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The Effects of Nonstoichiometry in High - T_c Superconducting Properties of $YBa_2Cu_{3+x}O_{7-\delta}$

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Abstract

Polycrystalline samples of high T_c superconductor $YBa_2Cu_{3+x}O_{7-\delta}$ with $0 \leqslant x \leqslant 0.5$ were grown. The effects of nonstoichiometry on the resistance slope above transition temperature T_c and X-ray diffraction patterns for these samples were investigated. In the diffraction patterns obtained, apart from some new features in the peaks, these polycrystalline samples show predominantly 123 phase.

Key Words: Effects of nonstoichiometry, high T_c superconductor, polycrystalline samples.

1. Introduction

After the discovery of the 90K superconducting phase $YBa_2Cu_3O_{7-\delta}$ [1-2], it was found that other superconducting materials can be obtained by substitution [3-8]. Although, many models [9] have been proposed for the high T_c superconductivity of Y-Ba-Cu-O it is yet not possible to favour one model over the other.

In the past few years, attempts were made to study the resistivity and X-ray diffraction pattern of $YBa_2Cu_3O_{7-\delta}$ superconductor. However, the results so far obtained are still far from being able to account for the complexity of these materials. In particular, no author has reported the effects of nonstoichiometry on resistance slope above transition temperature T_c in $YBa_2Cu_{3+x}O_{7-\delta}$ with $0 \leqslant x \leqslant 0.5$. Kishio et al., [10] determined the equilibrium value of oxygen nonstoichiometry of $YBa_2Cu_3O_{7-\delta}$ through thermogravimetric measurement in conjunction with chemical analysis. Recently, Mosqueira et al., [11] reported the resistivity versus temperature curve for the $YBa_2Cu_3O_{7-\delta}$ compounds and found the transition temperature at T_c = 91.8 K. Therefore, the study and interpretation of the reesistance and X-ray difraction in the high T_c superconductors are still crucial for practical purposes.

In this paper, the effects of the nonstoichiometry on resistance above the transition temperature T_c in the superconducting $YBa_2Cu_{3+x}O_{7-\delta}$ with $0 \leqslant x \leqslant 0.5$ is reported and the X-ray patterns obtained for these polycrystalline samples are studied. The layout of the paper is as follows: In Sec. 2 the experimental technique is described. The results and discussions is presented in Sec. 3 and the conclusion is drawn in Sec. 4.

2. Experimental Technique

The samples were prepared by mixing high purity samples of Y_2O_3,$BaCO_3$ and CuO powders on proper stoichiometry. The powders were mixed in isopropanol alcohol and were thoroughly ground with pestle mortar and pestle, then heated in is flowing oxygen atmosphere at 850°C. The oven was later quenched and the calcinated samples were mixed with isopropanol alcohol before re-grinding. The CuO powder was added to some of the powdered samples and the structures obtained became $YBa_2Cu_{3.1}O_{7-\delta}$, $YBa_2Cu_{3.3}O_{7-\delta}$ and $YBa_2Cu_{3.5}O_{7-\delta}$. Later the samples were pelletized and calcined at 950°C for 12 days before cooling for intermediate temperatures and then quenched.

The resistance was measured down to 91 K using a standard four-probe method and Si-diode thermometry.

Powder X-ray diffraction (XRD) was performed using a Siemens D_{500} diffractometer with a monochromatic CuK_α. radiation source. Intensities were recorded, as a function 2θ.

Compositions as follows were tested: $YBa_2Cu_3O_{7-\delta}$, $YBa_2Cu_{3.1}O_{7-\delta}$, $YBa_2Cu_{3.3}O_{7-\delta}$, and $YBa_2Cu_{3.5}$ $O_{7-\delta}$.

3. Results and Discussions

The electrical resistance measurements were carried out by a standard four-probe technique. The temperature was monitored by Si-diode thermometer which was attached behind the sample holder. Figure 1 shows the temperature dependence of the resistance for $YBa_2Cu_{3+x}O_{7-\delta}$, ($0 \leqslant x \leqslant 0.5$) and the nonstoichiometry effect on the resistance slope above the transition temperature T_c. The samples show sharp transitions at 91 K, implying that these samples have an almost pure superconducting phase. The resistance at 300 K increases with increasing copper content and, for all samples, the temperature dependence of the resistance shows metallic character. The sample with x = 0 attained the on set temperature at the minimum resistance (~0.47 mΩ), the other samples with $0.1 \leqslant x \leqslant 0.5$ showed higher resistance at the on-set temperature. It is interesting to observe the crossover in the crystals with x = 0.3 and x = 0.5 at T ∼ 175 K. This behaviour explains why the sample with x = 0.5 exhibited lower resistance than the sample with x = 0.3 at the on-set temperature. Also, it is observed that the resistance at the on-set temperature is the same in the samples with x = 0.1 and 0.5.

