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# Optimization of Matrix Chemical Ratio for High Critical Current Density $J_c$ in (Nd, Eu, Gd)$Ba_2Cu_3O_y$ Superconductors

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### Abstract

We have investigated the magnetic field dependence of the critical current density $J_c$ in (Nd, Eu, Gd)$Ba_2Cu_3O_y$ samples with various Nd: Eu: Gd ratios in the rare earth site at 77.3 K. It was found that all the samples showed well-developed peak effect depended on the chemical ratio in the matrix. We have also showed that a maximum flux pinning can be achieved in the whole magnetic field when 3 mol% NEG-211 particles were dispersed in a (Nd, Eu, Gd)$Ba_2Cu_3O_y$ matrix with an optimum Nd: Eu: Gd ratio. The enhancement of peak critical current density $J_{cp}$ with increasing matrix ratio suggests that the maximum pinning effect can be achieved by combining the appropriate concentration of 211 phase particles with the optimum chemical ratio in the matrix.

## Introduction

The RE-$Ba_2Cu_3O_y$ superconductors (RE-rare earth element, RE-123) are a broad family of perovskites-type oxides with unique properties. The first compound of this family, Y-123 was discovered in 1987 in USA and Japan[1, 2]. Today, the family of RE-123 materials consists of all simple compounds and a rich variety of their combinations; binary, ternary or querternary ones. It became a typical representative of high-$T_c$ materials. Nd-123 single crystals exhibit systematically better pinning at high fields (a higher peak effect) than Y-123 [3,4]. The reason is solid solution that Nd forms with barium[5,6]. As a result, the Nd-123 matrix properties strongly fluctuate on a nanometer scale, which produces a pinning disorder with similar properties as a system of oxygen vacancies. This behavior is common for all rare earth atoms (RE = La, Nd, Eu, Gd, Sm ). An excessive formation of the solid solution clusters leads to deterioration of macroscopic characteristics of the material, in particular to reduction of $T_c$ and eventually to suppression of superconductivity [7]. To avoid this, RE-123 materials are usually melt-textured in a reduced oxygen atmosphere. By this technique the amount of the RE/Ba solid solution is reduced to an optimum. One of the technologies of this type, called oxygen-controlled-melt-growth (OCMG)[8], has successfully used for fabrication of ternary (Nd, Eu, Gd)-123 [9, 10, 11], melt-textured samples with superior properties. In this way, the RE-123 samples easily reach $T_c$ values as high as 93-96 K and exhibit superior pinning properties at high magnetic fields and temperatures.

Magnetic studies showed that the peak effect appearance on magnetic hysteresis loop 'MHL' is sensitive to chemical ratio in the 123 matrix [12, 13]. This indicates that optimum configuration of the superconducting matrix in the NEG-123 system is not necessarily 1: 1: 1 and optimization of the chemical ratio with respect to macroscopic superconducting characteristics is possible and of course desirable. It was also found that to each particular chemical ratio an optimum concentration of the externally added 211 phase belongs. We have thus effective tool for tailoring material properties according to requirements of the particular application.

It was reported[], that the peak effect in RE-123 materials originate from field-induced pinning from oxygen deficiency and or RE-Ba chemical fluctuation. In general, $\Delta T_c$ pinning provided by chemical fluctuation is stronger than that provided by oxygen vacancies [17, 18], because of oxygen defects deteriorate superconducting properties of the parent phase. It is also true there is a limit in the enhancement of the peak effect without deteriorating $T_c$, since it requires the introduction of a large amount of oxygen vacancies. Chemical fluctuation or distribution of RE-123 as clusters in the RE-123 matrix is sensitive to the kind of RE element involved.[19]. And therefore the control of the matrix composition is important for engineering flux pinning.

Recently, it was showed that among the RE-123 compounds, the ternary (NEG-123) bulk superconductor exhibit superior properties. Typical for these materials is a strongly developed peak effect, providing a large $J_c$ at high magnetic fields that almost reach the required engineering values at 77 K [14, 16]. However, further optimization is important for engineering applications.

Hence, in this paper, we have investigated the magnetic field dependence critical current density $J_c$ with various matrix ratio of Nd, Eu and Gd to find out the optimized matrix ratio for largest $J_c$.

