



Effect of Ag₂O Doping on Some Physical Properties of Y156 Superconductor

Prepared by Solid State Reaction

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Abstract

The influence of Ag₂O doping on some properties of Y156 Superconductors (YBa₂Cu₃O_{7-x} + xAg₂O where x = 0, 0.05, 0.10, 0.15, 0.20) prepared by solid state reaction were studied. Yttrium (III)oxide (Y₂O₃), Barium Carbonate (BaCO₃), Copper Oxide (CuO) and Silver(I) Oxide (Ag₂O) were used as the precursors. All of samples obtained were investigated by SEM, EDX, resistivity measurement, and the standard iodometric titration. The resistivity measurement by four-point probe technique showed that the highest critical temperature was found in pure Y156 at 95 K. The amount of silver(I) oxide has significant effect to the critical temperature of Y156 superconductor. The highest critical temperature of Y156 doped Ag₂O was found in Y156 doped 0.15Ag₂O at 93K. However, it had lower critical temperature than Y156 without doping Ag₂O. The surface of Y156 doped Ag₂O were improved by adding Ag to the porous structure and had narrow grain boundaries. We studied the compositions of elements demonstrated uneven distribution of elements and non-homogeneous material. Also, the results showed that the higher Cu³⁺/Cu²⁺ ratio, the higher critical temperature was found.

Keywords: Superconductor doped Ag, Y156 Superconductor, Solid State Reaction

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1. Introduction

Since the discovery the first high-temperature superconductor having critical temperature higher than the boiling point of liquid nitrogen in Y-Ba-Cu-O compound has been investigated by Chu and his colleague [1] in 1987, called as Y123 ($\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$) superconductor. Then Y-Ba-Cu-O system has become interested superconductor of the scientists' research widely. The Y123 superconductor has the orthorhombic perovskites crystal structure with two different Cu sites: CuO chains and CuO_2 planes. The critical temperature (T_c) of Y-Ba-Cu-O superconductor sensitively depends on both the hole's concentration in the CuO_2 planes and the relative concentration of the oxygen within the planes [2]. Since this superconductor contains copper (Cu) and oxygen (O), called as the cuprate superconductor.

In 2010, Udomsamuthirun et al. [3] synthesized the new Y-Ba-Cu-O superconductors as compositions of Y5-6-11, Y7-9-16, Y5-8-13, Y7-11-18, Y156, Y3-8-11 and Y13-20-33, where the numbers indicate Y, Ba, and Cu atoms respectively. To improve the superconducting properties of superconductors, various elements were added in Y-Ba-Cu-O system for increasing its critical temperature (T_c) and critical current density (J_c) [4]. However, there are some result shown that Ag-doping can improve the electrical properties of Y123 superconductor [5] such as critical current density [6-8]. The effect of Ag-doping can be investigated by measurement the oxygen content on

sample that the oxygen content in the structure of the Y-Ba-Cu-O superconductor was correlated with the critical temperature. The amount of oxygen can be calculated from the amount of Cu^{2+} and Cu^{3+} in the superconductor. So the critical temperature has the relation to the amount of Cu^{2+} and Cu^{3+} . The ratio of trivalent to divalent copper of Y123 has been determined by Choy et al. [9]. They found that T_c and the ratio of $\text{Cu}^{3+}/\text{Cu}^{2+}$ depend on the annealing temperature and time. The higher T_c is higher of the $\text{Cu}^{3+}/\text{Cu}^{2+}$ becomes. Furthermore, the ratio of $\text{Cu}^{3+}/\text{Cu}^{2+}$ was investigated in Y134 and Y145 doped Ag_2O . The results reported that the higher $\text{Cu}^{3+}/\text{Cu}^{2+}$, the higher critical temperature found.

In this paper, we interested in the effect of Ag_2O on some physical properties of Y156 superconductors. All of samples obtained were investigated by SEM, EDX, resistivity measurement, and the iodometric titration. The effect of $\text{Cu}^{3+}/\text{Cu}^{2+}$ ratio on critical temperature of Y156 doped Ag_2O were investigated.

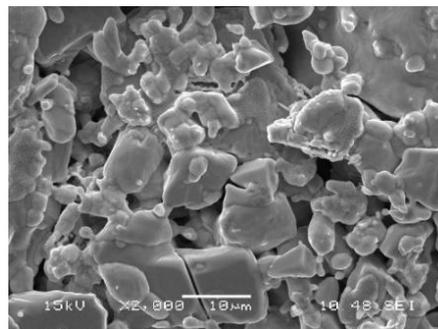
2. Materials and Experiment

The samples of $\text{YBa}_5\text{Cu}_6\text{O}_{13-y} + x\text{Ag}_2\text{O}$ series were prepared through conventional solid state reaction method with nominal composition $x = 0, 0.05, 0.10, 0.15$ and 0.20 . High-purity powders of Y_2O_3 , BaCO_3 , CuO and Ag_2O with the appropriate ratio contained Y156 by weight, were mixed and ground. After grinding, calcinations were carried out at 900°C for 24 hours two times.