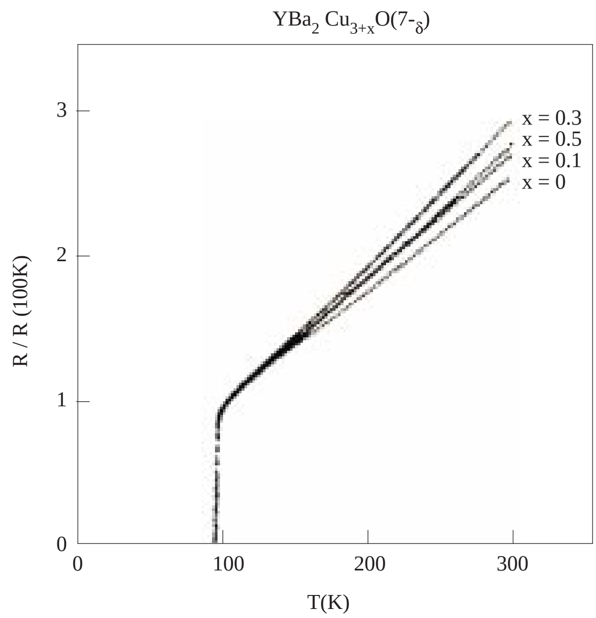

Figure 1. Graphs of Resistance versus Temperature for $YBa_2Cu_{3+x}O_7$, (x = 0, 0.01, 0.3, 0.5)

The CuK_α X-ray diffraction patterns of the polycrystals $YBa_2Cu_{3+x}O_{7-\delta}$ with $0.1 \leqslant x \leqslant 0.5$ are shown in Figure 2. The peaks obtained in this figure confirm the structure pattern of the 123 $YBa_2Cu_{3+x}O_{7-\delta}$ phase. This phase has the strongest peaks in the samples with x = 0. It is also interesting to point out that most of the previous works [12-14] reported the strongest peak at $2\theta = 32.5°$ in the (013) plane, for the range $20 \leqslant 2\theta \leqslant 40$, whereas the present work shows the strongest peak at $2\theta = 38.30°$ in the (005) plane which agrees with ref. [15] for $YBa_2Cu_3O_{7-\delta}$ sintered discs, and a small peak was observed at the same 2θ for $YBa_2Cu_3O_{7-x}$ powders. In the samples $YBa_2Cu_{3.1}O_{7-\delta}$ and $YBa_2Cu_{3.5}O_{7-\delta}$, the peaks at $2\theta = 22.65°$ and

31.00° in the (010) and (004) planes, respectively, have identical strength. This similarity in the magnitude of the peaks reconfirm the identical resistance exhibited by both samples (x = 0.1 and 0.5) at the on-set temperature which was shown in Figure 1. On the other hand, the peaks at $2\theta = 38.25°$ and 32.55° which correspond to the (005) and (103) planes respectively, decrease as x increases. This observation is associated with the trend exhibited at high temperature ($T \geqslant 175$ K) in the resistance measurements whereby the resistance increases with increase in x.