## Experimental

The melt-textured samples with nominal composition of $(Nd_{0.33}Eu_{0.66-x}Gd_x)Ba_2Cu_3O_y$ where x values 0.25, 0.27, 0.28, 0.29 were fabricated by the oxygen-controlled-melt-growth (OCMG) process, under oxygen-partial pressure of 0.1 % $O_2$ and gas flow rate of 300 ml/min. The details of the sample preparation and the OCMG process were described elsewhere [20]. Next, commercial Nd-422, Eu-211 and Gd-211 powders were mixed in the ratio 1: 1: 1 to prepare NEG-211 secondary phase particles. Then, the samples were added volume fractions of 3 and 7 mol% of NEG-211 were subjected to the OCMG process. The details of the heat treatment profile can be found in [21]. The samples with dimensions of $l \times w \times d = 1.5 \times 1.5 \times 0.5\ mm^3$ were cut from the as-grown crystals and annealed in flowing $O_2$ gas in the temperature range of 300-600 º C. The magnetization in a magnetic field along the *c*-axis was measured using a Quantum Design SQUID magnetometer. The onset superconducting transition temperature of all the samples was in about 93-96 ranged. The critical current density $J_c$ was estimated from the measured magnetization hysteresis of the magnetic moment using the Bean model [22],

$$J_c = \frac{6\Delta m}{w^2 d(3l - w)},$$

where $\Delta m$ is the magnetization width, d, l, and w are the thickness, length and width respectively. The magnetization was carried out in the 0.1-7 T magnetic field range and at 77.3 K temperature. The specimens were eight (Nd, Eu, Gd)$Ba_2Cu_3O_y$ bulk oxide superconductors (NEG-123) with addition of 3 and 7 mol% NEG-211 secondary phase particles. The specifications of the specimens are listed in Table 1.

**Table 1:** Specification of Specimens.

| **No.** | **Specimens** | **Addition of 211 Secondary phase particles** |
|---|---|---|
| 1 | $(Nd_{0.33}Eu_{0.41}Gd_{0.25})Ba_2Cu_3O_y$ | 3 mol% NEG-211 |
| 2 | $(Nd_{0.33}Eu_{0.39}Gd_{0.27})Ba_2Cu_3O_y$ | 3 mol% NEG-211 |
| 3 | $(Nd_{0.33}Eu_{0.38}Gd_{0.28})Ba_2Cu_3O_y$ | 3 mol% NEG-211 |
| 4 | $(Nd_{0.33}Eu_{0.37}Gd_{0.29})Ba_2Cu_3O_y$ | 3 mol% NEG-211 |
| 5 | $(Nd_{0.33}Eu_{0.41}Gd_{0.25})Ba_2Cu_3O_y$ | 7 mol% NEG-211 |
| 6 | $(Nd_{0.33}Eu_{0.39}Gd_{0.27})Ba_2Cu_3O_y$ | 7 mol% NEG-211 |
| 7 | $(Nd_{0.33}Eu_{0.38}Gd_{0.28})Ba_2Cu_3O_y$ | 7 mol% NEG-211 |
| 8 | $(Nd_{0.33}Eu_{0.37}Gd_{0.29})Ba_2Cu_3O_y$ | 7 mol% NEG-211 |

## Results

The DC magnetization measurements is investigated for a group of specimens of NEG-123 superconductors with changing matrix chemical ratio such as Eu/Gd ratio by SQUID magnetometer, which as shown in Fig. 1. The highest magnetization width $\Delta m$ was found for $(Nd_{0.33}Eu_{0.39}Gd_{0.27})Ba_2Cu_3O_y$ specimen with addition of 3 mol% NEG-211 particles. The magnetic field dependence of critical current density $J_c$ is estimated from the hysteresis measurements by using the Bean model, which as shown in Fig. 2. It is found that $J_c$-$B$ characteristics changes with changing the matrix ratio such as the ratio of Eu/Gd. The highest peak critical current density $J_{cp}$ is observed for sample-2 of $(Nd_{0.33}Eu_{0.39}Gd_{0.27})Ba_2Cu_3O_y$ specimen with addition of 3 mol% NEG-211 particles and it is about $1.77\times10^9$ A/m$^2$ at higher magnetic field of 2.5 T.

