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Oxygen purity effect oxygen deficiency by argon heat treatment on Y:123 superconductors

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ABSTRACT

Argon heat treatments at 450 °C for different times of 0, 12, 24, 36, 48 and 60 h on the two Y:123 oxygenated samples with different oxygen purity (Hp 99.99% and Lp 93%) are reported. The results of X-ray diffraction, oxygen deficiency and resistivity measurements are presented in details. Furthermore, the superconducting critical temperature T_c, normal resistivity ρ_{300}, residual resistivity ρ_0, resistivity slope $d\rho/dT$ and width of transition ΔT_c are extracted from resistivity data. It is found that both c-parameter and oxygen deficiency increase by increasing annealing time, while OD and effective Cu valance decrease. But, the relative increase/decrease is more in Lp samples than that of Hp samples. Although T_c is decreased by annealing time for the two samples, the relative decrease in T_c is more in Lp samples when compared with those of Hp samples. Our results are discussed in terms of oxygen vacancies and hole carriers which are produced by annealing for the considered samples.

1. Introduction

The peculiar dependence of normal and superconducting properties on oxygen stoichiometry of Y:123 high T_c superconductor has been interesting land mark in the field of superconductivity [1–5]. It is well known that argon heat treatment at a temperature above 400 °C on oxygenated Y:123 superconducting system can lose oxygen from the poor Cu–O chains and consequently the oxygen content is decreased from seven to six along with a gradual decrease in T_c [6,7], while the rich Cu–O_2 planes are supposed to stay unaffected. The T_c versus oxygen deficiency plot exhibits two successive plateaus at 90 and 60 K (orthorhombic I and II). Orthorhombic I is well established and associated with an optimal doping of the CuO_2 planes [8], while orthorhombic II results from an alternate ordering of oxygen rich and oxygen poor of Cu–O chains along the *a*-axis [9].

However, Y:123 samples should have similar mechanism of oxygen transfer through Ar heat treatment, independent on oxygen purity during sintering processes. If the mechanism of oxygen loss stays independent of the degree of oxygen purity, the normal and superconducting properties should be at least qualitatively similar in both samples, as documented for Y:123 systems before annealing [10–12]. However, the present study shows that the oxygen purity results in an entirely different set of properties for the two samples after Ar heat treatments, which are highlights of the present work. To clarify the above, two similar oxygenated Y:123 samples are prepared by the well known solid state reaction method. But one of them is sintered in high purity oxygen (99.99%), while the other is sintered in low purity oxygen (93%). After that Ar heat treatment at a temperature of 450 °C for five different times (0–60 h) is performed.

2. Experimental details

Two similar samples of the series $YBa_2Cu_3O_7$ are synthesized by the well-known solid state reaction method. The ingredients Y_2O_3, $BaCO_3$ and CuO of 4 N purity are thoroughly mixed in required proportions and calcined at 900 °C in air for a period of 16 h, and then the furnace is slowly cooled to room temperature. This exercise is repeated three times with intermediate grinding at each stage. The resulting powders are ground, palletized in to two pellets. One of them is sintered in high purity oxygen (99.99% Hp) at 940 °C for a period of 24 h and then the furnace is cooled to room temperature with an intervening annealing for 24 h at 600 °C, while the other is sintered in low purity oxygen (93% Lp) at the same conditions. The 93% oxygen produced from air by the molecular sieve process contains not less than 90% and not more than 96% by volume of oxygen. The remainder consists mostly of argon and nitrogen. Also, some other gases could be

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found such as carbon dioxide (0.03%) and carbon monoxide (0.001%). The samples are tested for phase purity by X-ray diffraction (XRD) using Semen's D-500 with CuKα radiation of 1.541838 Å. The oxygen content is measured by idometery titration method. The electrical resistivity of the samples are obtained using the standard four-probe technique in closed cycle refrigerator [cryomech compressor package with cryostat Model 810-1812212, USA] within the range of (10–300) K. Nanovoltameter Keithley 2182, current source Keithley 6220 and temperature controller 9700 (0.001 K resolution) are used in this experiment. After that, Ar heat treatments, at a temperature of 450 °C for different times of 12, 24, 36, 48 and 60 h are separately made on the two considered samples with intermediate characterization and measurements.

