MPMG법을 이용한 YBaCuO 초전도체의 임계특성에 영향을 미치는 파라미터

論 文
48C - 4 - 1

Parameters to Affect the Critical Characteristics of YBaCuO Bulk Prepared by MPMG

姜亨坤* · 林成勳* · 朴晟鎭** · 韓秉誠***
(Hyeong-Gon Kang, Sung-Hun Lim, Sung-Jin Park and Byoung-Sung Han)

Abstract - This paper shows the parameters to affect the critical characteristic of YBaCuO superconducting bulk fabricated by MPMG (Melt Powdered Melt Growth)process. In order to investigate proper processing variables, the effect of the holding time at the melting temperature and that of the slow cooling time in O_2 ambient on the J_c were experimented. And then with the above obtained heat treatment conditions, the effects of addition of Y_2BaCuO_5 and Ag on the J_c were also investigated. A proper slow cooling time yields phase transformation from Tetragonal ($YBa_2Cu_3O_6$) to Orthorhombic ($YBa_2Cu_3O_7$) during an annealing time in O_2. Ag addition plays a role in increasing the T_c and the J_c, but the magnetization decreases. The J_c and the magnetization increase with addition of Y211. The Jc of the sample added Ag 10wt% is superior over 3000 G. Proper holding time, slow cooling time and amount of impurity addition are important parameters in fabricating the YBaCuO bulk by MPMG process with high J_c.

Key Words : MPMG(Melt Powdered Melt Growth), slow cooling time(서냉시간), holding time(유지시간), magnetization(자화), $YBa_2Cu_3O_7$,(고온상) Y_2BaCuO_5 (저온상)

1. Introduction

Since the discovery of the high-T_c superconductor in 1987, intensive efforts have been made to understand the mechanism of superconductivity and find a way to apply it to mechanical and electrical devices operated in liquid nitrogen temperature. The important characteristics for application of high-T_c superconductors are the Meissner effect and high critical current density(J_c). So, application techniques such as wiring and thin film using High-Tc superconductor have been reported in many papers[1-3]. Especially, it is important to fabricate the bulk with large current density. The YBaCuO bulk fabricated by the solid-state reaction method has low current density because of its micro crack, weak link, and nonuniformity[4]. In order to solve these problems, various melting processings such as MTG (Melt-Textured-Growth).

QMG (Quench-Melt- Growth) and MPMG (Melt-Powdered-Melt-Growth) have been developed[5-7]. These melting processings can form strongly pinned impurity phases or non-superconducting crystalline phases by peritectic reaction.

We fabricated YBaCuO bulk by the MPMG process which has the more effective distribution of non-superconducting crystalline phases than any other melting method. Several parameters have been considered to be very important in determining J_c in bulk sintered YBaCuO. Especially, in this paper, the effect of the holding time at the melting temperature and that of the slow cooling time in O_2 ambient on the J_c were experimented in samples with various composition. And, the effect of Ag addition and Y211 addition on critical characteristics in Y123 were also experimented.

It was suggested by the measurements of X-ray, SEM, J_c and magnetization that holding time at melting temperature, slow cooling time and impurity such as Y_2BaCuO_5 or Ag play a major role in improving its critical characteristic.

*正 會 員 : 全北大 電氣工學科 博士課程
**正 會 員 : 全北大 電氣工學科 博士 卒業
***正 會 員 : 全北大 電氣工學科 教授
接 受 日 字 : 1998年 6月 5日
最 終 完 了 : 1999年 2月 10日

2. Experimental procedures

Powders of $YBa_2Cu_3O_x$ (Y123) and Y_2BaCuO_5 (Y211) were mixed at the ratios of 1:0.0, 1:0.2, and 1:0.4, respectively. Some 1:0.4 powder was then seperated. To one group, 10 wt.% Ag_2O was added. To another group, 20 wt.% Ag_2O was added. These 5 prepared kinds of powders were melted at 1410 ℃ for 40 minutes and quenched. And after grinding these powders for 3 hours, the pulverized powders were pelletized. Firstly, in order to investigate the influence of holding time on the J_c in the proper melting temperature, the prepared pellets with $YBa_2Cu_3O_x$:Y_2BaCuO_5 ratios of 1:0.4 and no Ag addition, were seperated into 5 samples maintained at 1120 ℃ for 0, 10, 20, 40, and 80 minutes, respectively.

