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Superconducting transitions of $LuBa_2Cu_3O_{7-z}$ heated under various atmospheres

M. S. Lee and H.K. Lee*

Kangwon National University, Chuncheon, Korea

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Abstract

The influence of quenching temperature, heating atmosphere and annealing time on superconducting characteristics has been studied for $LuBa_2Cu_3O_{7-z}$ compound which has been recently synthesized in a nearly phase-pure form. Resistivity measurements for the as-prepared sample heated at 300 ℃ in oxygen and subsequently quenched into liquid nitrogen revealed that there is no significant change in T_c. On the other hand, T_c of the sample slightly increased when the sample was heated at 300 ℃ either in air or in N_2 atmosphere. It was also found that T_c of the sample decreased when the annealing temperature in N_2 atmosphere increased above 400 ℃. The experimental results indicated that the as-prepared sample is under overdoped state. The enhanced superconducting transition observed in the samples after heating at 300 ℃ in air or N_2 atmosphere was discussed in conjunction with a slight removal of oxygen and ordering of oxygen atoms in the as-prepared sample.

Keywords: $LuBa_2Cu_3O_{7-z}$,Transition temperature , Overdoping, Heat treatment

1. INTRODUCTION

Soon after the discovery of high T_c superconductivity in $YBa_2Cu_3O_{7-z}$, it was found that Y in $YBa_2Cu_3O_{7-z}$ (Y-123) can be replaced by most of the rare earth (R) elements to form superconductors of the same crystal structure with T_c near 90 K except for R = Pr, Ce, Tb and Lu [1, 2], The $PrBa_2Cu_3O_{7-z}$ cuprate is known to be non-superconducting while the orthorhombic compounds of $CeBa_2Cu_3O_{7-z}$ and $TbBa_2Cu_3O_{7-z}$ does not form by solid-state reaction methods [3]. On the other hand, it has been shown that $LuBa_2Cu_3O_{7-z}$ prepared by standard solid-state reaction contains a second phase that accounts for at least half the intensity in the powder X-ray diffraction patterns [1, 2].

Hodorowicz et al. [4] reported an extensive investigation on the phase diagram of Lu-Ba-Cu-O system and concluded that the Lu-123 phase cannot be synthesized by direct combination of the reactants at temperatures between 900 and 1000 °C both in air and in pure O_2. However, it has been recently discovered that nearly phase-pure Lu-123 compound can be formed within a narrow range of oxygen partial pressure in the specific temperature range [5, 6]. In this work, we have investigated the effects of heat-treatment on superconducting properties of the newly synthesized phase-pure Lu-123 samples to obtain further information for optimization of superconducting properties of the sample.

* Corresponding author: hklee221@kangwon.ac.kr

2. EXPERIMENTALS

Polycrystalline samples with nominal composition of $LuBa_2Cu_3O_{7-z}$ were synthesized by a solid-state reaction technique described elsewhere [5]. The temperature dependence of the electric resistivity was measured by a standard 4-probe technique with silver paste as contacts within a temperature range of 10 K to room temperature. The room-temperature thermoelectric power was measured relative to copper electrodes by using a dc differential method and further calculated by correcting for the thermoelectric power of copper. Thermo-gravimetric analysis was carried out using a TA Inst. SDT-2960. The weight of the sample for analysis was about 20 mg and the heating rate was 10 °C /min in air atmosphere.

2. RESULTS AND DISCUSSION

Fig.1 shows the temperature dependence of electrical resistivity for $LuBa_2Cu_3O_{7-z}$ annealed at 300 °C for 10 h and 500 °C for 20 h in oxygen and subsequently quenched into liquid nitrogen. The as-prepared sample exhibits the onset transition temperature, T_c(onset) = 91.3 K and zero-resistivity temperature, T_c(zero) = 86.1 K and it can be seen that T_c(zero) of the sample hardly changed (within 1 K) after the annealing in oxygen atmosphere On the other hand, when the sample was heated at 300 °C in air atmosphere, T_c(zero) of the sample increased with the increase in the annealing times as shown in Fig.2. The

T_c(zero) values of the samples after annealing at 300 °C for 3 h and 14.2 h are 88.7 K and 91.0 K, respectively.

Fig.3 reveals the temperature dependence of electrical resistivity for $LuBa_2Cu_3O_{7-z}$ samples annealed at various temperatures in nitrogen and subsequently quenched into liquid nitrogen. This figure indicates that T_c(zero) at first increases slightly after annealing at 300 °C and then decreases with increasing annealing temperature above 400 °C.

