Preparation of $YBa_2Cu_3O_x$ Superconductor Prepared with Additives of PbO and Ag_2O

Soon-Nam Chu*
Department of Electrical Engineering, Kyung Won University, Bokjeong-dong, Sujeong-gu, Seongnam-si, Gyeonggi 461-701, Republic of Korea

Jung-Cheul Park
Department of Electronic Engineering, Kyung Won University, Bokjeong-dong, Sujeong-gu, Seongnam-si, Gyeonggi 461-701, Republic of Korea

Yong-Woo Jeon
Department of Electronic Engineering, Sung Duk College, 1135-5 Hwanam-ri, Sinnyeong-myeon, Yeongcheon-si, Gyeongbuk 770-811, Republic of Korea

(Received November 13 2008, Accepted January 15 2009)

The improvement of preparation process of $YBa_2Cu_3O_x$ superconductor and its conducting properties is important for practical applications. In this study, the additives such as Ag_2O and PbO were used to improve the preparation conditions of $YBa_2Cu_3O_x$ superconducting bulk samples and the properties of $YBa_2Cu_3O_x$ superconductors prepared with powders using sol-gel method and solid state reaction method were studied. The effects of the different powders and the additives to the density, grain alignment, and porosity of samples, that affect the critical current density of superconductor, also have been investigated. It is found that the properties of $YBa_2Cu_3O_x$ prepared with sol-gel synthesized powder and the additives showed better superconductivities than those of conventionally prepared superconductors.

Keywords: Superconductor, Additives, Density, Grain alignment, Porosity

1. INTRODUCTION

Since $YBa_2Cu_3O_x$ superconductor with zero resistance at 90 K was found[1], many researchers have been performed vigorously to improve its superconducting properties and the results have been applied to the fabrication of various film or bulk type superconductors. High T_c oxide superconducting materials can be used in liquid nitrogen. The cheap cryogenic medium makes the materials promise in many fields, especially the tapes (wires) are designable for power transmission, fault current limiter. In addition, high strong magnets that could be used for magnetic levitation of transportation vehicles, separation of some mines and polluted water, as well as energy storage system was applied[2].

As an attempt to improve the preparation conditions of $YBa_2Cu_3O_x$ superconducting powder or bulk samples, the preparation methods such as MTG (Melt Texture Growth), MCP (Melt Condensed Processing), LPP (Liquid Phase Processing) etc. have been widely applied[3,4]. As those kinds of methods have many drawbacks such as higher and longer heat treatment during preparing processes the studies to improve processes are being developed currently. For those the researches of an ambient gas, an oxygen pressure control, and a use of additives were performed[5,6]. It has well known that materials with low melting temperature can make the materials with higher melting temperature lower. But it is hard work to avoid degradation of their properties. Thus, it needs many additional steps for reducing the effect on the properties of the system.

In this study, Ag_2O and PbO powder as additives were used for improving the preparation processes of $YBa_2Cu_3O_x$ superconducting bulk samples and its electrical properties. $YBa_2Cu_3O_x$ powder synthesized by sol-gel method (Y-SG), comparing with powder conventionally synthesized by solid-state reaction (Y-SS), was used to find additional effect on the properties of samples.

2. EXPERIMENT

Bulk samples used for this study were prepared by solid-state reaction. $YBa_2Cu_3O_x$ powders, synthesized by sol-gel method and by conventional solid-state reaction as shown Fig. 1 and 2, were mixed (with/without additives) and pressed into pellet with 20 mm diameter and 5 mm height. The samples were then sintered for 8 hours at 920 °C and subjected to final heating in flowing oxygen for 10 hours at 400 °C. The synthesized powders have a size distribution of 5 (Y-SS) and 0.2~1 μm (Y-SG) and their critical temperatures were 92 and 93 K, respectively. In order to lower heat treatment temperature of $YBa_2Cu_3O_x$ superconductors, 5 ~ 15 wt% of Ag_2O, 5 ~ 20 wt% PbO, and both 10 wt% Ag_2O and 10 wt% of PbO powder were added in $YBa_2Cu_3O_x$ matrix.

