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## The study of melt growth temperature for synthesizing NdEuGd-Ba-Cu-O bulk superconductor

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### Abstract

Recently, bulk superconductors are widely fabricated due to the fact that materials exhibit various attractive properties, such as zero electrical resistance, stable levitation effect, very high magnetic field trapping and so on. The bulk superconductors are synthesized mainly in RE-Ba-Cu-O with melt texture growth process. The RE is a single element of rare earth, for example, Yttrium, Samarium, Gadolinium, Neodymium, and etc. A modern technique for melt texturing growth process is used with seed crystal placed on the top surface either in room temperature or hot seeding during the melt process. Scientists and Materials engineer are pursuing the better quality of bulk superconductor every day. However, only single RE element in barium cuprate system is not enough, then ternary rare earth elements bulk superconductors are therefore developed in barium cuprate system that exhibits superior properties, for instance, critical current density, and magnetic field trapped ability in comparison to single RE materials. However, the melt processing conditions for a large single domain of ternary bulk superconductor are undergoing study. The homemade initial powders of  $(\text{Nd}_{0.33}\text{Eu}_{0.33}\text{Gd}_{0.33})\text{Ba}_2\text{Cu}_3\text{O}_{7-y}$  (NEG-123) and  $(\text{Nd}_{0.33}\text{Eu}_{0.33}\text{Gd}_{0.33})_2\text{BaCuO}_5$  (NEG-211) were synthesized. The powders were mixed of NEG-123:NEG-211 for 10:4 molar ratio. The well mixed powders were pressed uni-axially to a cylindrical shape of 20 and 30 mm. in diameter at about 100 MPa. The optimal melting temperature was studied at 1060, 1070, and 1080°C. The slow cooling rate was fixed at 0.3°C/h. Following the melt growth process, an MgO [100] seed crystal was used and placed on the top surface of bulk precursor before it was subjected to the box furnace melting in air at atmospheric pressure. In this work, we successfully fabricated a single domain NEG-Ba-Cu-O bulk with optimum melting temperature. After the melt growth process, a bulk sample was chosen to observe for growth morphology following by M-H hysteresis measurement using Physical Properties Measurement System (PPMS) with Superconducting Quantum Interference Device (SQUID) magnetometer under cryogenic temperature. This research is therefore under collaboration of university in Thailand and Japan especially for advance measuring system. The sample demonstrated critical current density of 70,000 A/cm<sup>2</sup> at 10K.

**Keywords:** Ternary bulk superconductor, NEG-Ba-Cu-O, Top seed melt growth, Critical current density.

### 1. Introduction

Recently, bulk superconductors are widely fabricated due to the fact that materials exhibit various attractive properties, such as zero electrical resistance, stable levitation effect, very high magnetic field trapping and so on. The bulk superconductors are synthesized mainly in RE-Ba-Cu-O with melt texture growth process. The RE is a single element of rare earth, for example, Yttrium, Samarium, Gadolinium, Neodymium, and etc. A modern technique for melt texturing growth process is used with seed crystal placed on the top surface either in room temperature or hot seeding during the melt process. Scientists and Materials engineer are pursuing the better quality of bulk superconductor every day. However, only single RE element in barium cuprate system is not enough, then ternary rare earth elements bulk superconductors are therefore developed in barium cuprate system that exhibits superior properties, for instance, critical current density, and magnetic field trapped ability in comparison to single RE materials. However, the melt processing conditions for a large single domain of

ternary bulk superconductor are undergoing study.

A ternary bulk NEG-Ba-Cu-O demonstrates high critical current density ( $J_c$ ) than the single RE-Ba-Cu-O bulk superconductors. In general, the ternary bulk exhibits the  $J_c$  over 50 kA/cm<sup>2</sup>[1]. Highly critical current density is essential for engineering applications such as, magnetic drug delivery system, magnetic bearing, superconducting motor and so on [2-3]. Furthermore, a high quality bulk superconductor is needed to fabricate in a single domain as large as possible. The top seeding melt growth process (TSMG) is now commonly used in synthesizing bulk superconductor. It could be classified into two main methods which are hot seeding and cold seeding. The hot seeding is the process which needs to place a seed crystal on the top surface of the bulk when the temperature is above the peritectic temperature. That means the seed must be placed on the top at the temperature above 1000°C. As for this reason, it pushed the process difficulty and dangerous. Moreover, the seed needs to be placed one by one to the bulk which hot seeding technique is not applicable

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for mass production. In contrast, cold seeding method is one that the seed could be easily placed at the center of the bulk in room temperature. Further that cold seeding is applicable for several bulks in a batch production. However, the melt growth process condition is varied depending on the initial chemical composition, rare earth elements, bulk size and seed type. As for ternary NEG-Ba-Cu-O, a large bulk was not yet study broaden as likely Y-Ba-Cu-O bulk. So it still needs to optimal study for the melt processing condition in various aspects.

