloy particles Fe-Cu-Nb-Si-Cr-B (Fe-B) into the Gd123 contributes to the enhancement of the critical current density (\(J_c\)) under a wide range of applied magnetic fields up to 3 T. The Fe-B particles refined less than 10 \(\mu\)m by ball milling indicate no remarkable contribution on the \(J_c\) under the magnetic field. The reduction of the Ba content resulted in the appearance of a peak of \(J_c\) which has been observed in the Gd/Ba solid solution with rich Ba content. These results let us discriminate the effect of the magnetic particles from other conventional flux pinning mechanism. The peak of \(J_c\) under magnetic field was not only observed in the part along the \(c\)-axis under the seed of the sample but also in the growth sector around the periphery of the Gd123 bulk with Fe-B addition. It indicates that the magnetic particles inclusions play an important role on the homogeneous enhancement of \(J_c\) and the high flux pinning performance.

1. Introduction
Due to the small coherence length \(\xi\) of the high-\(T_c\) superconductor, the vortex core has become a nano-sized object. The flux pinning takes place at the nano-scale and the materials optimisation must be carried out on a similar or slightly larger length size [1, 2]. Accordingly, the further optimisation of the flux pinning forces and, hence, the critical current density (\(J_c\)) requires detailed measurement on an according length scale. It is well known that RE\(_2\)BaCuO\(_5\) (RE211) as the secondary phase particles dispersed in a REBa\(_2\)Cu\(_3\)O\(_{\delta}\) (RE123) phase have a strong influence on the microstructure and pinning properties of RE123 bulk materials. Many studies have shown that the employment of ultra-fine ball-milled RE211 powders resulted in a large enhancement of \(J_c\) values in RE123 system [3-6]. We have reported that the addition of 0.05 wt. % of soft magnetic alloy particles Fe-Cu-Nb-Si-Cr-B (Fe-B) into the Gd123 contributes to the enhancement of the \(J_c\) under a wide range of applied magnetic fields up to 3 T [7, 8]. Fe-B alloy is a soft magnetic material which sees the appearance of a large magnetic

---

3 To whom any correspondence should be addressed.
4 Present address: Kawasaki Heavy Industries Ltd., 1-1, Kawasaki-cho, Akashi-shi, Hyogo 673-8666, Japan
moment upon application of an external magnetic field. It is fascinating for the potential practical applications and attracts much attention. It is expected that as any introduced secondary phase particles, the smaller the magnetic powder Fe-B alloys, the bigger the role they would play in the enhancement of the flux pinning properties. To clarify the mechanism of the enhancement of \( J_c \) in this material system, we have to study from the viewpoints described as follows.

First, the size effect of the magnetic particles on the Gd123 bulk superconductor has to be studied, for which the Fe-B soft magnetic particles were prepared from their typical size of 10 \( \mu \)m -30 \( \mu \)m to less than 10 \( \mu \)m by ball milling process during 1 or 2 hours. These particles were added into the precursor phase of Gd123 bulk described in the following section.

Then, it is well known that Gd123 single grains fabricated in air tend to form Gd\(_{1+y}\)Ba\(_{2-x}\)CuO\(_{2.8}\) (\( x > 0 \)) type mixed solid solution, since the ionic radii of Gd\(^{3+}\) and Ba\(^{2+}\) ions are relatively close with each other [9, 10]. Nano-scaled solid solution has been reported as pinning centres which contribute to the second peak effect under higher magnetic fields [11]. However, the superconducting properties of samples with a higher value of \( x \) are degraded significantly compared to the compound without substitution. As a result, Ba-rich compounds such as BaO\(_2\), BaCuO\(_2\) and BaCO\(_3\) [12-15] have been added to precursor compositions in an attempt to suppress the extent of Gd/Ba substitution during melt processing of Gd123 single grains in air. But as mentioned before, we know that the RE/Ba solid solution is one of the reasons of the peak effect. In order to make clear the reason of the appearance of the peak effect after doping with the soft magnetic powder, we also prepared series of bulks without BaO\(_2\) in this study.

