Y-Ba-Cu-O THIN FILMS BY EVAPORATING THE THREE ELEMENTS USING RESISTIVE HEATING

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Abstract

High-T_c superconducting Y-Ba-Cu-O thin films were fabricated by evaporating the three metal elements Y, Ba, and Cu using resistive heating. Subsequently, the films were annealed in oxygen in order to obtain the superconducting phase. The critical temperature T_c (onset) and T_c (end) of the obtained film were 88 K and 65 K, respectively.

Introduction

For electrical applications, thin film preparation of high T_c materials such as Y-Ba-Cu-O is very important. Several studies on preparation of films of high-T_c materials have been reported. Films of high-T_c materials have been fabricated using 50 Hz ac sputtering,[1,2] screen printing method,[3,4] magnetron sputtering,[5-7] dual magnetron sputtering.[8]

This paper reports that Y-Ba-Cu-O thin films were fabricated by multi-layer evaporation of the three elements Y, Ba, and Cu. The three elements were deposited by resistive heating with three W boats. Subsequently, the films were annealed in a furnace in order to obtain the superconducting phase. In our best film, T_c (onset), T_c (end) were 88 K, and 65 K, respectively, and the resistivity decreased when temperature decreased.

Experimental

Evaporating Three Elements

Single crystalline $SrTiO_3$(100) were used as substrates. Evaporation were done in a conventional vacuum system shown in Fig. 1. The main pump of the system was oil-diffusion pump, whose ultimate pressure was 1×10^{-4} Pa. Source materials were pure metals Cu, Ba and Y, and three W boats and shutter were used for multi-layer-deposition. In order to minimize the reaction with the substrate, Cu was first evaporated, followed by layers of Ba and Y, as shown in Fig. 2. If we consider the annealing after evaporation of the three elements, decreasing the thickness of each layer and increasing number of total layers are desirable. However, the more the number of the total layer increases, the larger the amount of time and labor of the evaporation become. Therefore, in this study the evaporation sequence were repeated 6 times, and number of total layers was 18. The top layer was Y, so that Ba layer might not be the top layer, because Ba easily reacts with humidity in atmosphere. The thickness and the deposition rate were monitored with a Sloan thickness monitor. The deposition rate was in the range of 0.2 - 0.5 nm/s. Thicknesses of each layer of Cu, Ba, Y are listed in Tables I and II. In Table I, thickness of the individual layer, monitored by Sloan thickness monitor, was determined so that composition of the film would be Y : Ba : Cu = 1 : 2 : 3. In Table II, 10% compensation of Cu was done.

Annealing

In order to obtain the 1:2:3 superconducting phase, solid phase reaction of the multi-layers and supply of oxygen were done in a furnace in flowing oxygen. Typical annealing process of the films was based upon that of bulk Y-Ba-Cu-O, and was as follows: The film was put in the furnace and its temperature was increased to a value in the range of 850-940°C. The sample was then treated in flowing oxygen for 0.5 hour, and finally the furnace temperature was decreased at a rate of 2°C/min to 300°C.

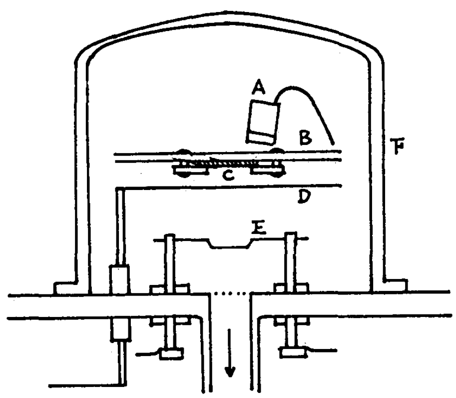

Fig.1. Schematic of evaporator.

A:Thickness monitor
B:Sample holder
C:Substrate
D:Shutter
E:Filament
F:Belljar

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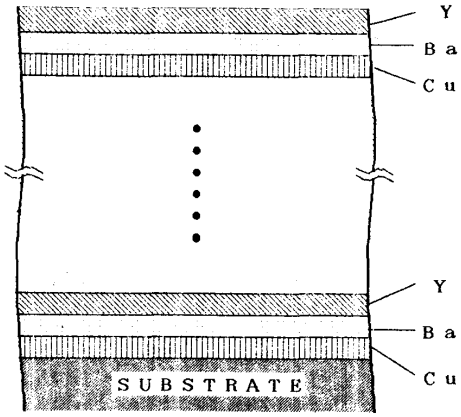

Fig.2. Built-up sequence of element.

Table I Thickness of each layer of evaporated metal films.

Cu (nm)	Ba (nm)	Y (nm)	number of layers
3.2	11.7	2.6	1
25.5	93.8	20.8	4
12.7	16.9	10.4	1

Total thickness: 647 nm.
Composition by thickness monitor; (Y:Ba:Cu =) 1:2:3.

Table II Thickness of each layer of evaporated metal films.

Cu (nm)	Ba (nm)	Y (nm)	number of layers
37.3	125	25.7	6

Total thickness: 1130 nm.
Composition by thickness monitor; (Y:Ba:Cu =) 1:2:3.3.

Results and Discussion

ICP Analysis

In order to confirm that composition (Y:Ba:Cu) of the obtained film was 1:2:3, ICP (Inductively Coupled Plasma) analysis of the films was done. Compositions of the films fabricated using the conditions listed in Table I, which was obtained by ICP, were Y:Ba:Cu = 1 : 2.11 : 2.75, so that Cu was about 8% short of the expected value. This shortage was thought to be attributed to the difference between amount of the deposited film thickness on the substrate and that on the crystal of the thickness monitor. The causes of this difference were thought as follows,

1) difference of temperature between the substrate, which was neither water-cooled nor heated, and the crystal of the thickness monitor, which was water-cooled.

