The effect of aging on the superconducting transition temperature and resistivity of Y-Ba-Cu-O ceramics after high temperature treatment

S.K.Nikoghosyan[1,2], V.V.Harutyunyan[2], V.S.Baghdasaryan[2], E.A.Mughnetsyan[1], E.G.Zargaryan[1] and A.G.Sarkisyan[1]
[1]International Scientific-Educational Center of NAS RA, Armenia,
[2]A.I. Alikhanyan National Science Laboratory (Yerevan Physics Institute) Foundation, Armenia

E-mail: nick@mail.yerphi.am

Absract. Some changes in superconducting transition temperature T_c and normal resistivity r of $YBa_2Cu_30_x$ ceramics are investigated as a result of the aging stimulated by high temperature treatment and slow cooling to room temperature followed by the storage under environmental conditions during 255 hours. It is shown that the changes of these parameters fall on three specific intervals of storage time t_a. In addition, different types of aging processes: fast and slow ones are observed for the heat-treated samples. It is found that the fast processes are accompanied by a slight increase of r and T_c within the first interval of storage time t_a ($t_a \leq 162$ hrs.), while the slow processes correlate with a sharp decrease of T_c and simultaneous increase of r within the second aging interval (162 hrs. $\leq t_a \leq$ 206 hrs.). It is revealed that the fast and slow aging processes result from the redistribution of atoms in intergranular weak and strong links, correspondingly. The probable reason of the observed results is the formation and coexistence of various structural phases including the semiconducing one.

1. Introduction

It is known that in the oxygen-deficient high-temperature superconducting (HTS) $YBa_2Cu_30_x$ samples obtained by quenching in liquid nitrogen after high temperature treatment and subsequent storage at room temperature, annealing of their characteristic parameters occurs [1-4]. Here, with increasing storage time, a significant increase in the temperature of superconducting (SC) transition T_c is observed due to processes of oxygen ordering in Cu-O chains of the sample unit cell [1,2]. This effect is known as "aging phenomenon", and a relationship between SC properties and structural parameters is revealed [1,2]. It is determined that the appearance of this phenomenon depends on the oxygen stoichiometry, as well as the sample treatment temperature and the conditions of its future storage (temperature, ambient atmosphere) [1,2,4]. Later, it was found that the phenomenon of aging takes place in HTS samples with intermediate shielding capacity, which in turn depends on temperature and the applied magnetic field [5]. Such a complicated dependence on various factors makes the results on aging effect of characteristic parameters (T_c and the corresponding values for resistivity r) of HTS rather ambiguous, and sometimes even controversial [1-4]. The above-mentioned data as well as the recent results of thermal annealing [6] show the importance of investigating the aging effects induced in heat-treated HTS materials not only after fast quenching, but also after slow cooling from 400°C to room temperature under environmental conditions and their subsequent aging in the same conditions. The paper is aimed at this problem.

2. Experimental methods

Two nearly identical rectangular $YBa_2Cu_30_x$ samples measured 2 mm×1 mm× 8 mm were used in our experiments. They were cut from the ceramic tablets synthesized in the air by the ordinary powder technology. Study of the aging phenomenon was carried out by registration of voltage-current characteristics using standard four- probe technique in the presence of the Earth's magnetic field [7]. Contacts on the samples were made using conductive silver paste. One of the two samples was heated in a tubular furnace at 400°C in the air for 30 minutes, cooled together with the furnace to room temperature at a rate of 2.5°C/min and then kept under environmental conditions during 255 hrs. (approximately 11 days). The other sample was kept in the same conditions and used as the control. During the whole storage time t_a, specific resistance r versus dc transport current was periodically recorded for both samples at 78 K. T_c was determined from the middle point of the sharp decrease on the r (I) curves within the temperature range from 77 to 300K. Note that the control sample parameters did not change during the whole storage period.

