Turkish Journal of Physics
http://journals.tubitak.gov.tr/physics/
Research Article

Turk J Phys
(2013) 37: 237 – 241
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doi:10.3906/fiz-1203-16

The effect of neutron irradiation on the properties of $Tl_{0.6}Pb_{0.3}Cd_{0.1}Ba_2Ca_2Cu_3O_{9-\delta}$ superconductors

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Received: 28.03.2012 • **Accepted:** 20.01.2013 • **Published Online:** 19.06.2013 • **Printed:** 12.07.2013

Abstract: The effect of neutron irradiation on phase purity, crystal structure, and electrical resistivity of $Tl_{0.6}Pb_{0.3}Cd_{0.1}Ba_2Ca_2Cu_3O_{9-\delta}$ superconductors has been studied before and after irradiation by electrical resistance measurements and X-ray diffraction techniques. The zero-resistance superconducting transition temperature, $T_{c(offset)}$, of samples systematically decreases by about 5 K with the increase of neutron irradiation fluence up 18.12 × 10^{10} n cm^{-2}. The crystalline phases showed that the compounds have 2 phases, a high-Tc phase (1223) and low-Tc phase (1212), in addition to the impurity phase. The irradiation led to a decrease in oxygen content and volume fraction of high-Tc phase (1223).

Key words: Irradiation, superconductors, crystal structure, X-ray diffraction, resistivity, T_c

1. Introduction

Superconductivity has been discovered in the Tl-Ba-Ca-Cu-O system with a variety of transition temperatures and compositions [1–4]. One of the problems in studying the superconducting behavior of these materials is the difficulty of obtaining single-phase samples. Partial replacement of Tl^{+2} in an oxygen-deficient Tl-Oδ layer by cations having higher oxidation states than Pb^{+4}, Bi^{+3}, and Sb^{+4} in order to improve the stability and make the synthesis of single-phase material more readily feasible has also been investigated [4-6]. Irradiation techniques are a powerful tool for assessing the influence of defects on superconductors, because they allow one to investigate the same sample prior to and after the irradiation, which excludes the problems of the variations of the sample. In particular, neutron irradiation was used in extended studies of the influence of disorder in superconducting material. The neutron-induced defects are also highly suitable for investigating flux pinning. This was done successfully in the cuprates [7–10]. Therefore, it should be fruitful to investigate the effect of irradiation on superconductors. In the present work, we have carried out irradiation experiments on a Tl(Cd)-1223 superconductor at room temperature by using neutron irradiation fluences of about 6.04 × 10^{10}, 12.08 × 10^{10}, and 18.12 × 10^{10} n cm^{-2}, irradiated for 1, 2, and 3 weeks, respectively, in order to study some properties of the proposed superconductors, such as $T_{c(offset)}$, $T_{c(onset)}$, oxygen content, lattice parameters, and volume fraction.

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2. Experimental

The synthesis of 4 samples (A, B, C, and D) of $Tl_{0.6}Pb_{0.3}Cd_{0.1}Ba_2Ca_2Cu_3O_{9-\delta}$ high-temperature superconductor phases were prepared by the solid-state reaction method. The samples corresponding to the original mixtures were prepared by the same initial chemical proportions by the mixing of Tl_2O_3, PbO, CdO, BaO, CaO, and CuO as a starting materials, according to the following general chemical formula:

$$0.3Tl_2O_3+0.3PbO+0.1CdO+2BaO+2CaO+3CuO \rightarrow Tl_{0.6}Pb_{0.3}Cd_{0.1}Ba_2Ca_2Cu_3O_{9-\delta}$$