2. Experimental
The soft magnetic particles (MP) of Fe-B alloy, which contain a small amount of Cu-Nb-Si-Cr, were treated by ball milling for 1 and 2 hours. We then observed the magnetic particles with SEM to verify their size. Commercially available Gd\(_2\)O\(_3\) (3 N), BaO\(_2\) (3 N) and CuO (3 N) powders were used to prepare Gd123 and Gd211 powders. In order to get fine particles of Gd211, we employed ball-milling treatment for 1 hour, using Y\(_2\)O\(_3\)–ZrO\(_2\) balls. The prepared Gd123 and Gd211 were weighed in a nominal molar 5:2 ratio and mixed thoroughly together with fine magnetic particles.

We have prepared three systems. The first and second systems’ precursor compositions were studied as follows: Gd\(_{123} + 0.4\) Gd\(_{211} + 0.1\) BaO\(_2\) + x wt. \% MP (1-hour ball-milled and 2-hour ball-milled), \( x =0.0, 0.02, 0.05, 0.08, 0.12 \). The third system, precursor compositions were studied as follows: Gd\(_{123} + 0.4\) Gd\(_{211} + x \) wt. \% MP, \( x =0.0, 0.02, 0.05, 0.08, 0.12 \), without BaO\(_2\). Both 10 wt. \% Ag\(_2\)O and 0.5 wt. \% Pt were added in order to improve the mechanical properties and hinder the coarsening of Gd211 second phase particles, respectively. The mixed powder was pressed into pellets of 20 mm in diameter and 10 mm in thickness. The melt-growth in air and the post annealing in oxygen gas were performed as described in the references [16].

Superconducting properties were measured by a SQUID magnetometer (Quantum Design Co Ltd.). The DC magnetization measurement was performed under an applied magnetic field parallel to the c-axis. \( J_c \) was deduced from the magnetization measurement using the extended Bean model [17]. The measured specimens were cut from \( a \)-growth sector and \( c \)-growth sector of the single-domain bulk.

3. Results and discussion

3.1. Effect of magnetic powder on superconducting properties
Figure 1 (a) shows the magnetic field dependence of \( J_c \) at 77 K under \( B // c \)-axis of the specimens cut from the \( a \)-growth sector with different amounts of original MP doping. It can be seen that the \( J_c \) was enhanced by doping with a suitable amount of MP. Especially, it seems that the peak effect appeared in these specimens. It means that MP doping enhances the \( J_c \) both in low and high fields. In addition, figure 1 (b) also gives the \( J_c - B \) curves at 77 K under \( B // c \)-axis of the specimens cut from the \( c \)-growth sector with different amounts of MP doping. For these specimens, the \( J_c \) in high field was also enhanced with a suitable amount of MP content. Generally speaking, for the bulk samples, the peak
effect appeared in the specimens just below the seed because of the RE/Ba compositional fluctuation which was induced by the seed crystal [18-20]. At this stage, we cannot definitively conclude that the reason of the enhancement of $J_c$ in high field is the effect of the magnetic particles itself or if it can be induced by the oxygen deficiencies and the RE/Ba compositional fluctuation in the bulk. The results show the present finding with the effect of soft magnetic particle is a promising candidate to improve the flux trapping of Gd123-bulk magnets from the viewpoint of the enhancement of homogeneity and pinning force.

3.2. SEM image of the doping magnetic powders

To check the change of the magnetic particles after the ball milling, SEM images for three kinds of MP are shown in figure 2. Clearly, for the original commercial magnetic particles in figure 2 (a), the average size of the particles is about 10 µm. After employing 1-hour and 2-hour ball milling, as shown in the figure 2 (b) and (c), the average size is about 3 µm, which is smaller than the original particles. However, from the image (b) and (c), it can be found that the size of the particles is almost the same, in both 1 and 2-hour cases. To check the size effect of the magnetic particles on the superconducting properties of the Gd123 bulk, 1 and 2-hour ball-milled magnetic particles were employed to prepare series of bulk samples.