For p-type cuprate superconductors, T_c is known to vary universally and empirically as an inverted parabolic function [7] of the hole concentration (p) in the superconducting planes with maximum T_c at $p = 0.16$. Both the change in oxygen content and the substitution of aliovalent elements in R-123 system bring about a change in hole concentration of the CuO_2 planes. Therefore, the variation of the critical temperature after thermal treatment under N_2 atmosphere may be associated with the change in oxygen content. To find more information on the change in oxygen content, a thermogravimetric analysis (TGA) was carried out on the as-prepared sample.

From Fig. 4, one can notice that the weight loss of the as-prepared sample takes place at temperature above 200 °C. The weight loss in the temperature between 200-400 °C is 0.24%. But the weight loss above about 400 °C becomes prominent as shown in Fig.4. Therefore, the decrease in T_c after annealing above 400 °C can be attributed to a decrease in oxygen content of the sample. A

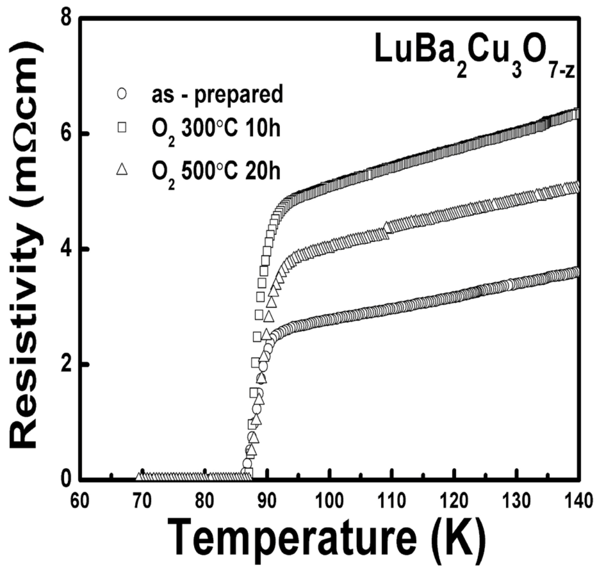

Fig. 1. Temperature dependence of electrical resistivity for $LuBa_2Cu_3O_{7-z}$ samples annealed at 300 °C for 10 h and 500 °C for 20 h in oxygen and subsequently quenched into liquid nitrogen.

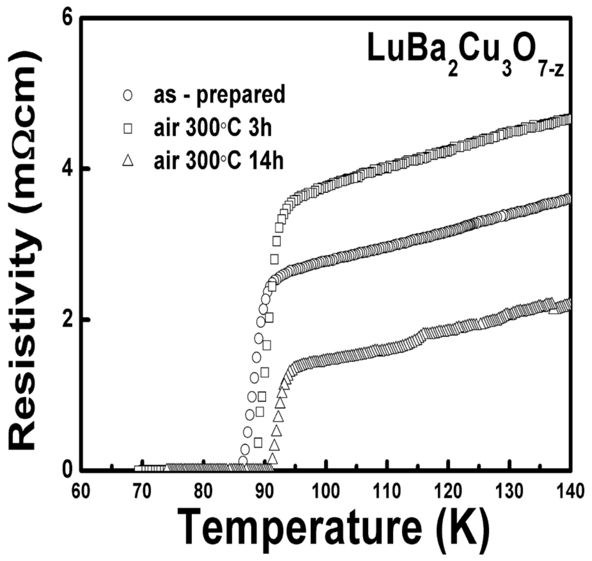

Fig. 2. Temperature dependence of electrical resistivity for $LuBa_2Cu_3O_{7-z}$ samples annealed at 300 °C for 3 h and 14.3 h in air and subsequently quenched into liquid nitrogen.

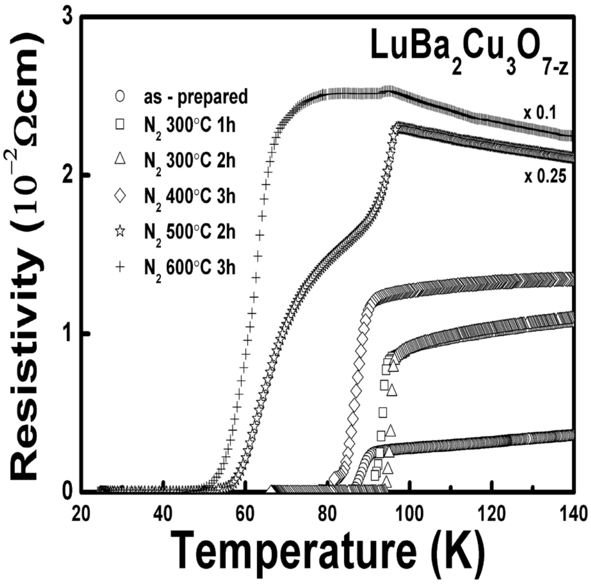

Fig. 3. Temperature dependence of electrical resistivity for $LuBa_2Cu_3O_{7-z}$ samples annealed at various temperature in nitrogen and subsequently quenched into liquid nitrogen.