The synthesis of the samples was carried out by a 2-step precursor method. In the first step, the powders BaO, CaO, and CuO were mixed together by using agate mortar to form slurry during the process of grinding for about 30–50 min. The mixture was put in a furnace for calcinations; for this process, the powder was heated to a temperature of 800 °C for 3 h with a rate of 200 °C h^{-1}. After that, it was cooled to room temperature at the same rate. In the second step, the $Ba_2Ca_2Cu_3O_7$ precursor was mixed with Tl_2O_3, PbO, and CdO to obtain the nominal composition $Tl_{0.6}Pb_{0.3}Cd_{0.1}Ba_2Ca_2Cu_3O_{9-\delta}$. The powder was pressed into disk-shaped pellets 1.2 cm in diameter and 0.2–0.3 cm in thickness by using a hydraulic press under a pressure of 7 t cm^{-2}. The prepared pellets were put in the usual quartz tube and presintered in air at 855–860 °C for 4 h with a rate of 200 °C h^{-1}, and then they were cooled to room temperature at the same rate. After that, the pellets were reground, repressed, and resintered in oxygen (oxygen rate: 0.2 L min^{-1}) at the same range of temperature for a further 12 h and were then cooled to 500 °C, annealed in oxygen for 2 h, and cooled to room temperature at the same rate. In order to make comparisons among samples A, B, C, and D (sample A was prepared without irradiation; samples B, C, and D were irradiated by using neutron fluences of about 6.04×10^{10}, 12.08×10^{10}, and 18.12×10^{10} n cm^{-2}, respectively). The neutron fluences were obtained from an available neutron source (^{241}Am-^{9}Be) with the assumption that the irradiation through the samples was uniform. The ρ–T (resistivity vs. temperature) characteristics of these samples were measured by means of a standard DC 4-probe technique in order to investigate their superconducting properties. The oxygen content could be determined by using a chemical method, the so-called iodometric titration [11]. Methods of measuring the critical temperatures as well as the volume fraction for any phase (V_{phase}) have been described elsewhere [6] and will not be mentioned here. The structures of the prepared samples were obtained by using X-ray diffraction (XRD) measurements in a $\theta°$-$2\theta°$ arrangement, from 20° to 60°. A computer program was established to calculate the lattice parameters a and c, and this program was based on Cohen's least square method [12].

3. Results and discussion

From the plot of the normalized resistivity vs. temperature (ρ–T) curves shown in Figure 1, one can obtain the values of critical transition temperatures $T_{c(offset)}$ and $T_{c(onset)}$ before and after neutron irradiations with fluences of 6.04×10^{10}, 12.08×10^{10}, and 18.12×10^{10} n cm^{-2}. These temperatures were found to be 116, 113, 112, and 111 K for $T_{c(offset)}$ and 136, 133, 132, and 130 K for $T_{c(onset)}$, respectively. The observed behavior may be attributed to the change of oxygen stoichiometry, which controls the hole concentrations in conducting CuO planes. Thus, irradiation induced changes in oxygen content as expected, which might bring changes in the carrier concentration, resulting in the observed values of Tc. The neutron irradiation might induce damage of the weak links [13] between grains and consequently it renders them largely unconnected after irradiation, as indicated by Sauerzopf and Wiesinger [14]. The values of oxygen content are shown in the Table, where it can be noticed that the values of oxygen content of the specimens after irradiation are decreased because irradiation will probably damage the bonds in the CuO planes and create defects. These defects decrease the number of holes in the lattice, which will decrease the critical transition temperature T_c [10]. According to the XRD data, the samples were found to consist of an almost phase-pure polycrystalline Tl(Pb,Cd)-1223 phase. However, very small amounts of Tl-1212 were also detected in all samples. Figure 2 shows XRD patterns taken in pelts

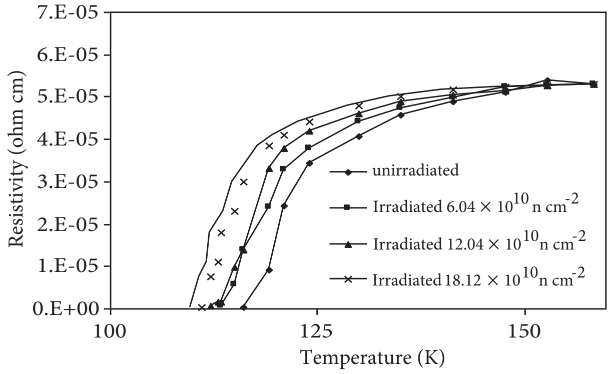

Figure 1. Temperature dependence of resistivity for $Tl_{0.6}Pb_{0.3}Cd_{0.1}Ba_2Ca_2Cu_3O_{9-\delta}$ samples.