3.3 Effect of refined magnetic particles on the superconducting properties

Figure 3 (a) shows the magnetic field dependence of $J_c$ – $B$ at 77 K under $B$ // c-axis of the specimens with different amounts of 1 h MP doping. These specimens were cut from the $a$-growth sector domain, on the side of the domain. It can be seen that the $J_c$ was enhanced by doping with suitable amounts of
MP. For the refined MP doped bulks, there is no peak effect appearance in the small amount additions, like 0.02 MP 1 h. The peak effect appears in the bulks doped with higher amounts of addition, like 0.08 MP 1 h. It can be seen that in the small amount range, with the increase of the MP doping, the \( J_c \) in low field increases significantly, the maximum \( J_c \) under self field appearing in the 0.02 MP 1 h bulk. It achieves 53,000 A/cm\(^2\). However, when the addition amount was more than 0.02 wt. % to Gd123, \( J_c \) in low field decreases again. For the 0.02 MP, \( J_c \) under self field is about 3 times what it under a higher magnetic field, 1.0 T. However, for the 0.08 MP, the \( J_c \) under self field only is about 1.3 times what it is under a 1.0 T magnetic field. It means that suitable amounts of refined MP doping can enhance the \( J_c \) in the low-field region and another suitable amount of refined MP doping can enhance the \( J_c \) in the high-field region, which is a little different of the system doped with original magnetic particles.

In figure 3 (b), the magnetic field dependence of \( J_c \) for the 0.05 MP and 0.08 MP specimens with different MP ball-milling time are given. The results show that both 1 h MP doping and 2 h MP doping almost have the same effect on the superconducting properties. It can be found that the refined MP doping cannot improve the \( J_c \) of the bulk obviously. This is the contrary to the fact that small-sized secondary phase particles enhance the \( J_c \) of the bulk. This maybe a evidence support the MP is not effect as the core pinning center. It’s mechanism should be discussed in the future.

![Figure 3.](image)

**Figure 3.** (a) The magnetic field dependence of \( J_c \) at 77 K under \( B \parallel c \)-axis of the specimens cut from \( a \)-growth sector with different amounts of 1-h milled MP doping. (b) Applied field dependence of \( J_c \) for 0.05 MP and 0.08 MP with different ball milling time were employed, at 77 K, \( B \parallel c \).

### 3.4 Effect of the Ba-rich phase on the superconducting properties

Except the size effect, we have to make clear the mechanism of the appearance of the peak effect with the magnetic particles doping as mentioned above. It is well known that the RE/Ba solid solution is one of the reasons causing the peak effect in the bulk superconductor. However, this kind of uncontrolled coarsening solid solutions deteriorates the superconducting performances. As a result, Ba-rich compounds such as BaO\(_2\), BaCuO\(_2\) and BaCO\(_3\) have been added to precursor compositions in an attempt to suppress the extent of Gd/Ba substitution during the melt processing of Gd123 single grains in air. We also employed BaO\(_2\) in our studies. However, in order to make clear the reason of the appearance of the peak effect after doping with magnetic powder, we wish to distinguish the contribution from the RE/Ba solid solution and the magnetic particles additions. So, series of Gd123 bulks doped with magnetic particles but without BaO\(_2\) were studied.
Figure 4. (a) The $J_c$-$B$ properties at 77 K under $B$ / / c-axis of the specimens cut from $a$-growth sector with different amounts of MP doping and without BaO$_2$ addition. (b) Applied field dependence of $J_c$ for 0.02 MP and 0.05 MP with and without BaO$_2$ doping, at 77 K, $B$ / / c.

Figure 4 (a) shows the magnetic field dependence of $J_c$ - $B$ at 77 K under $B$ / / c-axis of the specimens with different amounts of MP doping and without BaO$_2$ doping. These specimens were also cut from $a$-growth sector domain. It can be seen that the highest $J_c$ appeared in the 0.02 MP bulks, and the peak effect also appeared. With the doping content of MP increasing to more than 0.02 wt. %, the superconducting properties were destroyed. The results showed that, the $J_c$ observed in higher field was enhanced in all the samples. It means that the peak effect appeared in all the specimens doped with MP but without BaO$_2$.

As mentioned above, our purpose is to distinguish the effects of MP doping and the Gd/Ba solid solution on the peak effect. In figure 4 (b), the magnetic field dependence of $J_c$ for the 0.02 MP and 0.05 MP specimens with and without BaO$_2$ doping were given. The results show that the peak effect appeared in both the specimens doped with BaO$_2$ and without BaO$_2$. For the specimens without BaO$_2$, the appearance of the peak effect can be attributed to the formation of the Gd/Ba solid solution. However, the peak effect of the specimens doped with BaO$_2$ is more obvious. It indicates that the magnetic-particle doping contributes to the appearance of the peak effect in $a$-growth sector of Gd123 bulk, which indicates that it improves the homogeneity of flux pinning.