3. Results and discussion

Figure 1 presents a family of r (I) curves measured at 78 K after the storage of heat-treated samples during various t_a. Figure 2 shows r vs. t_a curves for fixed I measured at 78 K, and Figure 3-T_c and

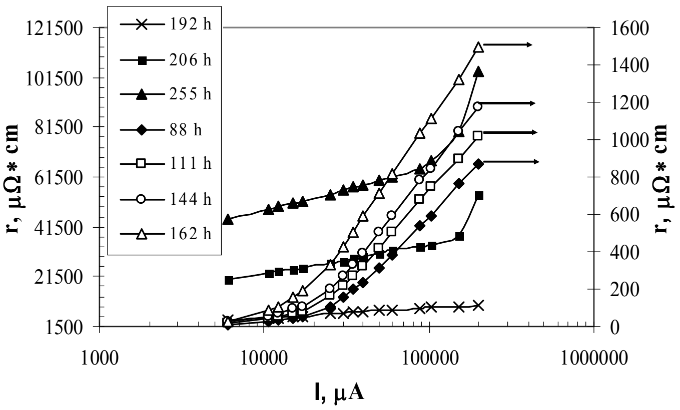

Figure 1. r (I) curves at 78 K for different storage times t_a.

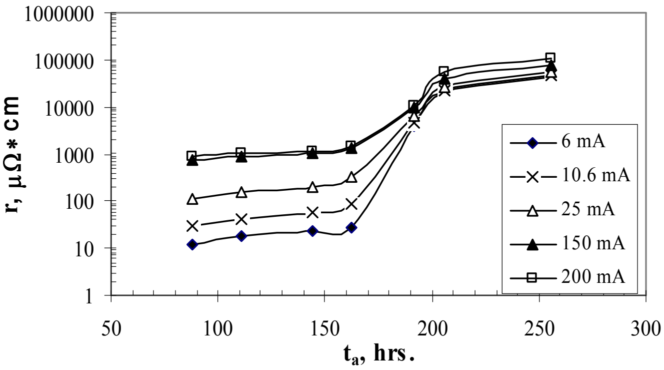

Figure 2. r (t_a) dependences of heat-treated samples for different transport currents I.

r(290K) dependences on t_a. For T_c and r (T, I) parameters, it is possible to distinguish three characteristic intervals of t_a. It is seen that for both measurement temperatures, r (t_a) curves for $t_a \leq 200$ hrs. are qualitatively similar. Besides, with increasing t_a, r in the first interval (88 hrs. $\leq t_a \leq$ 162 hrs.) grows much more slowly than in the second interval (162 hrs. $\leq t_a \leq$ 206 hrs.). Note that with increasing t_a, r (290K) in this interval increases reaching its maximum at t_a = 206 hrs. and then decreases with increase of t_a in the third interval (t_a > 206 hrs.). While in all intervals r(78K) increases with increasing t_a, in the third interval this increase occurs at much lower rates than in the second interval (see Figures 2 and 3). Moreover, at 78 K for the lower I and t_a the observed increase in r is smaller due to the fact that in this case the concentration of undestroyed weak links is small [7]. However, for the later aging stages (higher t_a) the concentration of destroyed weak links increases and, consequently, r of the sample increases. Note that in the first aging interval, when I changes within the range from 6 to 200 mA, the increase in r (78 K) makes 50 to 75 times, whereas for $t_a \geq 192$ hrs. the increase is only makes a factor of 2-3 (Figure 1). What this means is in the heat treated samples, rapid

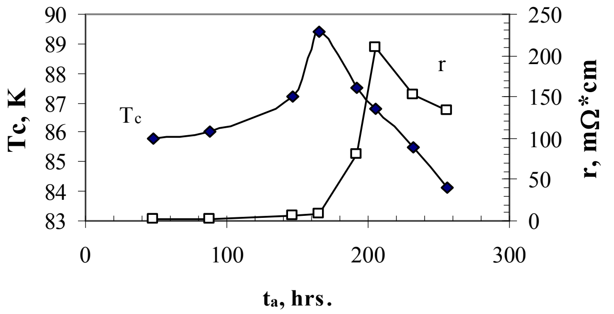

Figure 3. T_c and r (290K) dependences of heat-treated samples on the storage time t_a.

