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HgO-added $YBa_2Cu_3O_{7-\delta}$ superconductors

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Abstract. The HgO-added $YBa_2Cu_3O_{7-\delta}$ (YBCO) superconductor has been studied for its structural and superconducting properties. Polycrystalline YBCO samples were synthesized through solid-state reaction method by adding HgO in different concentrations without using oxygen annealing. All the samples showed a sharp superconducting transition temperature around 90 K. The X-ray diffraction patterns of all the samples revealed monophasic Y-123 nature. The structural studies were carried out by neutron scattering and Rietveld analysis. The neutron scattering revealed that Hg is not incorporated in the Y-123 system and has shown optimum oxygen concentration. The significant role played by the HgO is to provide oxygen ambient through its decomposition, thus changing the oxygen balance in favour of high Cu-valence state.

Keywords. Superconductivity; solid-state reaction method; HgO addition; neutron diffraction.

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1. Introduction

It is widely known that the superconducting properties and structural stability of Y-123 compound depend strongly on the oxygen content. Moreover, these compounds exhibit superconductivity only when annealed in the oxygen ambient [1–4]. An alternative way to improve the oxygen content is the use of some internal source of oxygen [5–7]. In this respect the HgO can be considered as a potential material because of its lower decomposition temperature (476°C) and high oxygen ambient created during decomposition. It decomposes into mercury, which escapes from the matrix leaving the crystal unaltered and oxygen, which provide an excellent ambient for the formation of a stoichiometric oxide compound [8–14]. We have already reported the synthesis of $YBa_2Cu_3O_y$ with the addition of HgO [15–17]. In this paper we report the studies on the structural and the superconducting properties of HgO-added Y-123 compound.

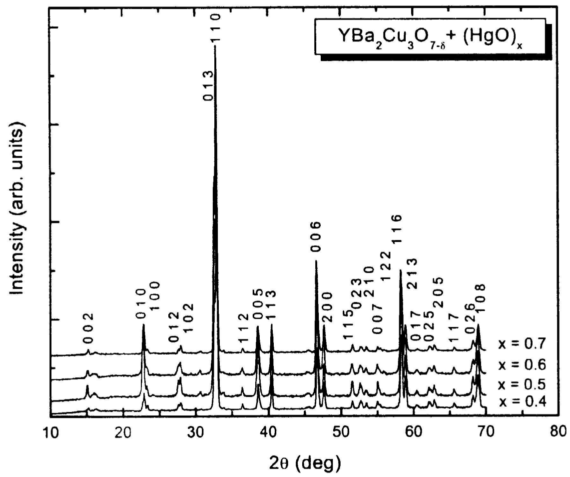

Figure 1. XRD patterns of HgO ($x = 0.4$, 0.5, 0.6 and 0.7) added Y-123 sintered at 950°C.

2. Experimental

The polycrystalline samples of $YBa_2Cu_3O_{7-\delta}$ were synthesized by solid-state reaction method. A stoichiometric amount of Y_2O_3, $BaCO_3$ and CuO was ground thoroughly. The calcinations were done twice at 925°C for 24 h with intermediate grinding. HgO was added to the calcined mass in the molar ratio $x = 0.4, 0.5, 0.6$ and 0.7 and were pressed to pellets. The pellets were sintered at 950°C for 24 h, followed by furnace cooling.

The identification of the phases in the samples was done by X-ray powder diffraction technique at room temperature in which CuK_α radiation was employed in the range $10° \leq 2\theta \leq 70°$ using Shimadzu XRD 6000 diffractometer. The resistance as a function of temperature was measured by the standard four probe method. The neutron powder diffraction data were collected at room temperature for the sample with $x = 0.4$ on the powder diffractometer ($\lambda = 1.249$ Å) at Dhruva reactor, BARC. The data were analyzed in orthorhombic Pmmm space group by using FullProf [18] to obtain structural parameters, including oxygen site occupancies for different oxygen sites.

3. Results and discussion

The X-ray diffractograms of all the samples synthesized with HgO addition are shown in figure 1. The diffraction peaks were compared with standard JCPDS data of $YBa_2Cu_3O_y$. It has been found that all the diffraction peaks correspond to Y-123 phase only, irrespective of the different x ratio. No peak corresponding to impurity or mercury related phase was observed. This reveals the mono-phasic nature of all the samples.

