Study of quenched YBa$_2$Cu$_3$O$_{7-y}$-WO$_3$ superconductors

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Abstract. Superconductivity and structure of YBCO with added WO_3 have been investigated. The result indicates that the addition of up to 5% WO_3 does not destroy the superconducting properties of YBCO. It is found that the samples with the WO_3 addition directly quenched from 900 °C in an oxygen atmosphere have a transition temperature of 88 K. The tetragonal-to-orthorhombic transformation takes place within a short time in the YBCO–WO_3 samples. Extra peaks can be observed in the x-ray diffraction pattern, indicating that there is a second phase in the samples.

1. Introduction

It has been found that in the $YBa_2Cu_3O_{7-y}$ (123) (YBCO) superconductors the crystal symmetry is a tetragonal type with space group $P4/mmm$ for $y = 1$ and is an orthorhombic type with space group $Pmmm$ for $y = 0$. The critical temperature T_c is highly sensitive to the oxygen content of the YBCO system. T_c decreases with decrease in the oxygen content until the superconductivity finally disappears. On the other hand, the crystal structure of YBCO changes from orthorhombic type to tetragonal type when the oxygen content decreases [1–3].

It has been widely reported that the oxygen content is closely related to the sintering process. As the orthorhombic transformation takes place at temperatures between 600 and 700 °C by obtaining a sufficient amount of oxygen, an additional heat treatment at 600–700 °C in oxygen or slow cooling in flowing oxygen is required to obtain the orthorhombic superconducting phase [4, 5]. After having studied the properties of the quenched YBCO system, Zhang *et al* [6] found that the samples quenched from temperatures above 880 °C had a typical tetragonal structure, showing no superconductivity above 4.2 K. However, additional annealing can cause severe inter-diffusion between the substrates and the 123 film. Consequently, T_c significantly decreases to a temperature much lower than 77 K. Here, we report the influence of the WO_3 addition on the superconducting properties and the structure of YBCO.

2. Experimental

The YBCO powder was fabricated by the coprecipitation method. The WO_3 powder was well mixed with the YBCO powder according to the appropriate ratio. The concentration of

$\mathrm{WO_3}$, i.e. X, is defined as follows:

$$X = W(\mathrm{WO_3})/\left[W(\mathrm{YBCO}) + W(\mathrm{WO_3})\right] \times 100\%$$

where $W(\mathrm{WO_3})$ and $W(\mathrm{YBCO})$ represent the weights of $\mathrm{WO_3}$ and YBCO powder, respectively. Then, the mixture was pressed and calcined at 900 °C for 24 h. Finally, the samples followed three different procedures:

(1) they were quenched to room temperature in flowing oxygen:
(2) they were slow cooled to room temperature at 2 °C min^{-1};
(3) they were quenched in a nitrogen atmosphere.

X-ray diffraction patterns were obtained on a Philips-1700 diffractometer with Cu Kα radiation at room temperature. The AC susceptibility was measured using mutual induction bridges.

3. Results and discussion

Figure 1 gives the x-ray diffraction patterns for the samples quenched from 900 °C in oxygen. It is found that an orthorhombic structure can be observed for the $\mathrm{WO_3}$-doped samples, while the phase of the undoped YBCO quenched from 900 °C is tetragonal. This result suggests that the tetragonal-to-orthorhombic phase transition is accelerated by the $\mathrm{WO_3}$ addition. Moreover, the peak position in the spectra does not change with increase in $\mathrm{WO_3}$. As seen in figure 1, some extra peaks at $2\Theta = 30.2°$, 35.7° and 43.4° are observed and their intensities are strengthened as the amount of $\mathrm{WO_3}$ is increased. This result reveals the presence of a second phase in the specimens. The impurity may be a Y–Ba–W–O compound. When the $\mathrm{WO_3}$ content is greater than 3%, another peak at $2\Theta = 18.5°$ can be seen.