After sintering and oxygenation, the samples cut into 5 (width) × 1 (length) × 1(height) mm^3. Then the properties of $YBa_2Cu_3O_x$ samples were investigated by using differential thermal analysis (DTA), scanning electron microscope (SEM), energy dispersive spectroscopy (EDS) and X-ray diffraction (XRD). The four point probe type measurement method was used to measure electrical conductivity in liquid nitrogen bath.

*Author to whom corresponding should be addressed: electronic mail: snchu@kyungwon.ac.kr

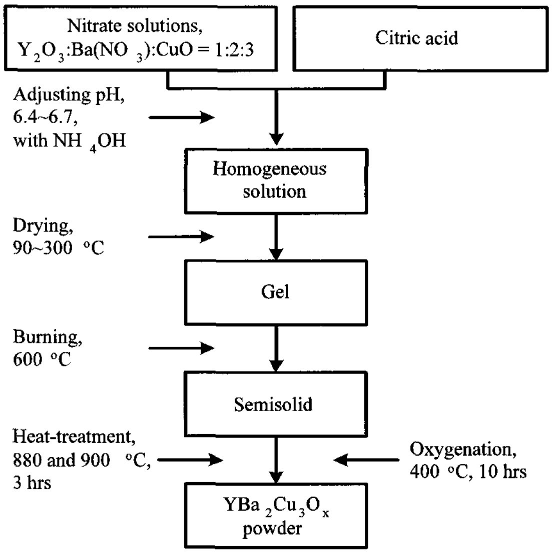


Fig. 1. Procedure of YBa_2Cu_3O powder prepared by sol-gel method.

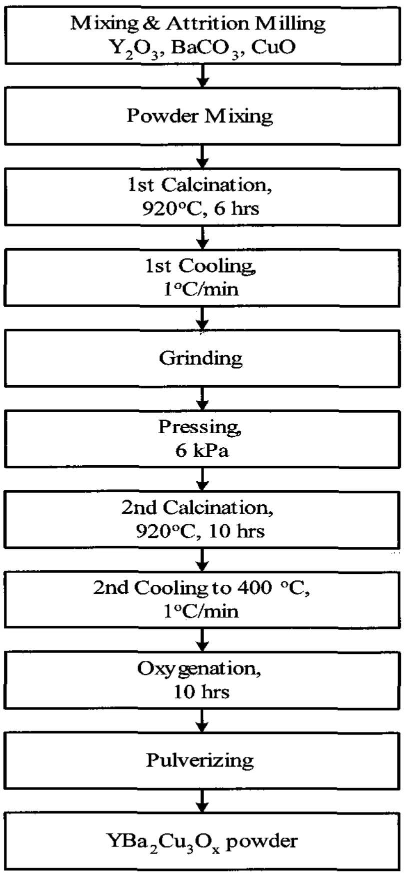


Fig. 2. Procedure of YBa_2Cu_3O powder prepared by solid-state reaction.

3. RESULTS AND DISCUSSION

3.1 *Melting temperature of samples with additives*

The melting point of relationship of $YBa_2Cu_3O_x$ and additives (Ag_2O and PbO) measured by using DTA are shown in Fig. 2 and 3, respectively.

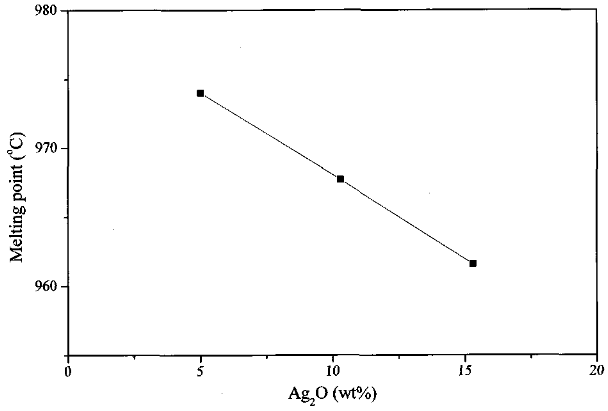


Fig. 3. Melting temperature changes of $YBa_2Cu_3O_x$ superconductor with different contents of Ag_2O.

