Study the Effect of Gamma Radiation on Superconducting Properties of $T_{0.5}Pb_{0.5}Ba_2Ca_2Cu_3O_{9-\delta}$ Compound

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Abstract. Two samples of the superconducting compound $Tl_{0.5}Pb_{0.5}Ba_2Ca_2Cu_3O_{9-\delta}$ were prepared by solid-state reactions. One sample was exposed to a Cesium-137 radiation source at a dose of 200 MR to study the electrical and structural properties of the superconducting transition temperature, while the other was left unexposed. The effect of gamma radiation on the electrical (transition temperature and resistivity) and structural properties was studied. The results showed that the normal-state resistivity increased with increasing gamma dose, while the transition temperature decreased with the same gamma dose. The structural properties were studied by using X-ray diffraction (XRD), and the results showed no significant change in the diffraction angles or peak widths. The intensity of the XRD peaks of the sample decreased with increasing gamma dose. No significant lattice expansion occurred, although the $Tl_{0.5}Pb_{0.5}Ba_2Ca_2Cu_3O_{9-\delta}$ superconductor was strongly affected by gamma radiation, as both samples were found to have a tetragonal crystal structure.

Keywords: Gamma radiation, Superconductor, Critical transition, Cesium-137.

INTRODUCTION

High-temperature superconductors (HTS) have attracted increasing interest for industrial purposes and research, due to their advantages over conventional low-temperature superconductors that require liquid Helium [1-3]. Basic thallium is one of the most interesting HTS superconductors. Two distinct superconductor phases exist in TlSr-Ca-Cu-O superconducting system, with several CuO layers in the Crystal structure [4-6]. The $TlSr_2Ca_2Cu_3O_{9-}$ (phase 1223) system has a zero-resistance transition temperature (T_c) of 105–118 K (depending on the microstructure) and contains three CuO layers. Similarly, the $TlSr_2Ca_1Cu_2O$ (Tl-1212 phase) system contains two CuO layers, and the copper oxide layer exhibits superconducting transition temperatures (Tc) ranging from 70–105 K. Since the discovery of superconductivity in $TlSr_2Ca_2Cu_3O_9$ ceramics, the preparation of single-phase 1223 materials have proven challenging because the 1212 phase is thermodynamically more favorable at high temperatures than the 1223 phase. Various preparation methods, as well as structural and superconductive properties, have been widely reported [7, 8]. It is worth noting that the Tl-1223 phase has been used in many techniques to process bulk ceramic superconductors, including co-deposition [9], sol-gel process [10], freeze-drying [11], and solid-state reaction [12]. $TlBr_2Ca_2Cu_3O_{9-\delta}$ (Tl-1223) powders were produced using the solid-state reaction method because it is a simple, efficient, and low-cost method. Adding lead to the thallium phase (Tl-1223) promotes phase diffusion, stabilizes the Tl-1223 phase, forms nucleation centers, and accelerates reaction kinetics. Lead (Pb) significantly influences phase composition, microstructure, and superconductivity. The presence of lead in the initial mixture results in the formation of the Tl

(Pb)-1223 phase. However, it is almost impossible to obtain a pure phase because it usually coexists with the Tl (Pb)-1212 phase. Lead also improves grain contact and grain growth, resulting in improved transfer current density (Jc), especially in low fields with 5% weight addition of lead. [12]. The cohesive force of the lattice atoms within the grains results in an improved critical current density (Jc) [13-15]. The high critical temperature of superconductors plays an important role in specific applications, such as particle accelerators, magnetic resonance imaging, and energy-saving technologies. [16, 17]. In this paper, we present a study of the effect of gamma radiation on structural and electrical properties, such as the critical temperature (T_c) and superconducting transition of a polycrystalline sample of $Tl_{0.5}Pb_{0.5}Ba_2Ca_2Cu_3O_{9-\delta}$. Irradiation was carried out using a Cesium-137 radiation source in air at room temperature, with a dose of 200 MR.