It is well known that $\mathrm{WO_3}$ has many oxygen-deficient states $2 \leqslant x \leqslant 3$. The crystal structure of $\mathrm{WO_3}$ has monoclinic, orthorhombic, tetragonal and cubic symmetries for different x-values [7]. These extra peaks in our spectra may be related to a structure with a higher symmetry than monoclinic, but the structure has not been identified and is under investigation.

X-ray diffraction patterns for the slowly cooled samples are shown in figure 2. All samples have an orthorhombic structure with some additional peaks. The positions and intensities of these peaks are similar to those of the specimens quenched from 900 °C in oxygen. The lattice constants were calculated by the least-squares method. As shown in table 1, the lattice parameters a, b and c are not affected by the mixing with $\mathrm{WO_3}$.

Table 1. Lattice constants of the slow-cooled samples.

No	a (Å)	b (Å)	c (Å)
C0	3.828	3.872	11.722
C1	3.820	3.867	11.704
C2	3.827	3.867	11.695
C3	3.825	3.864	11.705
C4	3.825	3.869	11.689

Note: C0 = YBCO, C1 = YBCO + 0.5% $\mathrm{WO_3}$, C2 = YBCO + 1% $\mathrm{WO_3}$, C3 = YBCO + 3% $\mathrm{WO_3}$, C4 = YBCO + 5% $\mathrm{WO_3}$.
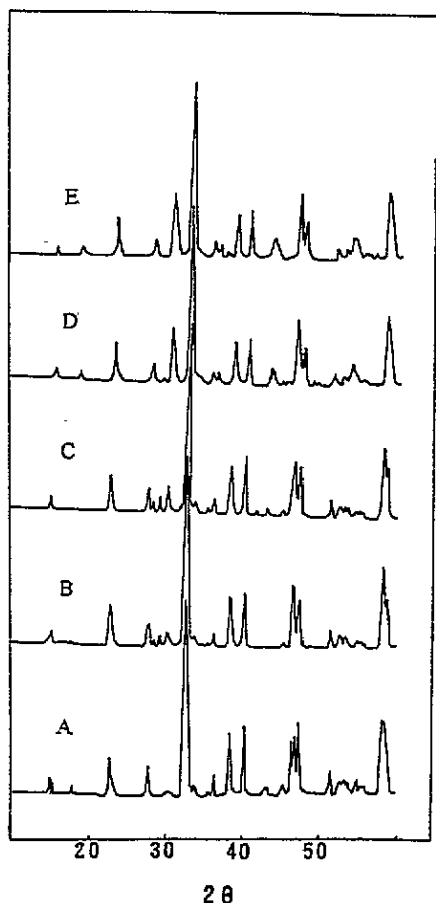
Figure 1. X-ray diffraction pattern for samples quenched in oxygen. A: YBCO; B: YBCO + 0.5% WO₃; C: YBCO + 1% WO₃; D: YBCO + 3% WO₃; E: YBCO + 5% WO₃.
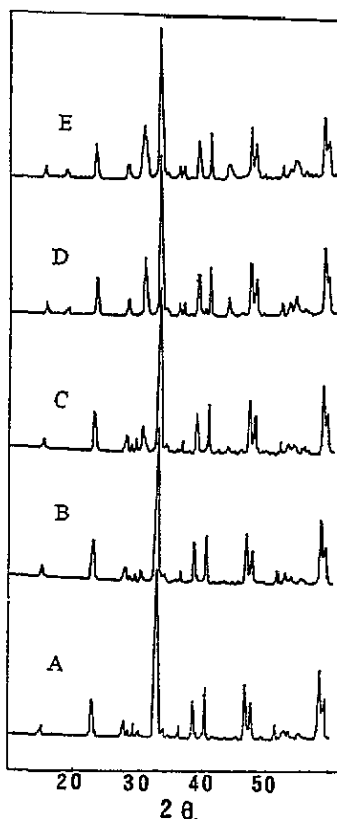
Figure 2. X-ray diffraction pattern for the slow-cooled samples. A: YBCO; B: YBCO + 0.5% WO₃; C: YBCO + 1% WO₃; D: YBCO + 3% WO₃; E: YBCO + 5% WO₃.