3.5 Magnetic-powder doping effect on the transition temperature

The onset superconductivity transition temperature ($T_c$) of these bulks is summarized in figure 5. It can be seen that the $T_c$ of the bulk decreased with the increase of the MP doping content, from 93.0 K without doping to 89.5 K with 0.12wt. % MP doping. It demonstrates that small amounts of MP inclusions can suppress the superconductivity of the bulk but cannot completely destroy it. The $T_c$ decreases more obviously in the system without BaO$_2$ doping, which also indicates the Gd/Ba solid solution destroys the superconducting properties. In addition, in the system doped with 1-hour ball-milled magnetic particles, $T_c$ only decreases a little. It may be linked to the fact that the 1-hour ball-milled magnetic-particle doping did not enhance the peak effect so much. It should be made clear in the future.
4. Summary
To clarify the size effect of magnetic particles of Fe-B alloys on the superconductivity properties of Gd123 bulks, the doped magnetic particles were refined by ball milling so that their size became smaller. The experimental results showed that suitable amounts of the fined magnetic particles doping enhance the $J_c$ of the Gd123 bulk superconductor. However, its effect on enhancement of $J_c$ is worse than that obtained by the doping of original magnetic particles. In addition, by reducing the Ba content of the composition, we obtain the peak effect caused by the Gd/Ba solid solution in the Gd123 system, which lets us discriminate the effect of the magnetic particles from that of the RE/Ba solid solution on the peak effect. The experimental results showed that the appearance of the peak effect in the $a$-growth sector of the magnetic-particle doped Gd123 bulk could not be attributed to the Gd/Ba solid solution. The magnetic-particle inclusions played an important role to enhance the homogeneity and the high magnetic flux pinning.

5. References
[1] Haugan T, Barnes P N, Wheeler R, Melsenkothen F and Sumption M 2004 Nature 430 867
[2] Iida K, Babu N H, Reddy E S, Shi Y H and Cardwell D A 2005 Supercond. Sci. Technol. 18 249
[3] Nariki S, Matsui M, Sakai N and Murakami M 2002 Supercond. Sci. Technol. 15 679
[4] Nariki S, Sakai N, Murakami M and Hirabayashi I 2004 Supercond. Sci. Technol. 17 S30
[5] Xu C, Hu A, Sakai N, Izumi M and Hirabayashi I 2005 Supercond. Sci. Technol. 18 229
[6] Nariki S, Sakai N, Murakami M and Hirabayashi I 2004 Physica C 412–414 557
[7] Xu Y, Izumi M, Tsuzuki K, Zhang Y F, Xu C X, Murakami M, Sakai N and Hirabayashi I 2009 Supercond. Sci. Technol. 22 095009
[8] Xu Y, Izumi M, Zhang Y F and Kimura Y 2009 Physica C, 469 1215
[9] Murakami M, Sakai N, Higuchi T and Yoo S I 1996 Supercond. Sci. Technol. 9 1015
[10] Shi Y, Babu N H, Iida K and Cardwell D A 2008 Physica C 468 1408
[11] Hu A, Schatzle P, Bieger W, Verges P, Fuchs G and Krabbes G 1999 Appl. Phys. Lett. 75 259
[12] Dai J Q, Zhao Z X and Xiong J W 2003 Supercond. Sci. Technol. 16 815
[13] Babu N H, Iida K, Shi Y and Cardwell D A 2005 Appl. Phys. Lett. 87 202506
[14] Shi Y, Babu N H, Iida K and Cardwell D A 2007 Supercond. Sci. Technol. 20 38
[15] Shi Y, Babu N H, Iida K and Cardwell D A 2007 IEEE Trans. Appl. Supercond. 17 2984
[16] Hu A, Sakai N, Ogasawara K and Murakami M 2002 Physica C 366 57
[17] Chen D X and Goldfarb R B 1989 J. Appl. Phys. 66 2489
[18] Koblishka M R, van Dalen A J J, Higuchi T, Yoo S I and Murakami M 1998 Phys. Rev. B 58 2863

Figure 5. Onset $T_c$ for specimens with different amount of magnetic particles doping.
Acknowledgements
This work was partially supported by Grant-in-Aid for Scientific Research (KAKENHI,18560642) and Iwatani Foundation.

[19] Hu A, Zhou H, Chikumoto N, Sakai N, Hirabayashi I and Murakami M 2004 Supercond. Sci. Technol. 17 856

[20] Hu A, Sakai N, Zhou H, Inoue K, Chikumoto N and Murakami M 2004 Physica C 402 127