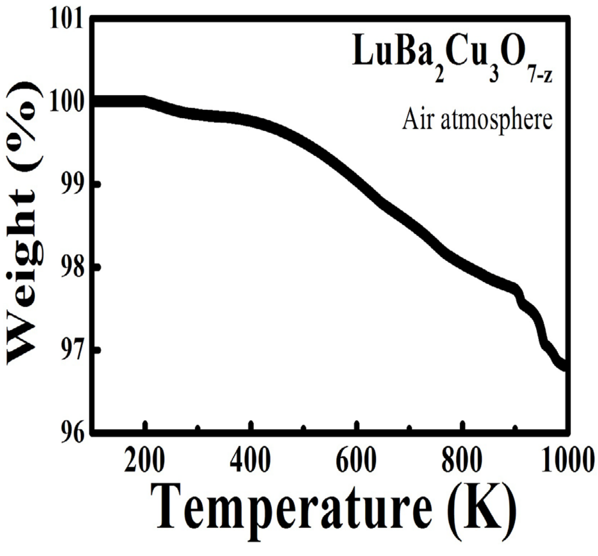

Fig. 4. Thermogravimetric curve for as-prepared $LuBa_2Cu_3O_{7-z}$. The heating rate was 10 ºC/min in air.

decrease in oxygen content in cuprate superconductor is known to result in a decrease in hole concentration in the CuO_2 planes. In order to address the change in hole concentration of the sample, room temperature thermoelectric power (TEP) measurements were performed because TEP is known to vary in a systematic manner according to the carrier concentration [8]. It has large positive values in the underdoped range and negative values in the overdoped range at room- temperature, with optimal doping being 1-2 ㎶/K. The room-temperature TEP for the as-prepared sample was -3.0 ㎶/K and the room-temperature TEPs for the samples after N_2 annealing at 300 °C for 1 h, 300 °C for 2 h, 400 °C for 3 h, 500 °C for 2 h and 600 °C for 3 h were 6.6, 6.1, 33.3, 42.9, and 61.4 ㎶/K, respectively. This result suggests that the as-prepared sample is overdoped state and N_2 annealing results in a decrease in hole concentration of the as-prepared sample. Therefore, the improvement of T_c after annealing at 300 °C in N_2 atmosphere can be explained by the decrease in hole concentration from the overdoped state and thereby reaching nearly optimum hole concentration. A similar change in T_c and hole concentration was also observed in overdoped $(Lu_{0.8}Ca_{0.2})Ba_2Cu_3O_z$ system [9, 10] after heat treatment under different temperatures and oxygen partial pressures including N_2 atmosphere.

On the other hand, the room-temperature TEPs for the samples after annealing at 300 °C for 3 h and 14.2 h in air were -3.1 and -3.0 ㎶/K, respectively. This result suggests a negligible change in hole concentration of the sample after the heat treatments in air. Therefore, in contrast to the case of the samples annealed at 300 °C in nitrogen atmosphere, the improvement of T_c(zero) for the samples annealed at 300 °C annealed in air cannot be attributed to the change in oxygen concentration. Related to this, Jorgensen *et al.* [11] reported that superconducting transition temperature in Y-123 system can be increased due to the local ordering of oxygen atoms with no significant changes in the average occupancies of oxygen sites. Therefore, the increase in T_c(zero) without significant change in hole concentration after annealing in air at low temperature of 300 °C is considered to be related with the local ordering of oxygen atoms.

2. CONCLUSIONS

The effects of heat treatment on superconducting properties of newly synthesized $LuBa_2Cu_3O_{7-z}$ have been investigated by characterizing the sample after annealing under various atmospheres, temperatures and times. It was observed that annealing under oxygen atmosphere below 500 °C hardly changed the superconducting transition temperature, while an annealing at 300 °C under air or N_2 atmospheres led to a slight increase in T_c(zero). We also observed that T_c of the Lu-123 sample decreased through the removal of oxygen when the sample is subjected to high temperature annealing above 400 °C in N_2 atmosphere. The increase in T_c observed for the sample annealed at 300 °C in N_2 atmosphere could be attributed to a slight deficiency in oxygen. However, the improvement of T_c(zero) for the samples annealed at 300 °C annealed in air could not be attributed to the change in hole concentration. From these results, it was concluded that the as-prepared sample is overdoped state.

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