dian J. Phys. 76A (1), 83-85 (2002)

IJP A
— an international journal

Synthesis of Y-123 superconductor without Oxygen annealing

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bstract This paper reports the synthesis of $YBa_2Cu_3O_{7-\delta}$ superconductors without external oxygen annealing. Polycrystalline samples are nthesized with nominal compositions. $YBa_2Cu_3Hg_xO_y$ $(0 \leq x \leq 0.3)$ The synthesis was carried out in an open atmosphere by two-stage, solid-state action method Initially, the stoichiometric composition of Y_2O_3, $BaCO_3$ and CuO was calcined twice at 925 °C for 24 hours and then the pellets of gO added Y-123 compound were sintered at 650 °C for 24 hours The X-ray diffraction patterns of the samples revealed monophasic Y-123 nature ithout any trace of Hg or related materials The superconducting behaviour was studied by low temperature *R-T* measurement The HgO added samples owed metallic behavior followed by the superconducting transition Some of the HgO added samples exhibited onset superconductivity around 100K he scanning electron micrographs of these samples showed non-uniform grain size with substantial grain inter-diffusion.

eywords : Y-123 superconductor, synthesis, solid-state reaction method

ACS No. 74 72 Bk

. Introduction

is well known fact that superconducting properties of the eramic superconductors are sensitive to the oxygen content of he sample. In particular, Y-123 compound exhibits uperconducting properties only when it is annealed in oxygen tmosphere [1-4]. This means that oxygen annealing is essential or Y-123 material and its transition temperature is totally lependent on the oxygen stoichiometry. An alternate way to icrease the oxygen content is to use some material as an internal ource of oxygen [5-7]. In this respect, HgO can be considered s a potential material due to its lower decomposition temperature 476°C) and high oxygen ambient created during decomposition. uch type of effect has already been studied with BiSrCaCuO ompounds [8-11] and also in YBCO compounds [12, 13]. In this aper, we report a novel technique for the synthesis of Y-123 ompound through the addition of HgO. It has been reported hat HgO decomposes into Hg metal and atomic oxygen during eating [14]. The Hg escapes from the matrix leaving the crystal naltered and atomic oxygen released during decomposition rovides an excellent ambient for the formation of a stoichiometric YBCO.

2. Experimental

The polycrystalline samples were prepared by solid-state reaction method in which appropriate quantities of high-purity Y_2O_3 (99.99%), $BaCO_3$ (99.98%) and CuO (99.99%) were weighed according to formula unit $Y_1Ba_2Cu_3O_{7-\delta}$, and ground in an agate mortar. The mixture was calcined in air at 925 °C for 24 h. The calcined mass was crushed, ground and recalcined for another 24 h. HgO was then added to this calcined powder in stoichiometric ratio 0, 0.1, 0.2 and 0.3 and mixed thoroughly. Circular pellets of 12-mm diameter were prepared by applying hydraulic pressure of 5 ton in a dye. These specimens were sintered in air at 650°C for 24 h, followed by furnace cooling.

All the samples were characterized by X-ray powder diffraction technique to identify the phase formation. For this purpose, Shimadzu Diffractometer (Model XRD-6000) with CuK_α radiation was employed in the range $5^0 \leq 2\theta \leq 60^0$. The superconducting behavior of the samples was studied by measuring electrical resistance as a function of temperature using the standard four-probe method. The low temperature measurements were carried out by using a closed-cycle He cryostat (CTI) with Lakeshore (model 330) temperature controller. A dc current of 10 mA was passed through the samples by a Kithley constant current source (Model 220) and voltage was

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measured by Kithley nano-voltmeter (Model 181). The microstructural features of the samples were studied through Scanning Electron Microscopic technique by using Cambridge stereoscan (ST50 MK3) microscope.

3. Results and discussion

The X-ray diffractograms of all the samples synthesized in the present work are shown in Figure 1. The diffraction peaks were compared with the standard JCPDS data of $Y_1Ba_2Cu_3O_y$ and with all the possible phases Y_2BaCuO_5, $BaCUO_2$, Ba_2CuO_3, $BaCO_3$, CuO, Y_2O_3 and BaO. Based on this, it has been found that all the diffraction peaks correspond to Y-123 phase only. No peak corresponding to impurity or mercury related phase was observed. The XRD patterns also reveal the change in the intensity of the diffraction peaks with variations in the composition of HgO. This may be the consequence of more or less absorbtion of oxygen. A systematic increase in the intensity of (013) and (103) peaks with increasing HgO concentration is the indication of the occurrence of superconducting orthorhombic phase.

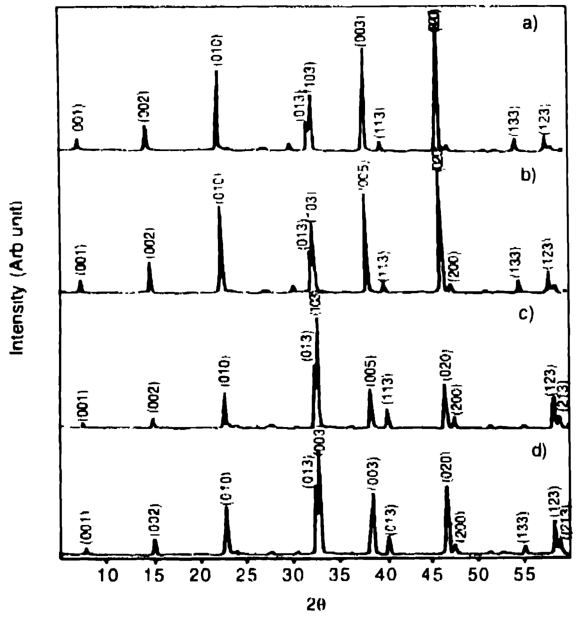

Figure 1. XRD patterns of $Y_1Ba_2Cu_3Hg_xO_y$ samples (a) $x = 0$, (b) $x = 0.1$, (c) $x = 0.2$ and (d) $x = 0.3$.