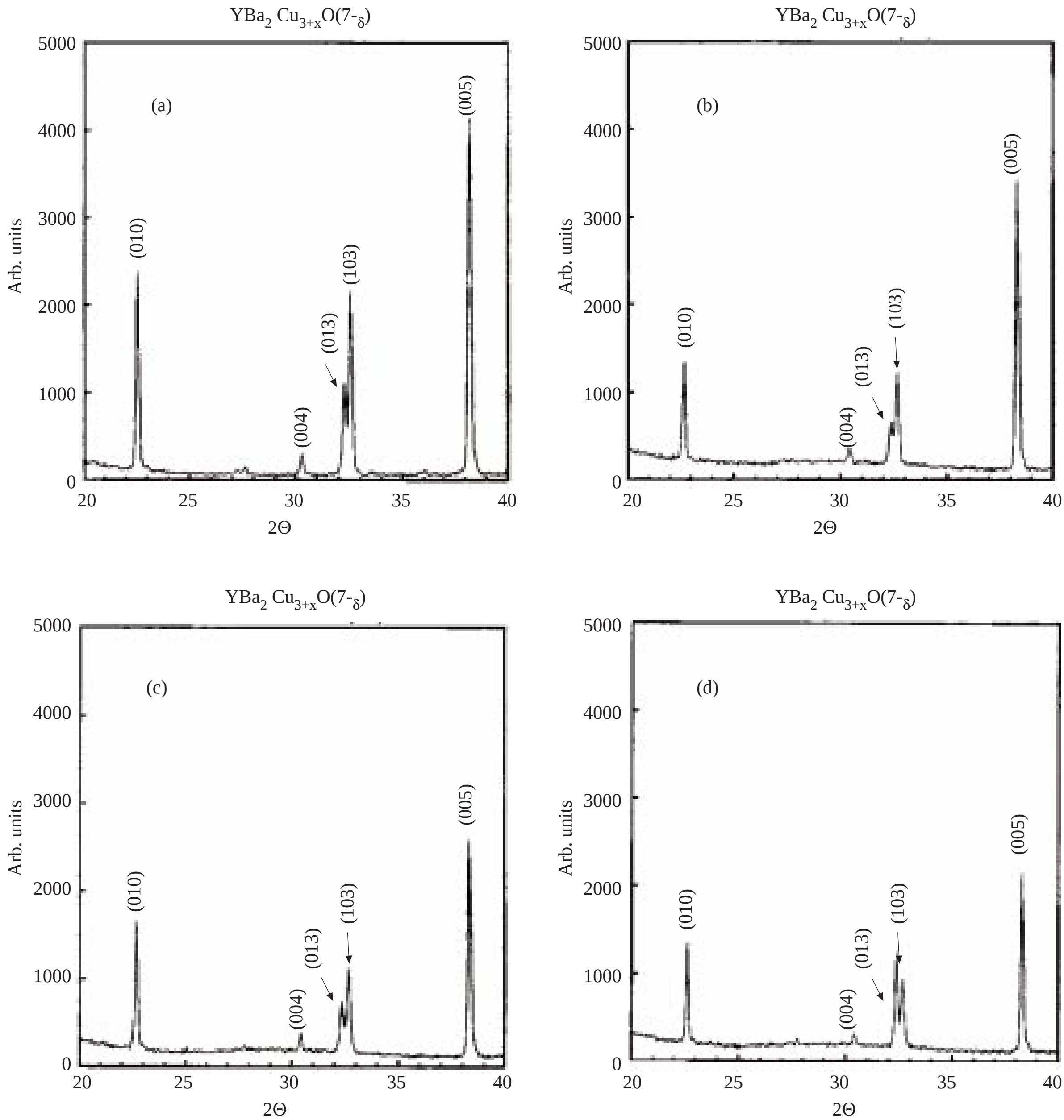

Figure 2. X-ray Diffraction Patterns
(a) $YBa_2Cu_3O_{7-\delta}$ (x = 0)
(b) $YBa_2Cu_{3.1}O_{7-\delta}$, (x = 0.1)
(c) $YBa_2Cu_{3.3}O_{7-\delta}$, (x = 0)
(d) $YBa_2Cu_{3.5}O_{7-\delta}$, (x = 0.5)

4. Conclusion

In this paper, it is shown that high quality polycrystalline $YBa_2Cu_{3+x}O_{7-\delta}$ with $0 \leqslant x \leqslant 0.5$ can be grown. The resistance was measured down to 91 K with a standard four-probe method and Si-diode thermomether. The X-ray diffraction pattern was obtained with Siemens D_{500} diffractometer with a monochromatic CuK_α radiation. The results obtained in the resistance measurements for the four samples (x = 0, 0.1, 0.3, 0.5) show the effects of the nonstichiometry in the resistance R(T) slope above the transition temperature T_c($\sim$91 K), as well as the variation in the resistance at the on-set temperature. Apart from the some new features in the peaks, the samples predominantly show the 123 phase.

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References

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

[2] P. H. Hor, L. Gao, R. L. Meng, Z. J. Huang, Y. Q. Wang, K. Forster, J. Vassilious, C. W. Chu, M. K. Wu, J. R. Ashburn and C. J. Thorng, *Phys. Rev. Lett.*, **58**, (1987), 911.

[3] K. Yamaya, K. Kodaira, M. Higuchi, T. Ishibashi, M. Ido, K. Nomura and T. Sambongi, *Jpn. J. Appl. Phys.*, **26**, (1987), L1095.

[4] H. Tajima, A. Maeda, M. Kobayashi, T. Satake, H. Shimada, S. Miyama and H. Kuroda, *Jpn. Appl. Phys.,* **26**, (1987), L845.

[5] A. Oota, Y. Kiyoshima and T. Hioki, *Jpn. Phys.*, **26**, (1987), L1356.

[6] I. Sankawa, M. Sato and T. Konaka, *Jpn. J. Appl. Phys.*, **28**, (1988), L28.

[7] K. Takita, H. Akinaga, H. Katoh, H. Asano and K. Masuda, *Jpn. J. Appl. Phys.,* **27**, (1988), L67.

[8] E. Talik, J. Szade and J. Heimann, *Physica C*, **165**, (1990), 434.

[9] L. F. Matheiss, *Phys. Rev. Lett.*, **58**, 1024 (1987); J. Yu, A. J. Freeman and J-H. Xu: ibid. 1035 (1987); B. V. Chakraverty, D. D. Sarma and C. N. R. Rao, *Physica C,* **413**, 156 (1988); J. M. Wheatley, T. C. Hsu and P. W. Anderson, *Phys. Rev.,* **B37**, 5897 (1988).

[10] K. Kishio, J. Shimoyama, T. Hasegawa, K. Kitazawa and K. Fueki, *Jpn. J. Appl. Phys.*, **26**, (1987), L1228.

[11] J. Mosqueira, J. A. Weira and F. Vidal, *Physica C*, **229**, (1994), 301.

[12] R. K. Siddique, *Physica, C.*, **228**, (1994), 306.

[13] A. M. M. Barus and J. A. T. Taylor, *Physica C*, **225**, (1994), 374.

[14] C-J. Kim, K-B Kim and G-Won Hong, *Physica C*, **232**, (1994), 163.

[15] A. Iwase, N. Masaki, T. Iwata and T. Nihira, *Physica C*, **174**, (1994), 321.