Secondly, to investigate the effect of slow cooling time on the J_c, the pellets with 20 wt% Ag addition were cooled quickly from 1120℃ to 1000℃ to produce Y123 from Y211 + liquid phase. After that, the pellets were cooled slowly to 950℃ at times of 0, 15, 40, and 80 hours. As the last heat treatment for the phase transformation, all pellets from all five samples were annealed at 500℃ for 24 hours in O_2.

Influences of the holding time at the melting temperature and the slow cooling time were measured by X-ray, SEM and J_c. In addition, in order to investigate the influence of added impurities such as Ag and Y211 on the J_c, the critical characteristics of pellets were investigated by M-H curve. And for the study of the flux pinning effect on impurity phases, the magnetizations dependent on the applied magnetic field at 77K and on the temperature in a 20 Gauss were compared.

3. Results and discussion

Figure 1 shows the Jc with holding time. The critical current density measured by 4 probe method was highest in 20 minutes. In the holding time more than 20 minutes, the J_c of samples was not inclined to increase. This result has relation to formation and distribution of non-superconducting crystalline phases. Long holding time prevents Y211 from being formed from Y_2O_3 + liquid phase. In fig 2, As the holding time increases, the pinlike phases of Y211 are covered with the increased liquid phase and changed to amorphous phases. It is also seen in the X-ray diffraction of Fig.3. The intensity of the Y123 peaks decreases with the increase of holding time. Critical values of sample were measured after quenched to 1000℃ and then slowly cooled down to 950℃.

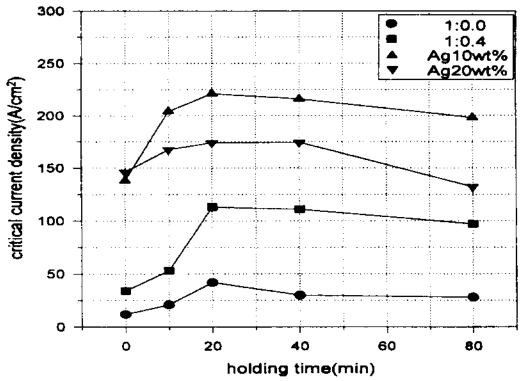

Fig. 1 The critical current density dependent on various holding times at the melting temperature of 1120℃

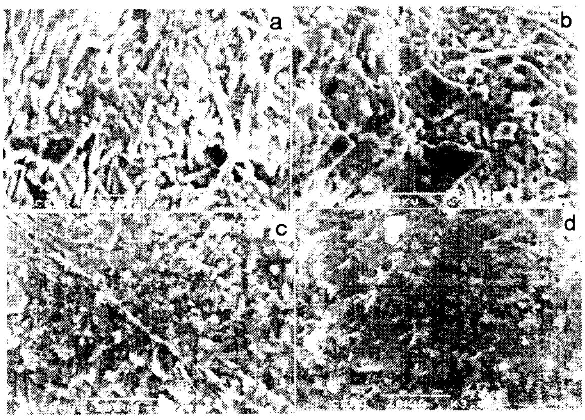

Fig. 2 SEM photographs of fracture surface of superconducting bulks dependent on various holding times at the melting temperature of 1120℃.($YBa_2Cu_3O_x$: Y_2BaCuO_5 = 1 : 0.4) (a) 0min (b) 10min (c) 20min (d) 40min

Figure 4 shows the surface morphology of the bulks at various slow cooling times from 1000 ℃ to 950 ℃ after being quickly cooled down from 1120℃ to 1000 ℃ in order to form Y123 from Y211 + liquid phase. The stripes which indicate phase transformation do not appear when the cooling time is 15 hours, but do begin to appear when it is 40 hours. And these stripes are clearer than those that appear at 80 hours. Tetragonal non-superconductor($YBa_2Cu_3O_6$) formed from insufficiency of oxygen is appeared after MPMG process. Therefore, the bulk needs sufficient cooling time in O_2 ambient in order to be orthorhombic superconductor ($YBa_2Cu_3O_7$). As a result, a slow cooling time can be seen to affect the phase transformation from Tetragonal to Orthorhombic during 40 hours annealing time