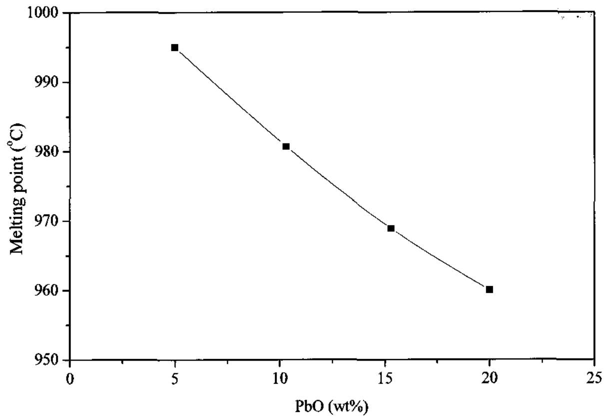


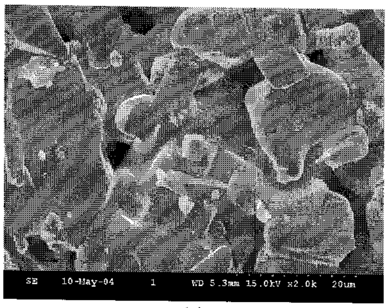
Fig. 4. Melting temperature changes of $YBa_2Cu_3O_x$ superconductor with different contents of PbO.

The melting point of pure $YBa_2Cu_3O_x$, Ag_2O and PbO were 1015, 961 and 880 °C, respectively. To improve the preparation condition, the additives were added in $YBa_2Cu_3O_x$ powders, which prepared by solid-state reaction and sol-gel method, respectively. The melting temperature of $YBa_2Cu_3O_x$ was decreased with increasing Ag_2O and PbO contents. The additives cause eutectic reaction between $YBa_2Cu_3O_x$ particles and additives. Thus, it could decrease the melting point of samples, resulting in reducing the heat treatment time.

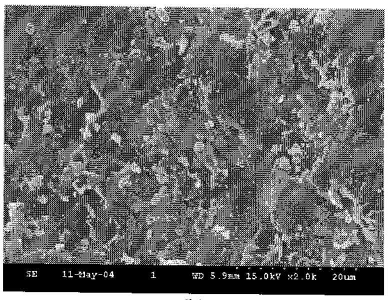
3.2 The superconductivities of samples

Figure 5 shows the sintered samples prepared with pure Y-SS and Y-SG. Powder, respectively. As Y-SG powder has relatively small particle size as tens times as Y-SS powder the packing condition was enhanced. Therefore, the surface structure of sample prepared with Y-SG powder appeared better densification and the connectivity. The density of sample was also improved comparing with the sample prepared with Y-SS powder.

The critical current density (J_c) changes of $YBa_2Cu_3O_x$ samples with different Ag_2O content was shown in Fig. 6. In the measurement of critical current density of pure



(a)



(b)

Fig. 5. Surface SEM photographs of $YBa_2Cu_3O_x$ superconductor prepared with solid-state synthesized powder (a) and sol-gel synthesized powder (b).

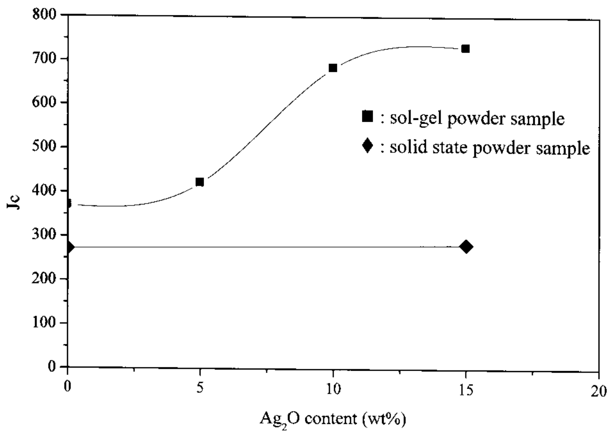


Fig. 6. J_c changes of $YBa_2Cu_3O_x$ samples with different Ag_2O content.