Figure 3 illustrates the AC susceptibility of the slowly cooled samples. T_c for all specimens except for the sample with 5% WO₃ is around 90 K and falls slightly with increasing WO₃ content. The critical temperatures of the specimens quenched in oxygen are given in figure 4. From figure 4, it can be observed that undoped YBCO shows no sign of the Messiner effects above 63 K; however, the transition temperatures of the doped samples are found to be around 88 K. This results suggests that superconductivity can be created by quenching of the YBCO–WO₃ sample in flowing oxygen from 900 °C. From the above discussion, it can be inferred that slow cooling is not necessary to obtain the YBCO–WO₃ superconductor with a critical temperature of 90 K. Thus, the severe diffusion between the substrate and YBCO thin film caused by additional annealing can be avoided.

The reason why the samples quenched from 900 °C in oxygen can show superconductivity above 88 K is still unknown. Maybe the temperature of the tetragonal-to-orthorhombic transformation is increased owing to the WO₃ addition. If this is true, the samples quenched in a nitrogen atmosphere should be the orthorhombic superconducting phase, but the specimens quenched from 900 °C in a nitrogen atmosphere are of tetragonal
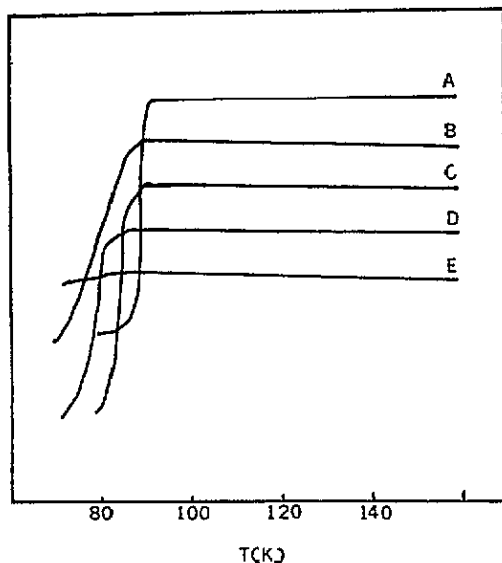
Figure 3. AC susceptibility of the slow-cooled samples. A: YBCO; B: YBCO + 0.5% WO₃; C: YBCO + 1% WO₃; D: YBCO + 3% WO₃; E: YBCO + 5% WO₃.

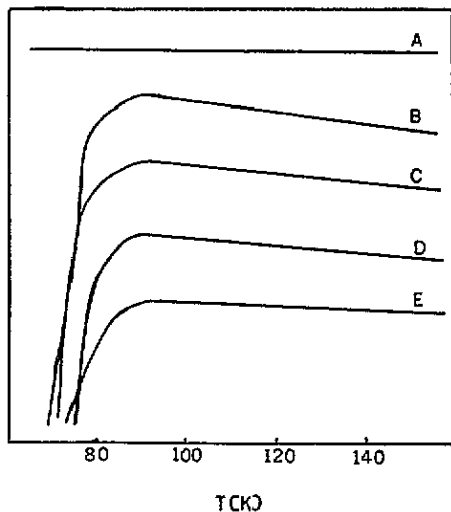Figure 4. AC susceptibility of the samples quenched in oxygen. A: YBCO; B: YBCO + 0.5% WO₃; C: YBCO + 1% WO₃; D: YBCO + 3% WO₃; E: YBCO + 5% WO₃.

structure and non-superconductors. This clearly demonstrates that the temperature of the tetragonal-to-orthorhombic transformation is not raised owing to the addition of WO_3. We consider that the rate of absorption of oxygen may be improved by the WO_3 addition and therefore the samples quenched in oxygen become superconducting.

It is not clear what accelerates the rate of oxygen absorption. The second phase in the samples may be responsible for this, but there is no direct proof and it should be further investigated.

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