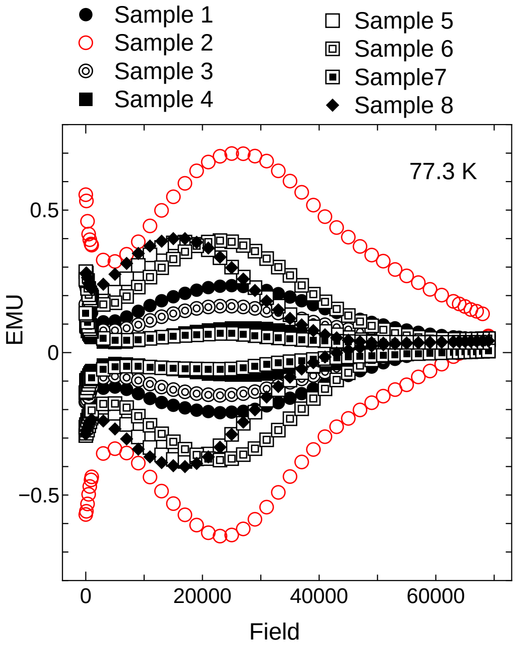

**Figure 1:** The magnetic field dependence magnetization for NEG-123 samples with varying Eu/Gd contents at 77.3 K.

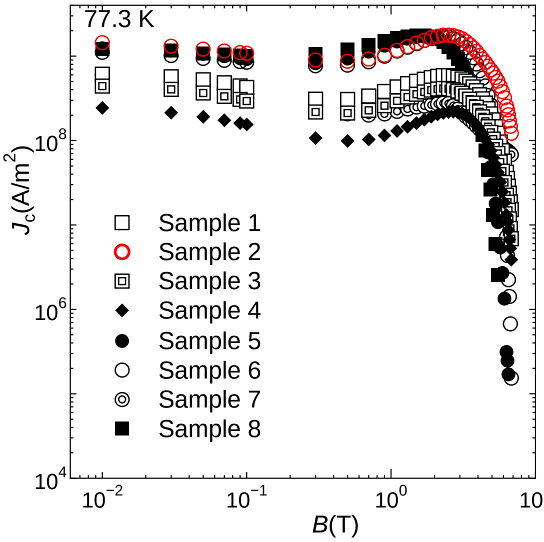

**Figure 2:** Field dependence critical current density $J_c$ for NEG-123 samples with varying Eu/Gd contents at 77.3 K.

## Discussion

In this work, we have measured the magnetic field dependence critical current density $J_c$ of NEG-123 samples with addition of 211 particles, we can called two sets of specimens. In the first set (sample 1-4), NEG-123 superconductors with addition of 3 mol% and in the second set (sample 5-8) with addition of 7 mol% NEG-211 secondary phase particles. In the both sets, the amount of Nd is fixed and the ratio of Eu and Gd are changed.

The Figure 2 shows the $J_c$-$B$ characteristics for 8 specimens of NEG-123 samples at 77 K for fields applied along the c-axis. In all cases, Nd was kept constant and the ratio of Eu:Gd varied from 1.24-1.64. It is evident that all the samples show well-developed peak effect with the peak position depended on the chemical ratio in the matrix, i.e., when Eu/Gd ratio changed, the peak critical current density $J_{cp}$ also changed. The amount of Eu increases from 0.37 to 0.41 while Gd decreases from 0.29 to 0.25, the $J_{cp}$ increases suggests that the presence of higher amount of Eu favors high $J_{cp}$. The discrepancy of $J_{cp}$ values by changing ratio of Eu/Gd suggests that a proper combination of RE elements in the ternary NEG-123 compound is effective in controlling flux pinning. A control of the Nd:Eu:Gd ratio in the NEG-123 matrix is an effective way in tailoring the pinning properties of this compound. A proper choice of the Nd:Eu:Gd ratio leads to the control of $J_{cp}$ at 77 K.

It is known that 211 particles act as strong pinning centers. In addition, it has been reported [23] that the pinning mechanism of normal 211 particles is the condensation energy interaction with the pinning force which decreases monotonically with increasing magnetic field. Hence, the pinning by 211 particles does not directly play the role in increasing $J_{cp}$. Another possibility of the peak effect by 211 particles is the order-disorder transition of the flux lines, i.e., a transitional change of flux lines from the ordered state with a low $J_c$ to the disordered state with a high $J_c$. The enhancement of the pinning efficiency at the peak effect is considered to be caused by the softening of elasticity of flux lines. It was shown [24] that the peak effect in a single crystal $NdBa_2Cu_3O_y$ superconductor is caused by the order-disorder transition of flux lines under the flux pinning by substituted regions with lower-$T_c$. For occurrence of the order-disorder transition, however, the pinning centers should be sufficiently small that an appropriate variation in the pinning efficiency could be realized by a slight deformation of flux lines. This requires a pinning potential quickly varying in space. On the other hand, the size of 211 particles is much larger than the flux line spacing. Hence, the 211 particles do not seem to contribute to the peak effect also from a softening of flux lines for the disorder transition.

