Zone-melting of EPD $YBa_2Cu_3O_x$ Thick Film under Low Oxygen Partial Pressure

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Abstract

The fine $YBa_2Cu_3O_x$ powder (0.2~1.0 ㎛) is produced by sol-gel method, and electrophoresis deposition is used for the preparation of $YBa_2Cu_3O_x$ thick films which are deposited on Ag wire. The oriented $YBa_2Cu_3O_x$ was tried to be prepared by the zone-melting method under low oxygen partial pressure. The orientation and the phase composition were examined by the X-ray diffraction and the superconductivities were measured by 4 line method. The critical current densities are still quite low, which may be due to unsuitable technical parameters for zone-melting of $YBa_2Cu_3O_x$ thick films. Therefore the heat treatment condition and controlling of low oxygen partial pressure should be improved in the future experiment.

Key Words : $YBa_2Cu_3O_x$ thick films, Electrophoresis, Zone-melting, Oxygen pressure

1. INTRODUCTION

Oxygen partial pressure is one of the important parameters in the processing of oxide superconductors. In the preparation of $Bi_2Sr_2Ca_2Cu_3O_y$ some scientists used the low oxygen partial pressure as the ambient atmosphere to increase the superconducting phase in the bulk sample [1,2,3,4]. 7.7 % of oxygen partial pressure was used in the heat treatment of Ag-sheathed $(Bi,Pb)_2Sr_2Ca_2Cu_3O_y$ tapes for improvement of the superconductivity [5]. 1 % or 0.1 % of the oxygen partial pretrial were used in the preparation of textured $Nd_{1+x}Ba_{2-x}Cu_3O_y$ in order to impress the substitution of Nd for Ba position, in the other words to decrease the x value in the equation of $Nd_{1+x}Ba_{2-x}Cu_3O_y$, because the x value of over 0.2 can reduce the superconductivities [6,7] and $Nd_{1+x}Ba_{2-x}Cu_3O_y$ would become non-superconductive tetragonal phase at larger x value. T.B Lindemer et al [8] measured the relation of oxygen partial pressures and melting temperatures as well as x value of $Nd_{1+x}Ba_{2-x}Cu_3O_y$ in the processing. The equation is as following:

$$\log (P[O_2][\text{MPa}])=33.872-17.9X+(-48226+24983X)/T$$

According to the equation the melting temperature of $Nd_{1+x}Ba_{2-x}Cu_3O_y$ is reduced with the decreasing oxygen partial pressure. In the previous work [9] of the authors for preparing the textured $Nd_{1+x}Ba_{2-x}Cu_3O_y$ the relation between oxygen pressures and melting temperatures is in accordance with above equation. Reducing the oxygen partial pressure may be an efficient way for reducing melting temperature of $YBa_2Cu_3O_x$ because $YBa_2Cu_3O_x$ compound is sintered from oxides and it is heat-treated in oxygen ambient.

In the experiment $YBa_2Cu_3O_x$ thick films are deposited on the Ag wires by the electrophoresis deposition(EPD). After EPD the films are tried to be processed by zone-melting method in order to make orientation in $YBa_2Cu_3O_x$ thick film. The zone-melting temperature can not be close to 961℃ of Ag melting point and $YBa_2Cu_3O_x$ has 1010℃ of melting point, therefore $YBa_2Cu_3O_x$ film on Ag substrate can not be melted. If the melting temperature of $YBa_2Cu_3O_x$ can be reduced to below the melting point of Ag by the controlling oxygen partial pressure and then the $YBa_2Cu_3O_x$ film can be melted in high temperature zone and Ag in solid state, the textured $YBa_2Cu_3O_x$ thick film can be realized.

2. EXPERIMENT

2-1 Preparation of $YBa_2Cu_3O_x$ fine powder by Sol-gel method

In order to reduce the micro-crack in the $YBa_2Cu_3O_x$ film fine powder of $YBa_2Cu_3O_x$ was used in the electrophoresis deposition, which was synthesized by sol-gel method.