In this work, it is continuous studied of top seed melt growth process in air with cold seeding technique [4]. The optimum melting temperature could be systematical studied with MgO [100] seed crystal. The bulk samples could be characterized in bulk morphology, microstructure and magnetic properties.

### 2. Experimental

Top-Seed Melt Growth (TSMG) with cold seeding method was used throughout this study. Starting powders were commercially used of high purity 99.9% of Nd<sub>2</sub>O<sub>3</sub>, Eu<sub>2</sub>O<sub>3</sub>, Gd<sub>2</sub>O<sub>3</sub>, BaO<sub>2</sub> and CuO powders. The well mixed powders were calcined to produce (Nd<sub>0.33</sub>Eu<sub>0.33</sub>Gd<sub>0.33</sub>)Ba<sub>2</sub>Cu<sub>3</sub>O<sub>7-y</sub> (NEG-123) and (Nd<sub>0.33</sub>Eu<sub>0.33</sub>Gd<sub>0.33</sub>)<sub>2</sub> BaCuO<sub>5</sub> (NEG-211). NEG-123 and NEG-211 powders were then weighted and mixed to 10:4 molar ratio. The mixed powder was pressed uni-axially at 100 MPa to obtain bulk precursor of 30 mm. in diameter weighted 40 g for each bulk. The compact precursor was placed on the top surface with MgO [100] seed crystal at a room temperature. The precursor was subjected to melt in a box furnace following the thermal profile as shown in Fig.1.

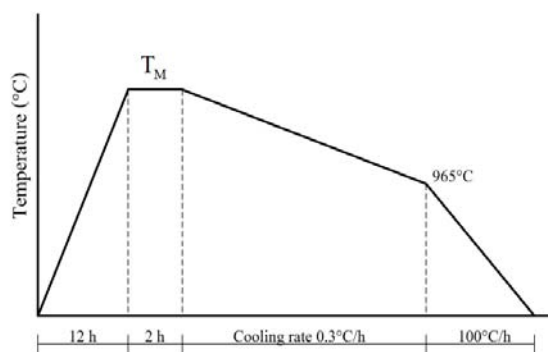


Fig. 1 Temperature profile of melt growth process

The sample was melted at a designated melting temperature ( $T_M$ ). Basically, the melting temperature could be several tens degree above the peritectic temperature ( $T_P$ ). In the previous work, the peritectic temperature was obtained at about 1050°C using TGA method [5]. Then the melting temperature was selected to study at 10, 20 and 30°C above peritectic temperature. Therefore,  $T_M$  was specified at 1060,

1070 and 1080°C. Resulted bulk samples were visually observed for growth morphology. A selected sample was crushed into a tiny piece of 2x2x1 mm<sup>3</sup> for measuring critical temperature ( $T_c$ ) and magnetic hysteresis using Physical Properties Measurement System with Superconducting Quantum Interference Device (PPMS-SQUID).

Critical current density ( $J_c$ ) was calculated from magnetic hysteresis (M-H) regarding the equation as shown below.

$$J_c = \frac{20 \times \Delta M}{a^2 d (b - \frac{a}{3})}$$

when  $J_c$  = critical current density (A/cm<sup>2</sup>)  
 $a \times b \times d$  = specimen width x length x height (cm)  
 $\Delta M$  = Difference of magnetic hysteresis (emu)

### 3. Results and discussion

The bulk samples came out of the furnace with  $T_M$  as 1060°C, 1070°C and 1080°C as shown in Fig. 2 (a)-(c). The sample in Fig. 2(a) was fabricated at maximum temperature of 1060°C. The sample morphology shows that the bulk surface consisted of small multi grain growth which reflected to inadequate melting temperature [6]. Fig. 2(b) was at 1070°C demonstrated more melting of solid-liquid solution in peritectic reaction then the liquid phase was solidified again during slow cooling across the peritectic temperature. At 1070°C, larger grain growth could be observed but the growth sector lines were interrupted by nucleation from outer rim of the bulk. Fig. 2(c) was the bulk at melting temperature of 1080°C which the sample morphology exhibited large and clear growth sector lines. A single domain growth of ternary bulk NEG-Ba-Cu-O could be obtained at 1080°C to ensure that the bulk was completely decomposed into solid-liquid solution in peritectic reaction. In order of quality, the bulk samples at 1060 and 1070°C were crushed into a tiny specimen of about 2x2x1 mm<sup>3</sup>. The specimen was randomly picked from the cloud to measure the critical temperature and magnetic hysteresis using PPMS-SQUID.

As a result, Fig. 3 shows the critical temperature which demonstrated superconductivity at on-set temperature of about 40 and 60K of the sample at 1060 and 1070°C respectively. The higher  $T_c$  is the better use in practical application. Then the specimen was soaking in cryogenic temperature to measure the magnetic responsive with variation of external magnetic field so called M-H loop. The difference of magnetic responsive was calculated to acquire the critical current density in respect to external applied field. The calculated critical current density of a sample at 1070°C is plotted in varied of temperature from 10-40K as shown in Fig. 4.