EXPERIMENTAL

Use appropriate weights of pure powders of Tl_2O_3, PbO, BaO, CaO, and CuO as a basic material based on their concentrations in the superconducting compound 1223 with the chemical formula $Tl_{0.5}Pb_{0.5}Ba_2Ca_2Cu_3O_{9-\delta}$. A solid-state reaction technique was employed to synthesize two samples of compounds A and B with identical elemental compositions. The powders were weighed using a sensitive balance to determine the weight of each reactant. Using an agate mortar, the powders were combined and ground for 30 to 50 minutes. The mixture was placed in a quartz glass tube and then evacuated using a vacuum device to ensure that the sample components did not react with air gases. (10^{-3}Toor) Centrifugation—a heat treatment—was carried out in a furnace equipped with a programmable controller. The powder was heated for three hours at 800 °C. The sample components were then removed and ground again. The mixture was divided into two parts and pressed into 1.2 cm diameter pellets under a hydrostatic pressure of approximately 8 tons/cm². The pellets were placed in a programmable sintering furnace and heated at 855-860°C for 24 h at 100°C/h, then cooled to room temperature at the same heating rate. Sample (A) was prepared without radiation, while Sample (B) was exposed to gamma radiation at room temperature using ^{137}Cs, up to a dose of ≈ 200 MR. The ρ-T (resistance versus temperature). Using a standard four-probe DC technique, the properties of these samples were measured to verify their superconducting state. The critical temperatures determined to obtain the structure of the prepared samples, X-ray diffraction (XRD) measurements were used within a range of 20–60°. The lattice parameters (a) and (c) were determined using a computer program that applied Cohen's least squares approach [18].

RESULTS AND DISCUSSION

A four-probe superconductivity measuring system was employed to determine and compute the critical transition temperature (Tc) of the prepared samples. Each specimen was mounted individually in the cryogenic chamber of the device and gradually cooled. During the cooling process, the current passing through the samples and the corresponding voltage drop across the terminals of sample A (without irradiation) and sample B (after irradiation) were continuously recorded as functions of temperature. From these measurements, the electrical resistivity was calculated at each temperature interval, allowing the construction of the full resistivity–temperature profile.

Figure 1 presents the normalized resistance versus temperature (ρ–T) behavior of the $Tl_{0.5}Pb_{0.5}Ba_2Ca_2Cu_3O_{9-\delta}$compound before and after γ-ray irradiation [19, 20]. As shown in the figure, both samples A and B exhibit a metallic-like trend in the normal state region: the resistance decreases progressively as the temperature decreases, which is consistent with previously reported results for high-Tc cuprate superconductors [21].

The superconducting transition temperatures of both samples were extracted from the ρ–T curves. A distinct reduction in Tc was observed upon γ-irradiation. From Figure 1, the displacement transition temperature Tc(displacement) decreases from approximately 117 K to 112 K, while the initial onset transition temperature Tc(initial) decreases from about 133 K to 129 K as the radiation dose increases. This systematic decrease in Tc indicates a clear deterioration in the superconducting properties caused by irradiation.

The reduction in the critical transition temperature can be attributed to increased disorder within the Cu–O planes, reduced atomic rearrangement, and modifications in the oxygen stoichiometry of the irradiated samples. These effects are consistent with changes in excess oxygen δ values presented in Table 1, which demonstrate that γ-ray exposure alters the oxygen content and consequently affects the electronic structure responsible for superconductivity [22, 23]. Overall, the observed behavior confirms that γ-irradiation introduces structural and electronic defects that weaken the superconducting phase of the Tl-based cuprate system.