Y_2O_3, $Ba(NO_3)_2$ and CuO (all with the purity of A.R.) powders were weighed respectively according to the atom ratio of Y:Ba:Cu=1:2:3. Nitrate solutions of Y, Ba metal elements were prepared, which were together with $Ba(NO_3)_2$ added into the excess citric acid solution. The value of pH was adjusted by dropping NH_3H_2O. By the time $Ba(NO_3)_2$ would be completely dissolved and the citric solution was deep blue. Then the solution was put in muffle furnace and heated to get rid of the solvent. During this period there existed the transition from transparent liquid to gel. With the color of the gel growing deeper and deeper, it would turn to be black netted viscose. Finally it was burned into ashes by the heat. The final ashes were grinded into powder and sintered at the temperature of 880℃ with flowing O_2. The ultimate black powder was YBaCuO superconductor powder. XRD examination proved the powder is single phase $YBa_2Cu_3O_x$. The size of Y123 powder was among 0.2 μm~1 μm. At liquid nitrogen temperature the critical transition temperature (T_c) was about 91K, and the critical current density (J_c) was 5-30A/㎠.

2-2 Preparation of $YBa_2Cu_3O_x$ thick films by electrophoresis deposition (EPD)

The YBaCuO thick film was deposited by the electrophoresis, of which the experimental details were described in the previous paper [10]. 20㎎ of iodine and 2㎎ of fine YBaCuO powder per 100㎖ acetone were used in the Pyrex beaker under which a magnetic stirrer stirred 5 minutes before each deposition in order to make homogenous suspension of the YBaCuO powder in the electrophoresis solution. A 40~50 ㎛ thickness of YBaCuO film on Ag plate could be deposited in about three minutes. After deposition the film is rolled in order to increase the density and the adhesion of the film to the Ag plate.

2-3 Zone-melting(Z.M.) of $YBa_2Cu_3O_x$ thick film under low oxygen partial pressure

A special furnace was designed with very narrow high temperature zone(fig. 1), the FeCrAl alloy wire was used as the heating element. The wideness of high temperature zone in the furnace is 4 ㎜ and the temperature gradient at the interface of liquid and solid is about 20 ℃/㎝. The large temperature gradient is benefit to the oriented growth of YBaCuO. The zone-melting process was carried out in low oxygen partial pressure (100 ㎩) and Ag was used as substrate. The mixture of 0.1 % O_2 and 99 % Ar was made from pure oxygen and pure argon in a plastic bag.

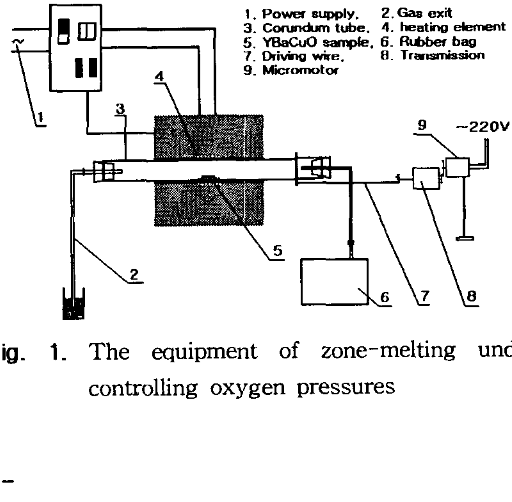

Fig. 1. The equipment of zone-melting under controlling oxygen pressures

3. RESULTS AND DISCUSIONS

3-1. The relation of melting temperature of $YBa_2Cu_3O_{6.5+\delta}$ and oxygen partial pressure

$YBa_2Cu_3O_{6.5+\delta}$ melts and decomposes based on following reaction:

$$2YBa_2Cu_3O_{6.5+\delta}=Y_2BaCuO_5(s)+3BaCuO_2(l)+2CuO(l)+\delta O_2(g)$$

From the thermodynamic principle in the above equation when oxygen partial pressure reduces the melting and decomposing temperature of YBaCuO will be reduced. Therefore it is possible to reduce the melting temperature of $YBa_2Cu_3O_{6.5+\delta}$ by lower oxygen partial pressure. According to Lindemer equation [8] for $Nd_{1+x}Ba_{2-x}Cu_3O_y$ at x=0 when oxygen partial pressure changes from pure oxygen to 0.1 %, the melting temperature dropped 110℃ (from 1385 K to 1275 K). In the practice of $Nd_{1+x}Ba_{2-x}Cu_3O_y$ zone-melting, the decreasing melting temperature was about 100~120℃ [9].