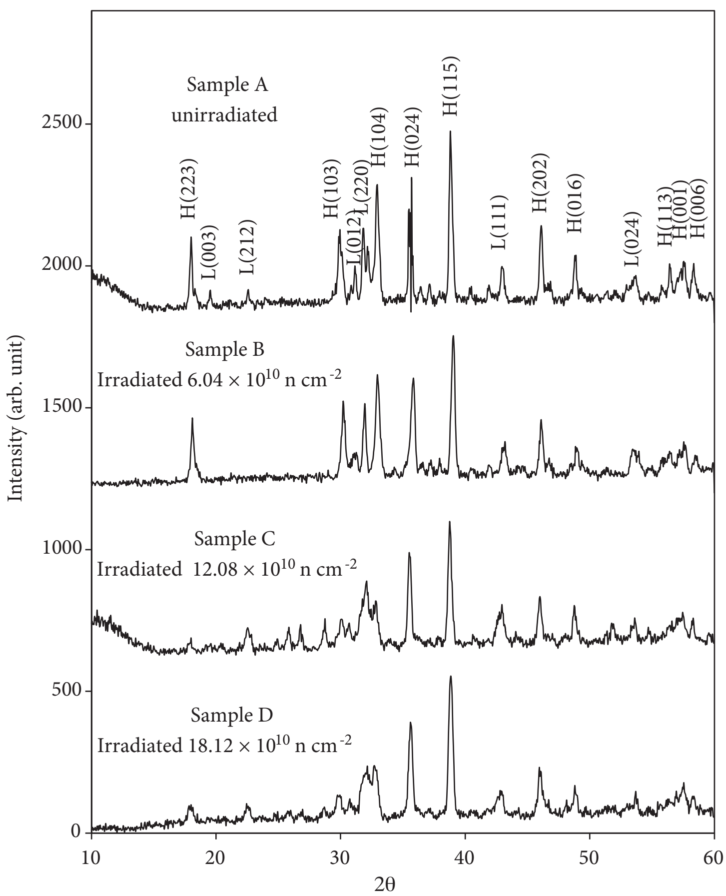

Figure 2. XRD patterns for the $Tl_{0.6}Pb_{0.3}Cd_{0.1}Ba_2Ca_2Cu_3O_{9-\delta}$ samples.

without irradiation for sample A (unirradiated) and samples B, C, and D irradiated with neutron irradiation fluences of 6.04 × 10^{10}, 12.08 × 10^{10}, and 18.12 × 10^{10} n cm^{-2}, respectively. From Figure 2, it can be observed that all the samples consisted of a major 1223 high-T_c phase (H-peaks), minor low T_c phase 1212 (L-peaks), and a very small amount of secondary phases. It can also be noticed from Figure 2 that as the irradiation fluences increase, the L-peaks increase, while the H-peaks decrease; the high-T_c phase reflections of sample A have higher intensity than samples B, C, and D.

Table. Values of transition temperatures $T_{c(offset)}$ and $T_{c(onset)}$, oxygen content, oxygen excess $\delta(O_2)$, lattice parameters a and c, c/a, and volume fraction $V_{ph(1223)}$ for the $Tl_{0.6}Pb_{0.3}Cd_{0.1}Ba_2Ca_2Cu_3O_{9-\delta}$ samples at different neutron irradiation fluences.

Neutron irradiation fluence (n cm^{-2})	$T_{c(offset)}$ (K)	$T_{c(onset)}$ (K)	O_2 content	$\delta(O_2)$	a (Å)	c (Å)	c/a	$V_{ph(1223)}$
0.0	116	136	8.78	0.22	3.847	15.89	4.130	81%
6.04 × 10^{10}	113	133	8.75	0.25	3.846	15.88	4.129	78%
12.08 × 10^{10}	112	132	8.73	0.27	3.846	15.81	4.111	77%
18.12 × 10^{10}	111	130	8.71	0.29	3.846	15.81	4.111	76%

The lattice parameters were estimated by using d-values and hkl reflections of the observed XRD pattern through a software program based on Cohen's least square method. Values of transition temperature parameters $T_{c(offset)}$ and $T_{c(onset)}$, oxygen content, excess oxygen $\delta(O_2)$, lattice parameters a and c, c/a, and volume fraction $V_{ph(1223)}$ are tabulated in the Table. The experimental errors in the present results were estimated to be about ±5%.