2) difference of material between the substrate ($SrTiO_3$) and the crystal of the monitor.

Therefore, in Table II total thickness of Cu was increased by 10 %. Then, compositions of the films fabricated with Table II, which was obtained by ICP, were Y:Ba:Cu = 1 : 2.18 : 3.09, so that Cu was compensated. Comparison of characteristics of the films prepared with Tables I and II is shown in Table III.

Temperature Dependences of the Resistivity

Temperature dependences of the resistivity of the obtained Y-Ba-Cu-O films were measured in a range from 8 to 300 K with liquid He refrigerator. Four electrodes of gold film were deposited on the Y-Ba-Cu-O film for ohmic contact[9,10] and the resistivity were measured by four-probe method.

Temperature dependences of the resistivity of the film prepared with the conditions listed in Table I, are shown in Fig. 3. The highest annealing temperature was 940°C shown in the figure caption, and T_c(onset) and T_c(zero) were 75K and 18K, respectively. When the annealing temperature was 900°C, resistance of the film did not become zero down to 10 K.

Temperature dependences of the resistivity of the film with Table II are shown in Fig. 4. Resistivity decreased when temperature decreased, and T_c(onset) and T_c(end) were 88 K and 65 K, respectively. The measured Tc versus annealing temperature of the films prepared with the conditions in Table II is shown in Fig. 5. When annealing temperature was as low as 850°C, and as high as 940°C, resistance of the film did not become zero down to 10 K, though T_c(onset) was about 80 K. Therefore, the resistance of the film did not become zero when the annealing process was not optimized, though composition is nearly 1 : 2 : 3.

Table III Comparison of the films prepared with two different conditions shown in Tables I and II.

	Table I	Table II
Composition (Y:Ba:Cu)		
by thickness monitor	1 : 2.0 : 3.0	1 : 2.0 : 3.3
by ICP	1 : 2.11 : 2.75	1 : 2.18 : 3.09
Total thickness	647 nm	1130 nm
T_c(onset)	75 K	88 K
T_c(end)	18 K	65 K

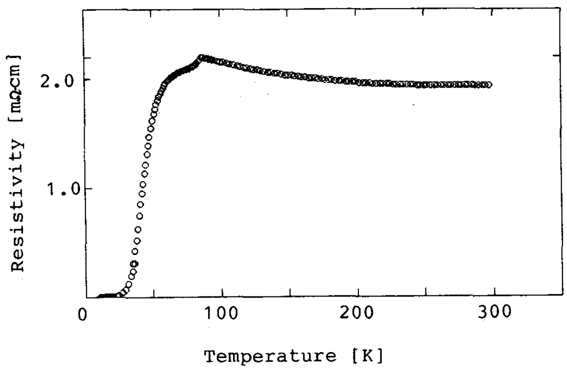

Fig.3. Temperature dependence of resistivity of the film with Table I.

composition by thickness monitor;
Y:Ba:Cu=1:2.0:3.0
by ICP;
Y:Ba:Cu=1:2.11:2.75
annealing conditions
650℃,3hour→750℃,1hour→
940℃,1hour→300℃
in flowing O_2
T_c(onset) :75K
T_c(zero) :18K

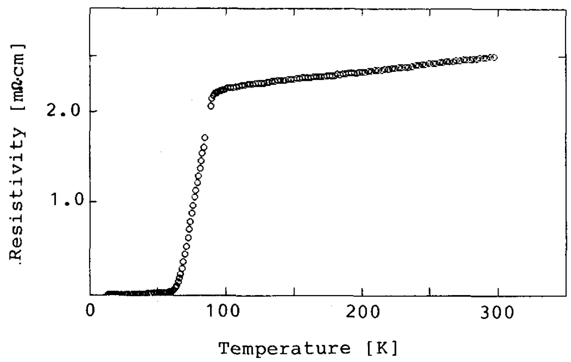

Fig.4. Temperature dependence of resistivity of the film with Table II.

composition by thickness monitor;
Y:Ba:Cu=1:2.0:3.3
by ICP;
Y:Ba:Cu=1:2.18:3.09
annealing conditions
900℃,30min $\xrightarrow[2℃/min]{}$ 300℃
in flowing O_2
T_c(onset) :88K
T_c(zero) :65K

Conclusions

In conclusion, high-T_c superconducting Y-Ba-Cu-O thin films were prepared by multi-layer evaporation of the three elements Y, Ba, and Cu. The three elements were deposited by resistive heating with three W boats. Subsequently, the films were annealed in a furnace in order to obtain the superconducting phase. In the best film, T_c (onset), T_c (end) were 88 K, and 65 K, respectively.

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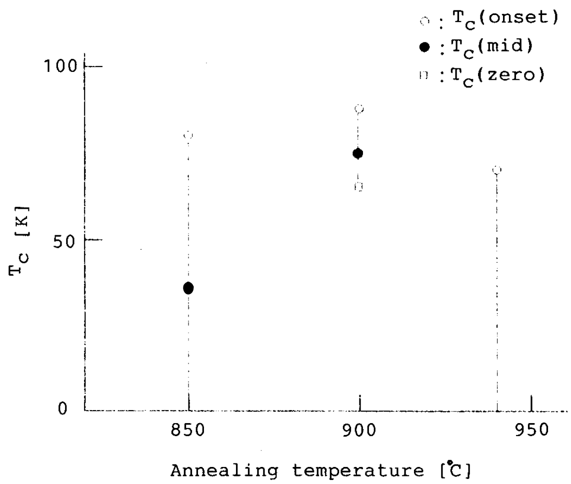

Fig.5. The measured T_c versus annealing temperature for Y-Ba-Cu-O films with Table II.

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