$YBa_2Cu_3O_x$ superconductor sample prepared with Y-SG powder showed 36 % higher than that of Y-SS powder. It is easily assumed that the sample prepared with Y-SG powder has higher density and lower porosity. $YBa_2Cu_3O_x$ superconductor samples prepared with Y-SS and Y-SG powder, added with 15 wt% of Ag_2O were found to improve the critical current by 6 % and 200 %, respectively.

Based on showing better superconductivities of $YBa_2Cu_3O_x$ superconductor, Y-SG powder is used for preparation of samples. To find out the optimal condition of PbO addition, the samples added with 5 ~ 20 wt% of PbO were prepared. As increasing the content of PbO additives, the melting temperature of $YBa_2Cu_3O_x$ sample was decreased as mentioned before. It was also shown that the packing effect was enhanced as shown in Fig. 7.

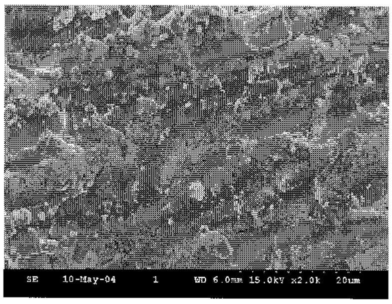


Fig. 7. Surface SEM photographs of $YBa_2Cu_3O_x$ samples prepared with Y-SG powder added with 10 wt% PbO.

Table 1. J_c changes of $YBa_2Cu_3O_x$ samples with different PbO content.

Contents of impurities, (wt%)	Critical current density, (A/cm^2)
0	370
5	415
10	374
15	349
20	283

But PbO is not independent phase in $YBa_2Cu_3O_x$ matrix there is a reaction between them. Thus, the critical current densities of samples decreased as increasing PbO content (Table 1).

The reaction produces non-superconducting phase such as $BaPbO_3$ and liquid phase as shown in XRD analysis (Fig. 8), resulting in reducing superconductor phase. Therefore, the result may cause the effect on the superconductivities of $YBa_2Cu_3O_x$ superconductor.

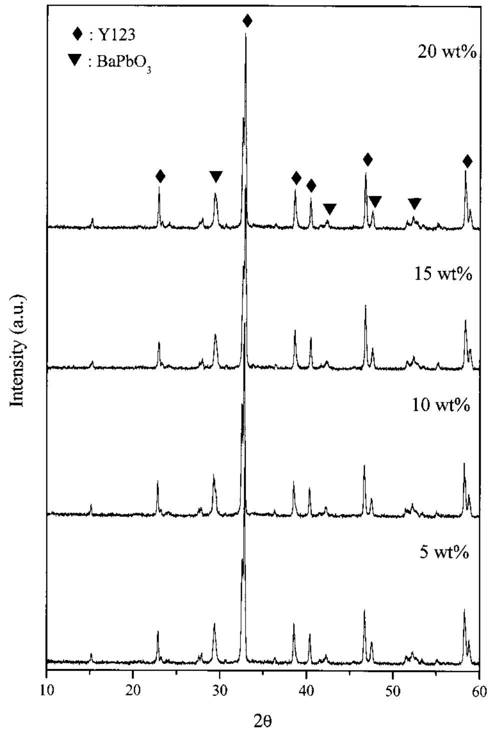


Fig. 8. XRD analysis of $YBa_2Cu_3O_x$ samples prepared with sol-gel synthesized powder and 5 ~ 20 wt% PbO.

EDS analysis (Table 2 and 3) shows the presence of liquid PbO and $YBa_2Cu_3O_x$ superconductive molecular ratio known as superconducting phase. Therefore, the increase of non-superconducting phase such as PbO and $BaPbO_3$ resulted in the decrease of critical current density.