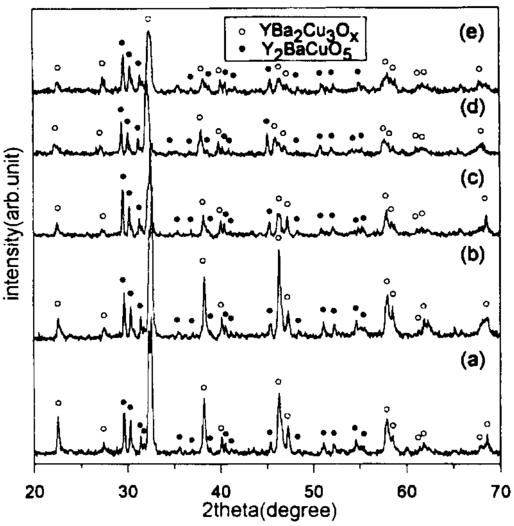

Fig. 3 X-ray diffraction patterns of superconducting bulks dependent on various holding times at the melting temperature of 1120℃ ($YBa_2Cu_3O_x$: Y_2BaCuO_5 = 0.4)
(a)0min (b)10min (c)20min (d)40min (e)80min

. Figure 5 shows that J_c is dependent on the slow cooling time. The highest J_c appeared at 40 hours as expected.

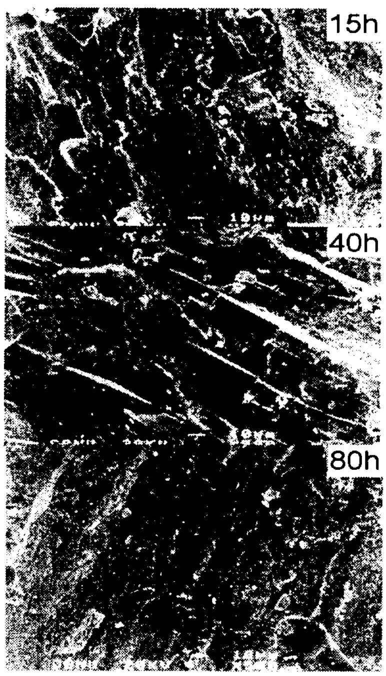

Fig. 4 SEM photographs of fractured surfaces of superconducting bulks dependent on slow cooling time from 1000℃ to 950℃.
($YBa_2Cu_3O_x$: Y_2BaCuO_5 = 1 : 0.4)

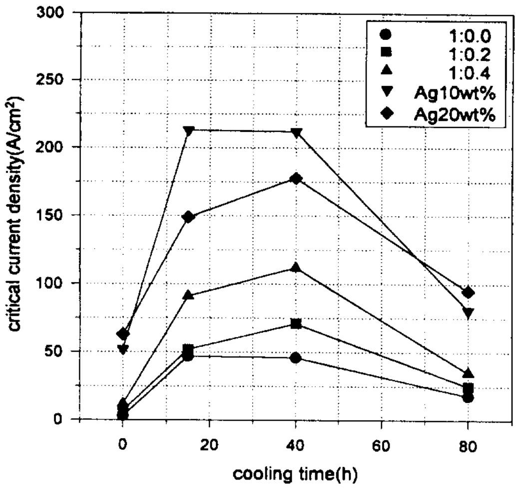

Fig. 5 Critical current densities of superconducting bulks resulted from a slow cooling time

In the next experiment, the effects of the Y_2BaCuO_5 and Ag addition on the J_c and on the flux pinning were investigated. In order to study the flux pinning effect, the temperature dependence of magnetization at an applied magnetic field of 20 Gauss was measured. Then sample size was 1.5x1.5x 1.5 mm^3. And as shown in Fig. 6(a), the magnetization and the critical temperature improved as the amount of Y211 increased in the ZFC step. In the case of addition of Ag(Fig. 6(b)), the critical temperature approached almost 90K, but the magnetization decreased. This result shows that proper Ag addition plays a role in increasing T_c, but the magnetization decreases. At the FC step, the magnetization values of all specimens, approached zero as the temperature decreased due to the flux confined to the impurity phase.

Figure7 shows the M-H (Magnetization -Hysterisis) curve measured at 77K, which has a retroprogressive characteristic. The difference between negative and positive magnetization increased as Y211 or Ag was added to Y123. The J_c in a superconducting grain was calculated by the Bean's critical state model (or magnetization method) followed as[8,9].

$$J_c = 30\triangle M/d \qquad (1)$$

where $\triangle M$ is the difference between negative and positive magnetization and d is an average diameter of a grain. Then, the average size of an observed grain in all samples with Y211 and Ag was 80-100 ㎛. The result calculated was shown in Fig. 8.