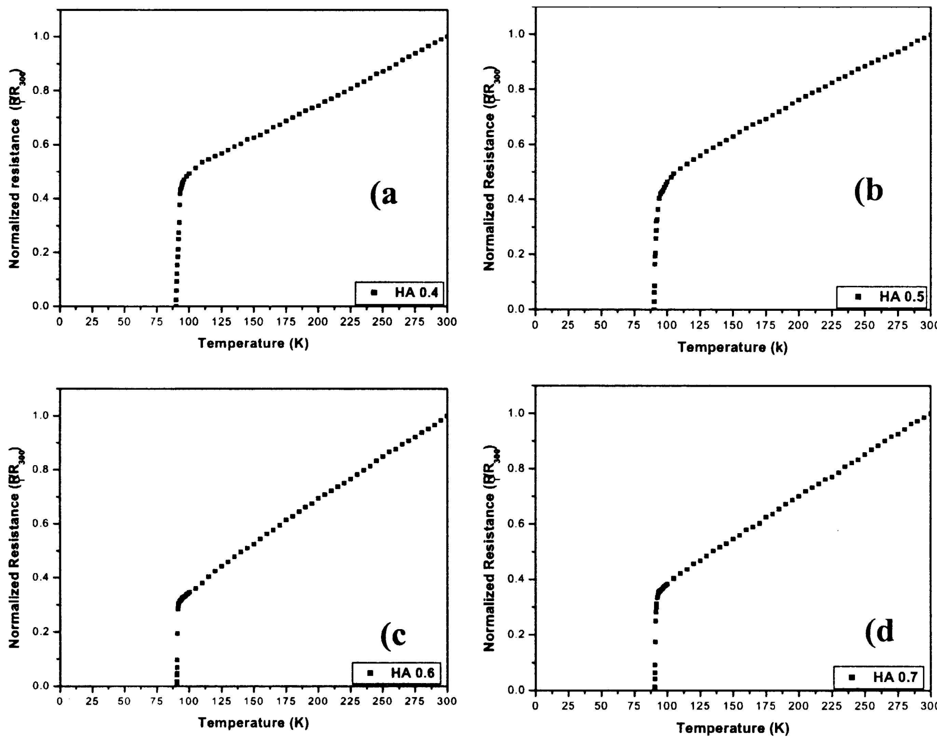

Figure 2. Normalized resistance plotted with temperature of HgO-added Y-123 samples (**a**) $x = 0.4$, (**b**) $x = 0.5$, (**c**) $x = 0.6$ and (**d**) $x = 0.7$ sintered at 950°C.

Table 1. Structural parameters for the 0.4 HgO-added Y-123 sample sintered at 950°C. Rietveld refinements were done in the orthorhombic Pmmm space group and the refined unit cell parameters are $a = 3.82290$ Å, $b = 3.88845$ Å, $c = 11.70163$ Å and $V = 173.947$ Å^3. R_p (%) = 3.55, R_{wp} (%) = 3.80, R_e (%) = 4.46, $\chi^2 = (R_{wp}/R_e)^2 = 1.37$.

Atom	x	y	z	B	N
Y	0.5	0.5	0.5	0.405	0.125(0)
Ba	0.5	0.5	0.18327	0.537	0.250(0)
Cu(1)	0.0	0.0	0.0	0.497	0.125(0)
Cu(2)	0.0	0.0	0.35421	0.379	0.250(0)
O(1)	0.0	0.0	0.15856	0.724	0.250(0)
O(2)	0.5	0.0	0.37971	0.592	0.238(2)
O(3)	0.0	0.5	0.37618	0.592	0.245(2)
O(4)	0.0	0.5	0.0	1.116	0.124(2)

The variation of normalized resistance of the samples with temperature is shown in figure 2. All the samples show metallic-to-superconducting transition around 90 K and there is not much variation in the transition temperature even though

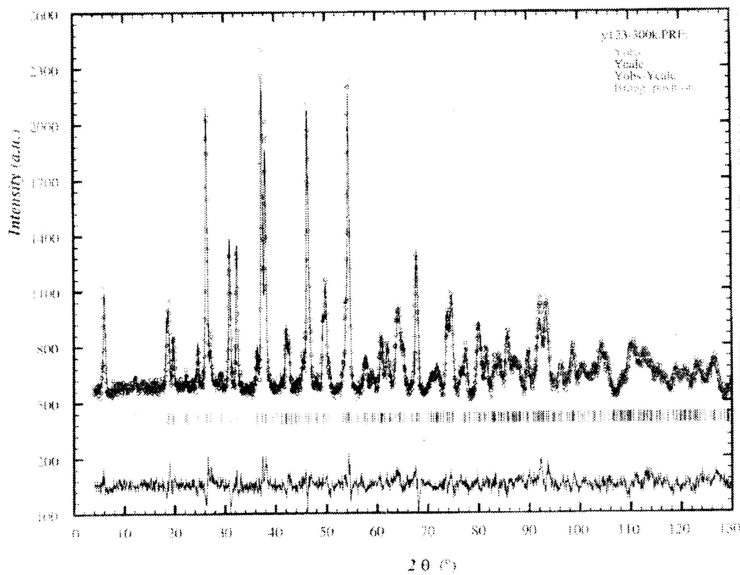

Figure 3. Rietveld refinement profiles for HgO added Y-123 ($x = 0.4$) sample. The dots are the observed diffraction data and solid curve is the calculated profile. The difference pattern for the sample is shown at the bottom.

HgO is added in different molar ratio. It is noteworthy that the samples were sintered in open atmosphere without any oxygen annealing. Still, the value of transition temperature (90 K) is in the close vicinity of the value reported for Y-123 compound annealed in O_2 ambient [3]. The most probable reason is the decomposition of HgO during sintering, which acts as an internal source of oxygen and improves the in-diffusion of oxygen in Y-123 system [17].

Rietveld refinement profiles for orthorhombic Y-123 with HgO addition is shown in figure 3. The essential results of the Rietveld refinement are summarized in table 1. The B values were used from ref. [19] and the refinement was done for oxygen. The neutron diffraction data which is extremely sensitive to oxide phases reveal single phase Y-123 nature. The oxygen content determined from the neutron diffraction data is 6.814 which is less than that of the reported oxygen annealed samples [1–4]. Further improvement of oxygen content may enhance the T_c value beyond 90 K.

4. Conclusion

We have synthesized Y-123 sample with HgO (0.4, 0.5, 0.6 and 0.7) addition using solid state reaction method in open atmosphere. The HgO addition in Y-123 leads to increase in oxygen content within the bulk material during sintering. As a result, Y-123 material exhibits superconductivity even though no oxygen annealing is done. The neutron diffraction and Rietveld refinement revealed that no HgO impurity is found in the sintered sample and oxygen occupancy in the sample is comparable to the sample annealed in oxygen atmosphere.

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