In addition, in the present case we get higher $J_{cp}$ in sample 3mol% NEG-211 phase particles instead of sample higher 7 mol% addition. The enhancement of $J_{cp}$ with increasing matrix ratio suggests that the maximum pinning effect can be achieved by combining the appropriate concentration of 211 phase particles with the optimum chemical ratio in the matrix.

## Conclusion

The investigation of $J_c$-$B$ characteristics in NEG-123 materials with addition of NEG-211 particles of 3 and 7 mol% for the various matrix ratio at 77.3 K suggests that the proper choice of the Nd: Eu: Gd ratio leads to the control of peak effect that is, peak critical current density $J_{cp}$ and the maximum pinning effect in a broad field range can be achieved by combining the appropriate concentration of 211 particles with the optimum ratio of Nd: Eu: Gd in the matrix.

## Reference

[1] Wu M. K., Ashburn J. R., Torng C. J., Hor P. H., Meng R. L., Huang Z. J., Wang Y. Q., Chu C. W., Phys. Rev. Lett., 58, 908 (1987).
[2] Hikani S. et al., Jpn. J. Appl. Phys., 26, L314 (1987).
[3] Koblischka M. R., Van Dalen A. J., Higuchi T., Sawada K., Yoo S. I., Murakami M., Phys. Rev. B54, R6893 (1996).

[4] Higuchi T., Yoo S. I., Murakami M., Phys. Rev. B59, 1514 (1999).

[5] Egi T., Kuroda K. Unoki H., Koshizuka N., Appl. Phys. Lett., 67, 2406 (1995).

[6] Chikumoto N., Yoshioka J., Murakami M., Physica C, 291, 79 (1997).

[7] Hasegawa T., Kishio K., Kitazawa K., Fueki K., in: High Temperature Superconductivity (Ed. R. M. Met) pp 37-51, Gordon & Breach, London (1988).

[8] Muralidhar M. Chauhan H. S., Saitoh T., Segawa K., Kamada K., Murakami M., Physica C, 282-287, 503 (1997).

[9] Muralidhar M. Chauhan H. S., Saitoh T., Segawa K., Kamada K., Murakami M., Supercond. Sci. Technol., 10, 663 (1997).

[10] Muralidhar M., Jirsa M., Sakai N., Murakami M., Appl. Phys. Lett., 79, 3107 (2001).

[11] Muralidhar M., Sakai N., Chikumoto N., Jirsa M., Machi T., Nishiyama M., Hu Y., Murakami M., Phys. Rev. Lett., 89, 2370011 (2002).

[12] Muralidhar M., Koblischka M. R., Murakami M., Physica C, 313, 232 (1999).

[13] Muralidhar M., Murakami M., in: Studies of high temperatures Superconductors, (Ed. A. V. Narlikar) 41, 105 (2002).

[14] Yokoyama K. Oka T., Okada H., Noto K., Physica C, 392-396, 739 (2003).

[15] Wang S., et al., IEEE. Trans. Applied Superconductivity, 13, 2134 (2003).

[16] Murakami M., Supercond. Sci. Technol., 5, 185 (1992).

[17] Ohara T., Kumakura H., Wada H., Physica C, 13, 1272 (2001).

[18] Day A. C. et. al., Supercond. Sci. Technol., 15, 838 (2002).

[19] Tanaka S., Physica C, 392-396, 1 (2003).

[20] Muralidhar M., Murakami M., Supercond. Sci. Technol., 13, 131 (2000).

[21] Muralidhar M., Koblischka M. R., Saitoh T., Murakami M., Supercond. Sci. Technol., 11, 1349 (1998).

[22] Gyorgy E. M., Van Dover R. M., Jackson K. A., Schneemayer L. F., Waszczak J. V., Appl. Phys. Lett., 55, 283 (1989).

[23] Hasan M. N., Kiuchi M., Otabe E. S., Matsushita T., Muralidhar M., Supercond. Sci. Technol. 20, 345 (2007).

[24] Hasan M. N., Kurokawa T., Kiuchi M., Otabe E. S., Matsushita T., Chikumoto N., Machi T., Muralidhar M., Murakami M., Physica C 426-431, 295 (2005).