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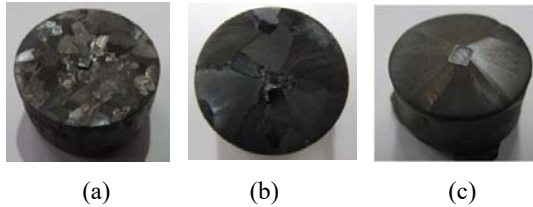


Fig. 2 The bulk sample at melting temperature of (a) 1060°C, (b) 1070°C, and (c) 1080°C

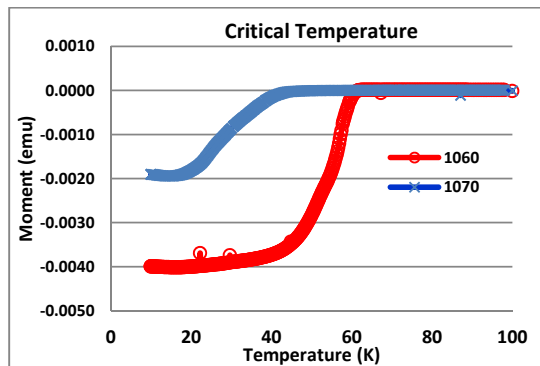


Fig. 3 Critical Temperature measured by PPMS-SQUID

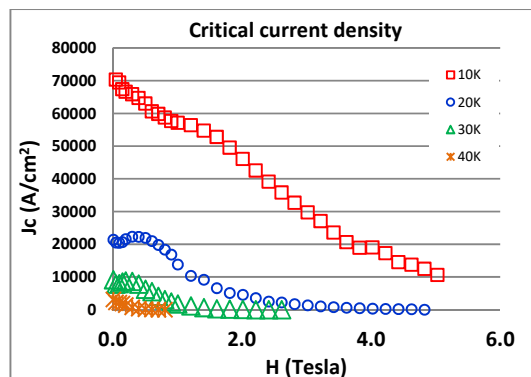


Fig. 4 Critical Density measured by PPMS-SQUID

Obviously, the critical current density can be seen that the lower temperature is the higher  $J_c$ . The maximum critical current density is obtained in Meissner state at nearly zero applied field which exhibits maximum  $J_c$  of about 70,000 A/cm<sup>2</sup> at 10K. For instance, the specimen demonstrates critical current density higher than conventional conductor about 140 times which a copper wire has critical current density of about 500 A/cm<sup>2</sup>.

### 4. Conclusion

The optimal melting for synthesizing of ternary NEG-Ba-Cu-O bulk superconductor has been studied with cold top seeding melt growth technique using MgO single crystal as a seed. The study resulted that the optimum melt temperature was obtained at 30°C above peritectic temperature which was at 1080°C for

a bulk precursor size of 30 mm in diameter. A single domain bulk was successfully synthesized at the optimum temperature with good growth morphology. The specimen was selected to characterize by PPMS-SQUID for measurement of critical temperature and magnetic hysteresis. The sample demonstrated superconductivity below the on-set temperature of 60 K. The maximum critical current density was achieved at 70,000 A/cm<sup>2</sup> in constant temperature of 10K.

### 5. Acknowledgement

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### 6. References

- [1] Muralidhar, M., Nariki, S., Jirsa, M., and Murakami, M. (2003). Superconductors for high-field use at 77K, *Physica C*, vol.392-396, October 2003, pp. 567-570.
- [2] Muralidhar, M., Tomita, M., Suzuki, K., Jirsa, M., Fukumoto, Y., and Ishihara, A. (2010) A low-cost batch process for high-performance melt-textured GdBaCuO pellets, *Superconductor Sci. Technol.*, vol.23, April 2010, pp. 040533.
- [3] Muralidhar, M., Nariki, S., Jirsa, M., and Murakami, M. (2002). Single-domain NEG-123 bulk superconductors for high magnetic field applications, *Physica C*, vol.372-376, August 2002, pp. 1134-1136.
- [4] Suzuki, K., Muralidhar, M., Koshizuka, N., Inoue, K., Murakami, M. (2014). Development of Melt-processed (Nd,Eu,Gd)Ba<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub> Superconductors in Air, *Physics Procedia* vol.58, October 2014, pp.70-73.
- [5] Kikuchi, T., Wongsatanawarid, A., Homma, Y., Suzuki, K., Koshizuka, N., and Murakami M. (2012). Processing conditions for (Nd, Eu, Gd)-Ba-Cu-O ternary bulk superconductors, *Physics Procedia*, vol.27, April 2012, pp.132-135.
- [6] Iida, K., Babu, N.H., Pathak, S., Shi, Y., Yeoh, W.K., Miyazaki, T., Sakai, N., Murakami, M., and Cardwell, D.A. (2008). Optimum processing conditions for the fabrication of large single grain Ag-doped YBCO bulk superconductors, *Materials Science and Engineering B*, vol.151, June 2008, pp. 2-6.