In the case of YBCO sample fabricated by the solid state reaction, the J_c decreases as the applied magnetic field increases, but the J_c in this sample fabricated by MPMG process slowly decreased as the amount of impurity increased.

from $0.69\times10^{3}\sim0.16\times10^{5}$ A/cm^2 at 1000G to $0.204\times10^{3}\sim0.114\times10^{4}$ A/cm^2 at 11000 G.

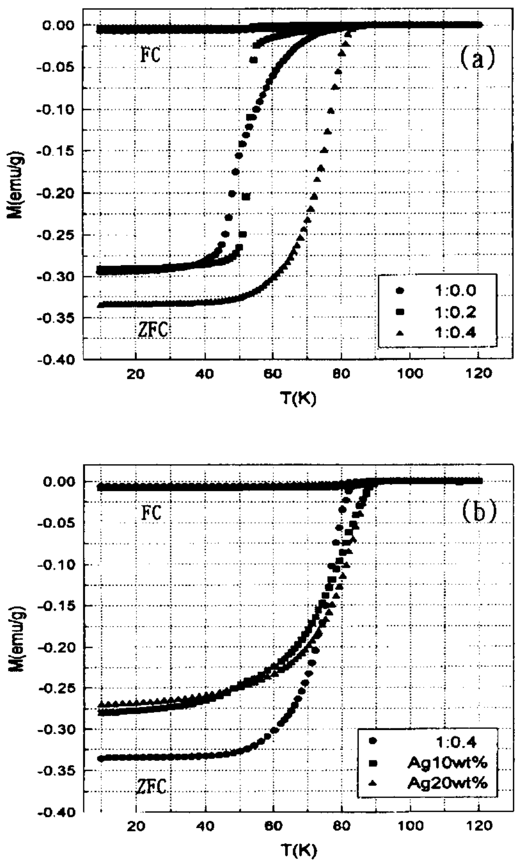

Fig. 6. Temperature dependence of magnetization measured in an applied magnetic field of 20 G
(a) Temperature dependence of magnetization due to Y_2BaCuO_5 addition
(b) Temperature dependence of magnetization due to Ag addition

4. Conclusions

The critical characteristic of bulk fabricated by the MPMG process is affected by the holding time, the cooling time and the amount of impurity.

A proper holding time forms large grains and textural structures in the MPMG process. A proper slow cooling time yields phase transformation from Tetragonal ($YBa_2Cu_3O_6$) to Orthorhombic ($YBa_2Cu_3O_7$) during 40 hours annealing time in O_2. Ag addition plays a role in increasing the T_c and the J_c, but the magnetization decreases. The J_c and the magnetization increase with addition of Y211. Especially, the Jc of the sample added Ag 10wt% is superior in over 3000 G.

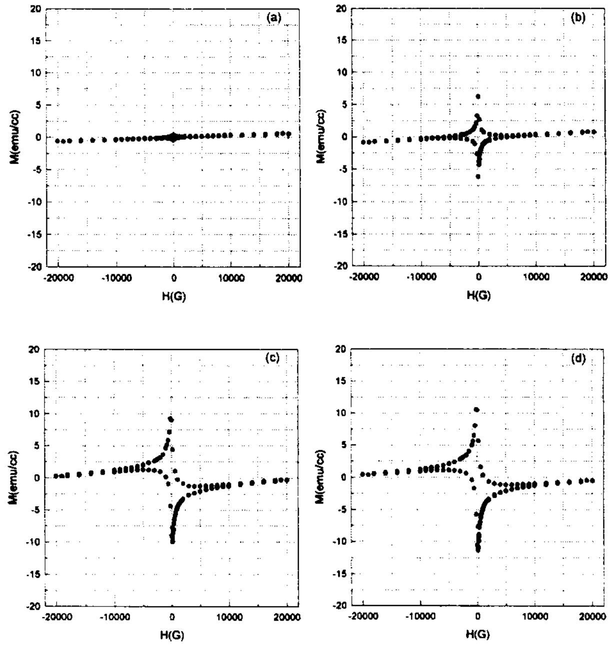

Fig. 7. Magnetization hysteresis curve with varying amounts of Y_2BaCuO_5 and Ag at 77K
(a) $YBa_2Cu_3O_x$: Y_2BaCuO_5 = 1 : 0.0
(b) $YBa_2Cu_3O_x$: Y_2BaCuO_5 = 1 : 0.4
(c) Ag10wt% (in Y123 : Y211=1:0.4)
(d) Ag20wt% (in Y123 : Y211=1:0.4)