Table 2. EDS analysis of $YBa_2Cu_3O_x$ sample prepared with 10 wt% PbO.

Element	wt%	Atomic %
O	11.57	47.85
Cu	23.51	24.47
Y	10.86	8.08
Ba	38.34	18.46
Pb	3.54	1.13
Totals	87.82	

The drawback condition of PbO additives, which reduce superconductor phase during the preparation process of samples, could be suppressed by using oxygen or non-active gas.

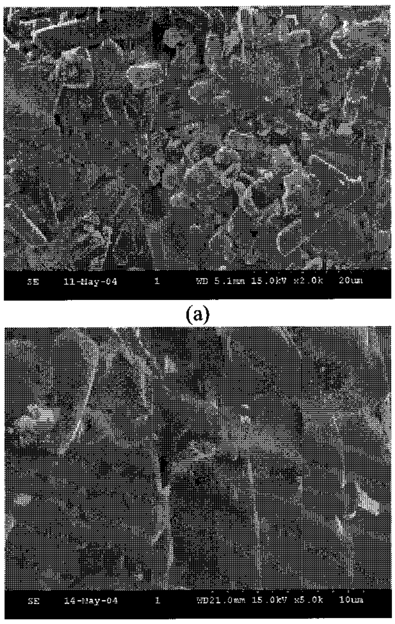
Table 3. EDS analysis of $YBa_2Cu_3O_x$ sample prepared with 15 wt% PbO.

Element	wt%	Atomic %
O	9.09	46.03
Cu	20.21	25.77
Y	7.72	7.04
Ba	33.31	19.65
Pb	3.87	1.51
Totals	74.20	

From the effect of Ag_2O and PbO additives on superconductivities, the samples added with both Ag_2O and PbO, were prepared to find out the optimal contents of additives. The sample prepared with the addition of both 10 wt% Ag_2O and 10 wt% PbO showed the highest critical current density of 435 A/m^2. Figure 9 shows the sample SEM photograph, and the additives could promote grain growth and decrease porosities and weak links between grain boundaries of $YBa_2Cu_3O_x$ superconductors.

4. CONCLUSION

$YBa_2Cu_3O_x$ superconductor samples with additives were prepared to improve the preparation processes. The



(a)

(b)

Fig. 9. Surface (a) and side (b) SEM photographs of $YBa_2Cu_3O_x$ samples prepared with Y-SG powder added with 10 wt% Ag_2O and 10 wt% PbO.

additives of Ag_2O and PbO were effective for decreasing the melting point of $YBa_2Cu_3O_x$ superconductors. Thus, it could reduce the preparation processes of heat treatment. In addition, the additives increased grain growth and interconnections of grain boundaries, so that it could effectively improve the $YBa_2Cu_3O_x$ superconductor properties and preparation conditions. It was also found that the powder synthesized by sol-gel method as well as applying additives could enhance the preparation processes.

REFERENCES

[1] M. K. Wu, J. R. Ashburn, C. J. Torng, P. H. Hor, R. L. Meng, L. Lao, Z. J. Huang, Y. Q. Wang, and C. W. Chu, Phys. Rev. Lett. **58**, 908 (1987).

[2] Y. Iijima, M. Hosaka, N. Tanabe, N. Sadakata, T. Saitoh, O. Kohno, and K. Takeda, J. Mater. Res.**12**, 2913 (1997).

[3] H. Asaoka and H. Takei, Jpn. J. Appl. Phys. **33**, 923 (1994).

[4] S. Jin, T. H. Tiefel, R. C. Sherwood, R. B. van Dover, M. E. Davis, G. W. Kammlott, and R. A. Fastnacht, Phys. Rev. B, **37**, 7850 (1988).

[5] G. W. Kammlott, T. H. Tiefel, and S. Jin, Appl. Phys. Lett. **56**, 2459 (1990).

[6] Z. G. Fan, Y. Q. Shan, W. H. Wang, X. Y. Wang, D.W. Soh, and Z. X. Zhao, Physica C, **282-287**, 495 (1997).