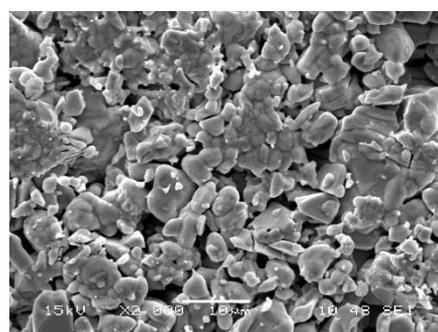
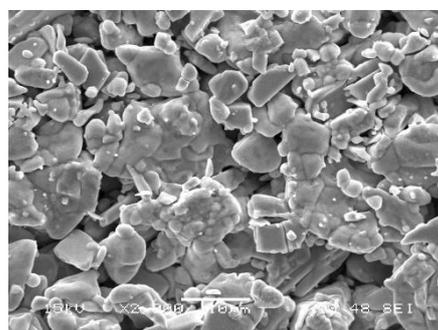
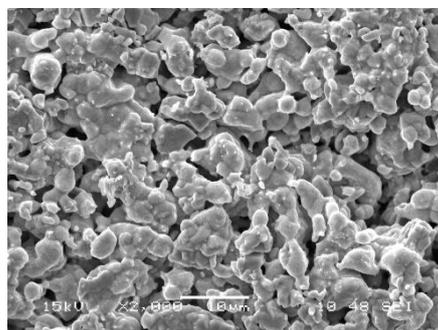
In each calcination cycle cooling was done slowly ($2\text{ }^{\circ}\text{C}/\text{min}$) and samples were ground before the next cycle. After calcination process, the samples were pressed into pellets form and took into sintering process at $900\text{ }^{\circ}\text{C}$ for 24 hours. Finally, the samples were annealed with flowing oxygen at $550\text{ }^{\circ}\text{C}$ for 24 hours and cooled slowly ($2\text{ }^{\circ}\text{C}/\text{min}$) to room temperature. All of samples were characterized by SEM and EDX (JEOL JSM 6380). The electrical resistivity has been measured by standard four-point probe method. The standard iodometric titration was used to determine the amount of Cu^{2+} and Cu^{3+} and oxygen content.

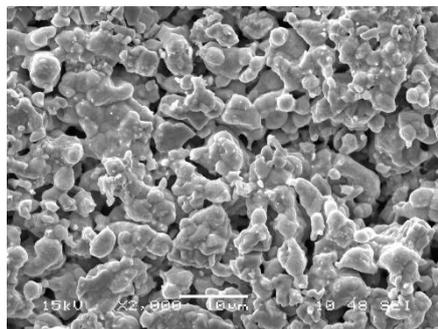
3. Results and Discussion

All samples were characterized by SEM and EDX that images were shown in fig. 1. The surfaces with inhomogeneous texture without any impurities were found. The EDX analysis of all samples were shown in Table 1. We found that the larger pores of pure Y156 were eliminated from the composites by the addition of Ag_2O . Our results shown that the surface of Y156 doped Ag_2O were improved by Ag adding on the porous structure. This result was consistent with the research of Azambuja et al [10] and Plesch et al [11] that studied Ag doped superconductor in Y123, they found that the sample doped Ag had the narrow grain boundaries but the superconducting properties were not significantly influenced by Ag addition.



Y156

Y156 + 0.05 Ag_2O Y156 + 0.10 Ag_2O Y156 + 0.15 Ag_2O

Y156 + 0.15Ag₂O**Figure 1** The SEM images of Y156 doped Ag₂O.**Table 1** EDX analysis of all samples.

Sample	Element (Atomic %)				
	Y	Ba	Cu	O	Ag
Y156	4.5	17.36	21.76	57.27	-
Y156+0.05Ag ₂ O	3.53	13.97	20.42	62.59	0.51
Y156+0.10Ag ₂ O	3.55	17.47	20.75	57.89	0.83
Y156+0.15Ag ₂ O	2.62	15.86	20.63	60.59	0.77
Y156+0.20Ag ₂ O	2.45	15.15	19.19	62.93	0.7

The resistivity measurement depending on temperature of our samples obtained was conducted with four-point probe technique in range 77-120 K as shown in the summation of T_c onset and T_c offset shown in fig. 2. The T_c onset temperature was taken as the temperature at which tangent of the resistivity versus temperature curve intersects with the tangent of the part where resistivity dropped abruptly and T_c offset was defined as the temperature at which the resistivity reached zero. The T_c onset and T_c offset were read out from the normalized resistivity curves that were shown in Table 2.