and slow aging processes are stimulated. For fast processes, the redistributions of oxygen atoms in weak linked copper-oxygen chains of the sample unit are responsible [1,2], and for slow processes - the redistributions of oxygen atoms in strongly bound copper-oxygen planes. Figure 3 also shows that t_a dependences of both T_c and r (290K) parameters have maxima at 162 hrs. and 205 hrs., correspondingly. Here, the increase in T_c with increasing t_a correlated with the simultaneous weak increase of r, and the sharp decrease in T_c after reaching its maximum is accompanied by rather rapid increase in r. The increase in T_c suggests that during the aging, first the (fast) ordering processes in the oxygen sublattice of the unit cell occur due to the redistribution of weakly bound oxygen atoms in the boundaries of superconducting grains, which is consistent with the result of [3]. However, if for yttrium based ceramics heat-treated at 78 K the observed temporary growth of T_c^i (responsible for intergranular weak links) correlates with the corresponding decrease in r [3], in our case it correlates, on the contrary, with the weak growth in r. It is seen that within the same interval of aging, at fixed t_a r(I) curves reveal a certain threshold I = I_s, below which with increasing I a relatively weak increase in r(78K) occurs, and for I> I_s the increase becomes significantly steeper (Figure 1). With increasing t_a, I_s (that creates the sample self-magnetic field), shifts to lower values due to the fact that the concentration of destroyed weak links increases, and the change of r(I) growth rate already occurs at lower I_s [4,7]. Qualitatively similar pattern is also observed for r(I) curves in the second aging interval (slow processes). However, the initial growth rate for slow processes is less than that for fast processes almost by an order of magnitude, and the corresponding I_s is more by a factor of 20. It speaks in favor of the assumption that in these aging processes, sharp increase in r with increasing I in I> I_s region is caused by the destruction of strong links in grains (or in superconducting planes). This behavior of r(I) (see Figure 1) is probably due to the depinning of self-magnetic field vortex lines from amorphous interface regions inside the grains and their movement into the grains [6]. Note that if with increasing t_a in the first interval I_s decreases from 30 to 10 mA, in the second interval I_s decreases from 150 to 100 mA. The probable reasons for the sharp increase of r in the first interval can also be the co-existence of two structural phases and changes in their volume fraction as well as in their degree of order during aging. This hypothesis is supported by our result on the availability of semiconducting phase and increase of their concentration (not included in this paper), and by the results of other investigations [6,8]. In particular, the observed increase in T_c can be explained by the transfer of O1 oxygen atoms from O5 position to their original sites, and the sharp growth of r in the second interval of t_a by the transition of atoms from O4 site into vacant O5 sites [2,8]. As for the formation of amorphous defect regions causing a sharp increase of r (I) for I> I_s in the third t_a interval, OH^- groups, which penetrate into the sample from ambient air moisture and then occupy O2 or O3 positions in Cu-O planes of the unit cell, could be responsible [6]. However, to get final clarification on the causes of the obtained results, further research is required. Such research is important in view of both the practical application and obtaining/expanding our understanding of the high-temperature superconductivity mechanism.

4. Conclusions

Study of the phenomenon of aging induced in $YBa_2Cu_30_x$ samples after heat treatment at 400°C and slow cooling to room temperature followed by the storage under environmental conditions, leads to the following conclusions. The changes of characteristic parameters during the progress of aging fall on three specific intervals of storage time t_a. In all intervals, r(78 K) increases with t_a. In the first and second intervals of storage time $t_{a,}$ the behaviors of r (78K) and r (290K) curves are qualitatively similar: their growth in the first interval with increasing t_a is much slower than in the second interval. However, in the second interval this increase for r(78 K) is more than two orders of magnitude faster than for r(290K). In the third interval, r(78 K) continues to increase with increasing t_a, but much more slower than in the second interval. While r(290K) decreases after reaching its maximum at t_a = 206 hrs., T_c also increases with increasing t_a in the first interval, passes through a maximum at t_a = 162 hrs. and decreases rapidly in the third interval. Moreover, in this range the increase of T_c correlates with weak increase in r(78 K) and r(290K). It is possible to assert that ordering processes are fast processes, and degradation processes are slow ones. Besides, the ordering is due to the redistribution of intergranular weak links, and slow processes are due to disordering in both weak and strong links (or inside the grains) of the HTS sample unit cell.

5. References

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