The variation of resistance of the samples prepared with temperature, for $x = 0, 0.1, 0.2, 0.3$ is shown in Figure 2. The samples prepared without HgO showed semiconducting behaviour followed by non-superconducting metallic behaviour. It is a well-documented fact that the as- synthesized compound has oxygen deficiency $\delta \geq 0.5$ and they exhibit non-superconducting tetragonal phase [1,4]. Further, it can be seen that with $x = 0.1$, there is an improvement in the low temperature conductivity of the samples. However, the samples could not exhibit superconductivity even at very low temperature. On the other hand, the samples with $x = 0.2$ and 0.3 have shown superconducting nature, although with broad transitions. The samples with $x = 0.2$ showed a metallic behaviour followed b superconducting transition with $T_{c\,(onset)}$ value 130 K and $T_{c\,(zer}$ value 51K. For $x = 0.3$, the curve showed semiconductor t

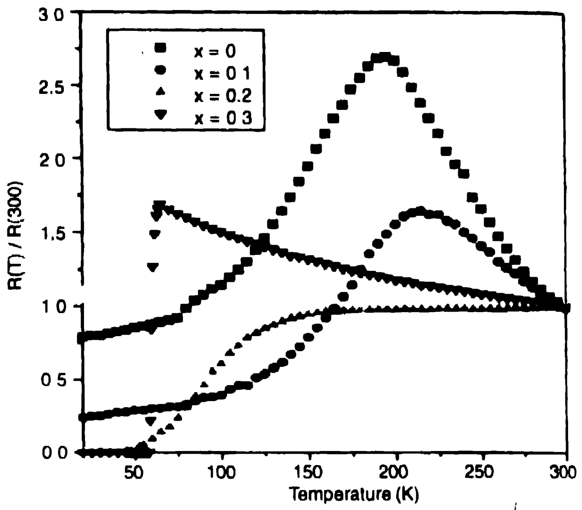

Figure 2. Normalised resistance plotted against temperature $Y_1Ba_2Cu_3Hg_xO_y$ samples

superconductor transition at around 60K with $T_{c\,(zero)}$ value 56K. The gradual improvement in the resistive behaviour these samples with x, is an indication that the oxygen conte are increasing. As per mechanism of thermal decomposition HgO at high temperature [14], the proceeding reaction is

$$HgO\,(s) \rightarrow Hg\,(g) + O\,.$$

Thus, the atomic species of oxygen being liberated throu the decomposition must be incorporated in YBaCuO syst while the Hg vapour escapes out. As the HgO compositi increases, more and more oxygen is made available, which resu in improvement of the resistive behaviour. Therefore, it possible to synthesize superconducting Y-123 compou

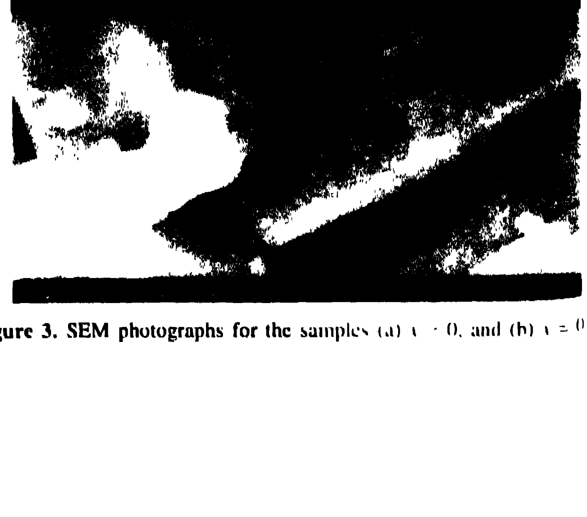

Figure 3. SEM photographs for the samples (a) $x = 0$, and (b) $x = 0$

without the external oxygen annealing. Further, it may be possible to tune the broad transitions precisely to yield an enhanced T_c value through proper optimization of HgO concentration and the process parameters.

The scanning electron micrographs of the $x = 0$ and $x = 0.3$ samples are shown in Figure 3. The micrograph reveals the granular structure of the material. In both the cases, plate-like grains of approximately 10 micron size were observed. Inspite of low sintering temperature (650°C), the samples showed better grain connectivity.

. Conclusions

In conclusion, we have found an internal oxygen source for Y-123, which appears to solve the problem of surface barrier to external oxygen diffusion. The addition of HgO leads to uniform oxygen stoichiometry within the bulk material during sintering. As a result, Y-123 material exhibited superconductivity even though no oxygen annealing was carried out. Mercury is not found to be incorporated in the system nor does it alter the crystal structure. By analyzing the R-T measurement results, it is clear that a good quality superconducting Y-123 samples could be prepared without oxygen annealing as the value of T_c increases with the increase in the x-value for HgO addition. Thus, HgO addition can be treated as a powerful tool for auditing the oxygen content of Y-123 samples.

Acknowledgment

The authors are thankful to the University Grants Commission for providing financial support through R&D and Special Assistance Programs.

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