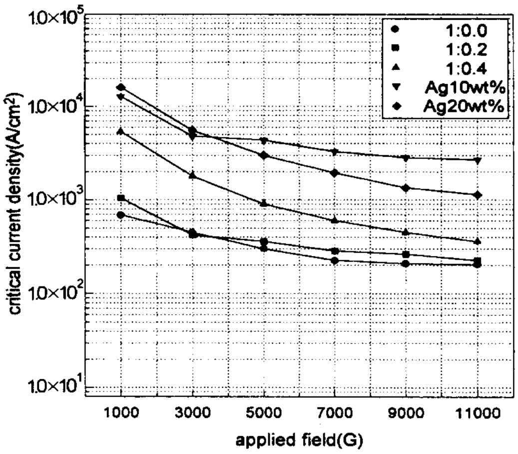

Fig. 8. The critical current density along applied magnetic field at 77K

References

1) J. Azero, L. et al., "Current limiter based on Melt processed YBCO bulk superconductors", IEEE Trans. on Applied superconductivity, Vol. 5, No.2, June, 1996.
2) Michiya Okada, et al., "Bi-2211/Ag Superconducting Insert Magnet for High Magnetic Field Generation over 22 T", Jpn. J.

Appl., Vol. 35, pp. L623-L626, May, 1996.

3) 김제하, 한석길, 강광용, "고온 초전도 $YBa_2Cu_3O_{7-x}$박막으로 구현한 오픈-스터브선방식의 다극저역통과필터의 마이크로파 특성", 한국물리학회지, 응용물리, Vol. 9, No. 3, pp. 405-411, May, 1996.
4) M. Murakami, "processing of bulk YBaCuO", Supercond. sci. Technol.,pp.185-202, April, 1992.
5) P. X. Zhang, et al., "The effect of annealing on stacking faults and Jc values of PMP processed YBCO", Supercond. sci. Technol., pp.15-19, Jan., 1995.
6) M. Murakami, K. Yamaguchi, H. Fujimoto, N. Nakamura, T. Taguchi, N. Koshizuka and S. Tanaka, "Flux pinning by non-superconducting inclusions in melt-processed YBaCuO superconductors", Cryogenics, pp.930-935, 1992.
7) Kamel Salama and Dominic F Lee, "progress in melt texturing of $YBa_2Cu_3O_x$ Superconductor", Supercond.sci. Technol., pp.177-193, July, 1994.
8) C.B.Bean, Rev. Mod. Phys. 36, 31(1964)
9) Shinjiro Tochihara, et al.,"Magnetization of type-II superconductors in the modified Bean model", Physica C 268, pp.241-256, 1996.

저 자 소 개

강 형 곤 (姜 亨 坤)

1967년 11월 20일생. 1995년 2월 전북대학교 일반대학원 전기공학과 석사 졸업 1999년 현재 동대학원 전기공학과 박사과정. 주관심 분야 반도체 및 초전도재료

Tel : 0652-270-2396
E-mail : joshuah@sinbiro.com

박 성 진 (朴 晟 鎭)

1964년 10월 11.일생. 1992년 2월 전북대학교 공대 전기공학과 석사 졸업. 1996년 2월 동대학원 박사 졸업. 주관심 분야 반도체 및 초전도재료.

Tel : 0652-270-2396
E-mail : psj6763@chollian.net

임 성 훈 (林 成 勳)

1973년 11월 1일 생. 1996년 전북대 전기공학과 졸업, 1998년 전북대 대학원 전기공학과 졸업(석사), 현재 동대학원 전기공학과 박사과정

Tel : 0652-270-2396
E-mail : dolphine@sinbiro.com

한 병 성 (韓 秉 誠)

1951년 12월 22일생. 1988년 프랑스 루이파스퇴르대학 박사학위취득. 1988년 프랑스 CNRS(프랑스 국립과학연구소)연구원. 현재 전북대 공대 전기공학과 교수. 공업기술연구소 연구원.

Tel : 0652-270-2396
E-mail : hbs@moak.chonbuk.ac.kr