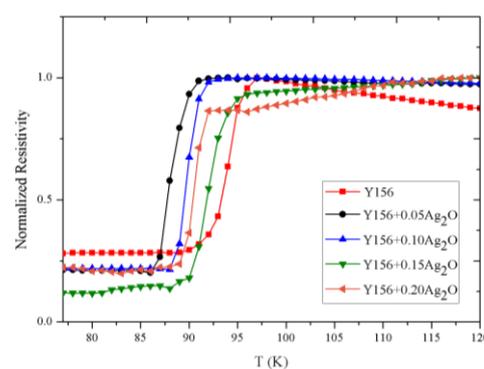
**Figure 2** The normalized resistivity versus the temperature of Y156 with Ag₂O-doped superconductors.

Table 2 The critical temperature of all samples.

Sample	T _c offset (K)	T _c onset (K)
Y156	92	95
Y156+0.05Ag ₂ O	86	89
Y156+0.10Ag ₂ O	88	91
Y156+0.15Ag ₂ O	90	93
Y156+0.20Ag ₂ O	89	92

From Table 2, we found that the highest critical temperature was found in pure Y156 sample with T_c onset 95 K and Y156+0.05Ag₂O sample had

the lowest critical temperature at T_c onset 89 K. Our result showed that Ag₂O doping has significant effect to the critical temperature of Y156.

The standard iodometric titration was used to determine the amount of Cu²⁺ and Cu³⁺ and oxygen content. We calculated the oxygen content by using the sum of the oxidation numbers of Y156 (YBa₃Cu₆O_{13-y} + xAg₂O; x = 0, 0.05, 0.10, 0.15, 0.20). The Cu³⁺/Cu²⁺ ratio and oxygen content of all samples were shown in Table 3. Also, the effect of Cu³⁺/Cu²⁺ ratio on T_c onset were shown in fig. 3.

Table 3 Cu³⁺/Cu²⁺ ratio and the oxygen content of all samples obtained.

Sample	Average mole (×10 ⁻⁶)		Cu ³⁺ /Cu ²⁺	Oxygen content
	Cu ²⁺	Cu ³⁺		
Y156	8.46	2.22	0.26	12.87
Y156+0.05Ag ₂ O	8.02	1.08	0.14	12.88
Y156+0.10Ag ₂ O	8.55	1.91	0.14	12.90
Y156+0.15Ag ₂ O	8.30	1.96	0.24	12.85
Y156+0.20Ag ₂ O	8.71	1.49	0.17	12.96

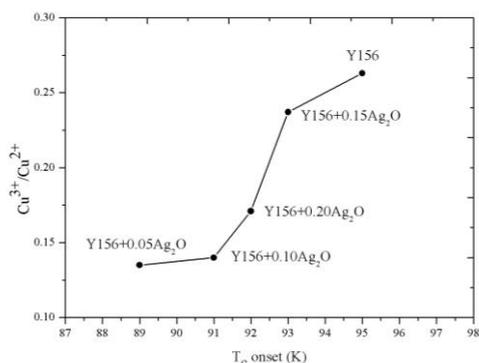


Figure 3 The ratio of Cu³⁺/Cu²⁺ versus the critical temperature onset of all samples.

From the fig. 3, the highest critical temperature in pure Y156 sample with Cu³⁺/Cu²⁺ equal to 0.26, and the lowest critical temperature was found in Y156 doped 0.05Ag₂O with Cu³⁺/Cu²⁺ equal to 0.14. Our results shown that the higher Cu³⁺/Cu²⁺ ratio, the higher critical temperature was found. This is in agreement with earlier report by Choy et al. [9]. According to the literature, the oxygen content is an important factor to improve the superconductor properties. The amount of

oxygen can be determined from the amount of Cu^{2+} and Cu^{3+} in superconductor. So the superconductor with the appropriate $\text{Cu}^{3+}/\text{Cu}^{2+}$ ratio, it had the high critical temperature.

4. Conclusions

We have prepared Y156 superconductor doped Ag_2O ($\text{YBa}_5\text{Cu}_6\text{O}_{13-y} + x\text{Ag}_2\text{O}$; $x = 0, 0.05, 0.10, 0.15, 0.20$) by solid state reaction. The calcinations and sintering temperature were at 900°C and annealing temperature was at 550°C . The highest critical temperature was in pure Y156 sample with T_c onset 95 K and the lowest critical temperature was in Y156 doped $0.05\text{Ag}_2\text{O}$ with T_c onset 89 K. We found that the surface of Y156 was improved by Ag_2O adding on the porous structure. Furthermore, the higher $\text{Cu}^{3+}/\text{Cu}^{2+}$ ratio, the higher critical temperature was found.

5. Acknowledgements

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