3. Results and discussion

Fig. 1 shows XRD patterns for Hp samples before and after Ar heat treatment. The same is done for Lp samples. The structure of the samples, after annealing by Ar up to 60 h, maintain a clearly single phase and no additional peaks could be formed as before Ar treatment. The obtained peaks (0 1 0), (0 1 3), (1 0 3), (0 0 5), (1 1 3), (0 2 0), (2 0 0), (1 2 3) and (2 1 3) belong to the well known peaks reported for Y:123 superconductors. Although no extra peaks are observed in the XRD patterns for all samples, there is a little variation of the lines intensity of the peaks with increasing annealing time. The marginal shift in the position of lines is mainly attributed to the change in the *c*-lattice parameter produced by Ar annealing. The samples annealed at $t=0$ and 12 h are completely orthorhombic, being nearly evident from crystallographic splitting of (0 2 0), (2 0 0) and (1 2 3), (2 1 3). But with increase in annealing time up to 60 h, the crystallographic splitting of (0 2 0), (2 0 0) and (1 2 3), (2 1 3) disappears and seem to be one broader peak which indicates that the structure is changed to tetragonal. The c-parameters, orthorhombic distortion (OD) [(b − a)/b] and oxygen deficiency for all samples are listed in Table 1(a and b). It is clear that c-parameter and oxygen deficiency are increased by increasing annealing time for all samples, while OD continuously decreases. The c-parameter and oxygen deficiency for LpAr samples are comparatively more than observed in HpAr samples, while the vice is versa for OD. The change of c-parameter after annealing is probably related to the

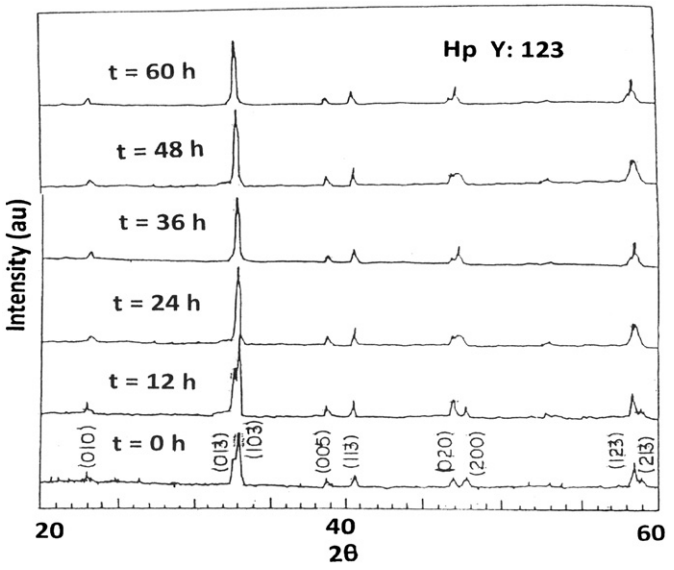

Fig. 1. XRD patterns of Hp Y:123 samples before and after annealing.

Table 1
(a) C- parameter, orthorhombic distortion, oxygen deficient and effective Cu valance, width of transition, resistivity slope , residual resistivity and normal resistivity of Hp samples

HP	C (Å)	Orth. Dis.	Oxy. def.	P	T_c (K)	ΔT_c (K)	ρ_0 (mΩ.cm)	ρ_{300} (mΩ.cm)	(dρ/dT) (mΩ.cm/K)
Ar0	11.683	0.019	0.09	2.273	90	8	0.09	1.07	0.003
Ar12	11.689	0.017	0.16	2.226	84	11	1.94	2.86	0.003
Ar24	11.695	0.012	0.20	2.200	72	18	6.38	4.98	−0.005
Ar36	11.702	0.009	0.28	2.147	60	25	7.67	6.13	−0.006
Ar48	11.713	0.001	0.51	1.993	40	40	12.04	7.33	−0.017
Ar60	11.717	0.000	0.64	1.907	00	85	18.84	8.92	−0.037

(b) C parameter, orthorhombic distortion, oxygen deficient and effective Cu valance, width of transition, resistivity slope, residual resistivity and normal resistivity of Hp samples.

LP	C (Å)	Orth. dis.	Oxy. def.	P	T_c (K)	ΔT_c (K)	ρ_0 (mΩ.cm)	ρ_{300} (mΩ.cm)	(dρ/dT) (mΩ.cm/K)
Ar0	11.683	0.018	0.08	2.280	89	6	0.329	1.47	0.004
Ar12	11.690	0.015	0.21	2.193	76	19	3.29	3.36	0.000
Ar24	11.699	0.008	0.32	2.120	52	33	10.35	6.76	−0.013
Ar36	11.705	0.005	0.39	2.073	22	63	19.29	8.19	−0.041
Ar48	11.726	0.000	0.66	1.893	00	00	25.09	9.55	−0.056
Ar60	11.731	0.000	0.75	1.833	00	00	38.94	10.16	−0.107