From Figures 3–5, it can be observed that there is, in general, a decrease in volume fraction, ratio of lattice parameters c/a, and oxygen content by increasing the neutron irradiation fluence.

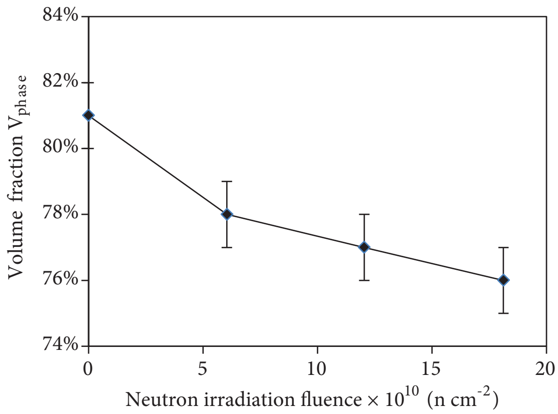

Figure 3. Volume fraction $V_{(phase)}$ as a function of neutron irradiation fluence for $Tl_{0.6}Pb_{0.3}Cd_{0.1}Ba_2Ca_2Cu_3O_{9-\delta}$ samples.

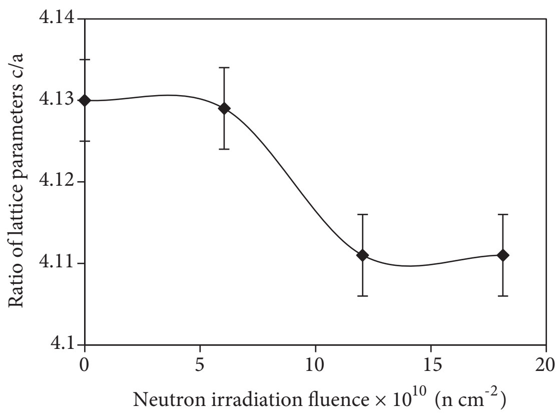

Figure 4. Ratio of lattice parameters c/a as a function of neutron irradiation fluence for $Tl_{0.6}Pb_{0.3}Cd_{0.1}Ba_2Ca_2Cu_3O_{9-\delta}$ samples.

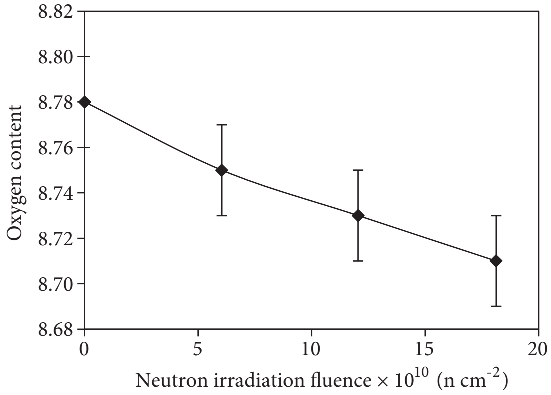

Figure 5. Oxygen content as a function of neutron irradiation fluence for $Tl_{0.6}Pb_{0.3}Cd_{0.1}Ba_2Ca_2Cu_3O_{9-\delta}$ samples.

4. Conclusions

In the present study, we have investigated the effect of neutron irradiation on the physical properties of $Tl_{0.6}Pb_{0.3}Cd_{0.1}Ba_2Ca_2Cu_3O_{9-\delta}$ superconductors, synthesized by the usual solid-state reaction method. The transition temperature of as-grown samples was found to be sensitive to the irradiation; it decreased from 116 K to 111 K after neutron irradiation fluence of 18.12×10^{10} n cm^{-2}. The variation of T_c might be explained by the mobility of smaller defects and their interaction with the radiation-induced defect cascades [15]. Volume fraction, ratio of lattice parameter c/a, and oxygen content were found to decrease as the neutron irradiation fluence was increased. The observed decrease in the volume fraction might be due to displacement of oxygen atoms in the CuO chains, which is confirmed from the observation of the c/a ratio decrement. It should be mentioned that the influence of neutron irradiation might also create effective pinning centers, and so it would be desirable to study evolution of the flux pinning mechanism with neutron irradiation.

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