Here the same equipment is used for the preparation of YBaCuO superconductor. It was estimated that when oxygen partial pressure was 100 Pa, the melting point of YBaCuO may decrease to below 961℃(the melting point of Ag).

3-2 Superconductivities of zone-melted EPD $YBa_2Cu_3O_x$ thick film

After the oxygenation of the zone-melted EPD $YBa_2Cu_3O_x$ thick film, the transition temperature (T_c) of the sample is 92 K and the transition wide(ΔT_c) is 3 K. The critical current densities of the zone-melted thick films are shown in table 1.

Table 1. Critical current densities(78 K, 0 T) of zone-melted $YBa_2Cu_3O_x$ thick film

Sample	1	2	3	4	5	6	7	8
Z.M.Speed, ㎜/h	6	6	6	6	2	2	2	2
Z.M.Temp., ℃	953	953	958	958	963	963	963	963
Ic (A)	0.16	0.20	0.24	0.12	0.12	0.24	0.32	0.66
Jc (A/㎠)	160	120	160	80	80	160	213	442

3-3 X-ray diffraction of zone-melted $YBa_2Cu_3O_x$ thick films

After the zone-melting there are cracks and micro-cracks observed by microscopy on the surface of YBaCuO. The X-ray diffraction pattern (XRD) of a typical sample is shown in fig. 2. Besides the (001) diffraction of $YBa_2Cu_3O_x$ there are some diffraction peaks from other crystal planes and Y_2BaCuO_5 (Y211 phase).

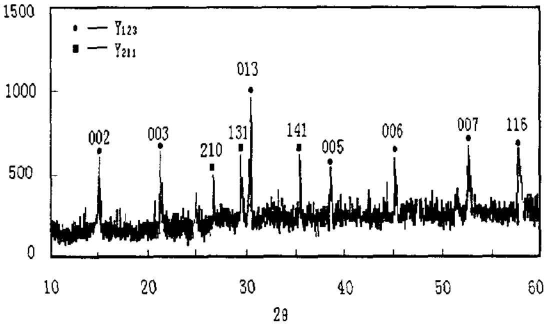

Fig. 2. X-ray diffraction of zone-melting EPD $YBa_2Cu_3O_x$ thick film

3-4 Discussion about zone-melting results of $YBa_2Cu_3O_x$ thick films

The critical current density is not high in table 1, and there is a large difference between predicting aim and practice results. The main reasons may be suggested as followings.

There are some cracks and micro-cracks in the zone-melted $YBa_2Cu_3O_x$ thick films, which may be caused by the difference of thermal expansion coefficient between the $YBa_2Cu_3O_x$ thick film and Ag substrate. These cracks reduce the I_c of the samples seriously. From XRD pattern it is seen that the zone melted sample is not perfect (001) oriented, besides the (001) diffraction peaks there are some peaks form other crystal planes and other impurities phases. The temperature of zone-melting of 953℃~963℃(see table 1), which is close to the melting point of Ag, may be too low for the melted texture growth of $YBa_2Cu_3O_x$ thick films. Based on the analysis of the relation between oxygen partial pressure and melting temperature of $YBa_2Cu_3O_x$, the melting temperature could be reduced to below the melting point of Ag (961℃) if the oxygen partial pressure was controlled to 0.001.

Therefore it is the most important to control the low oxygen partial pressure accurately for reducing the melting temperature of $YBa_2Cu_3O_x$ thick films. After the zone-melting the sample should be heat treated in a suitable condition in order to overcome the cracks caused by thermal expansion

4. CONCLUSIONS

The fine $YBa_2Cu_3O_x$ powder(0.2~1.0 ㎛) can be produced by sol-gel method, and electrophoresis deposition can be used for the preparation of $YBa_2Cu_3O_x$ thick films. The oriented $YBa_2Cu_3O_x$ was tried to be prepared by zone-melting method under low oxygen partial pressure. However the critical current densities are still quite low which may be due to unsuitable technical parameters for zone-melting of $YBa_2Cu_3O_x$ thick films. Therefore the heat treatment condition and control of low oxygen partial pressure should be improved in the future experiment.

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