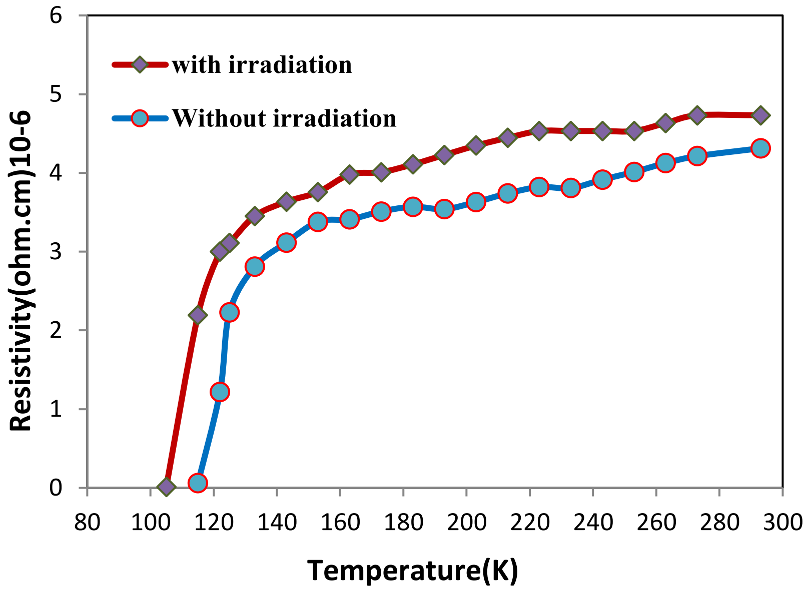

FIGURE 1. The relationship between temperature and resistance, (A) without irradiation and (B) with gamma irradiation.

The XRD patterns of samples (A) and (B) are presented in **Figure 2**. The diffraction results confirm that both samples contain a dominant high-Tc phase (H-1223), a weaker low-Tc phase (1212-L), and a very small amount of secondary phases. Notably, the overall peak intensity of sample (B) — the γ-irradiated specimen — decreases compared to the unirradiated sample (A). This reduction in intensity indicates structural degradation and reduced crystallinity as a consequence of radiation exposure.

A decrease in mass density (ρm) and in the c/a lattice parameter ratio was also observed for sample (B), as documented in **Table 1**. These variations arise from radiation-induced dissolution of Cu–O bonds within the CuO2 planes and the formation of additional structural defects, both of which contribute to a reduction in carrier concentration (hole density) within the lattice [24]. The breakdown of CuO bonding and the accumulation of lattice defects—particularly oxygen vacancies and Cu-O site disruptions—lead to a measurable decrease in charge carriers, thereby lowering the material's overall electrical conductivity. This mechanism is well known in cuprate superconductors and is a major pathway through which structural imperfections influence electronic transport properties [25–27].

Historically, more than 100 distinct copper-oxide phases exhibiting high-temperature superconductivity have been identified. These structures are generally composed of alternating layers of oxygen-deficient alkaline-earth metals (such as Ba and Ca) and vacancy-free CuO2 planes, meaning that most cuprate phases originate from ordered perovskite-type frameworks containing controlled oxygen vacancies [28–30]. Because superconductivity in cuprates is highly sensitive to oxygen stoichiometry, any change in the number of holes within the CuO2 planes directly influences the superconducting transition temperature. Thus, the reduction in hole concentration caused by irradiation is an essential factor behind the observed decrease in Tc.

Based on the XRD results in **Figure 2**, both samples exhibit a tetragonal crystal structure; however, subtle changes in the c/a ratio indicate lattice distortion in the irradiated sample. This distortion is accompanied by a noticeable reduction in the fraction of the high-Tc H-1223 phase. Since the superconducting properties of Tl-based cuprates depend strongly on the proportion of the high-phase component, the diminished H-1223 content is the primary reason for the reduced transition temperature.

Overall, the findings suggest that γ-ray irradiation effectively promotes phase decomposition and delamination within the CaO2 layers, leading to partial transformation of the high-Tc phase into lower-Tc phases. This phase degradation is consistent with the reduced Tc observed in the irradiated sample and demonstrates that γ-irradiation is a highly effective mechanism for inducing high-phase deconvolution and suppressing superconductivity in Tl-based cuprate systems [31].

TABLE 1. Oxygen content (δ), Values of lattice parameter, T_c, c/a, and ρ_M for the samples.

Sample	$T_{c\,(OFF)}$(K)	$T_{c\,(ON)}$ (K)	$\delta(o_2)$	a(A^0)	c (A^0)	c/a	r_m
A	117	133	0.213	3.421	13.280	3.881	4.92
B	112	129	0.254	3411	13.195	3.868	4.86

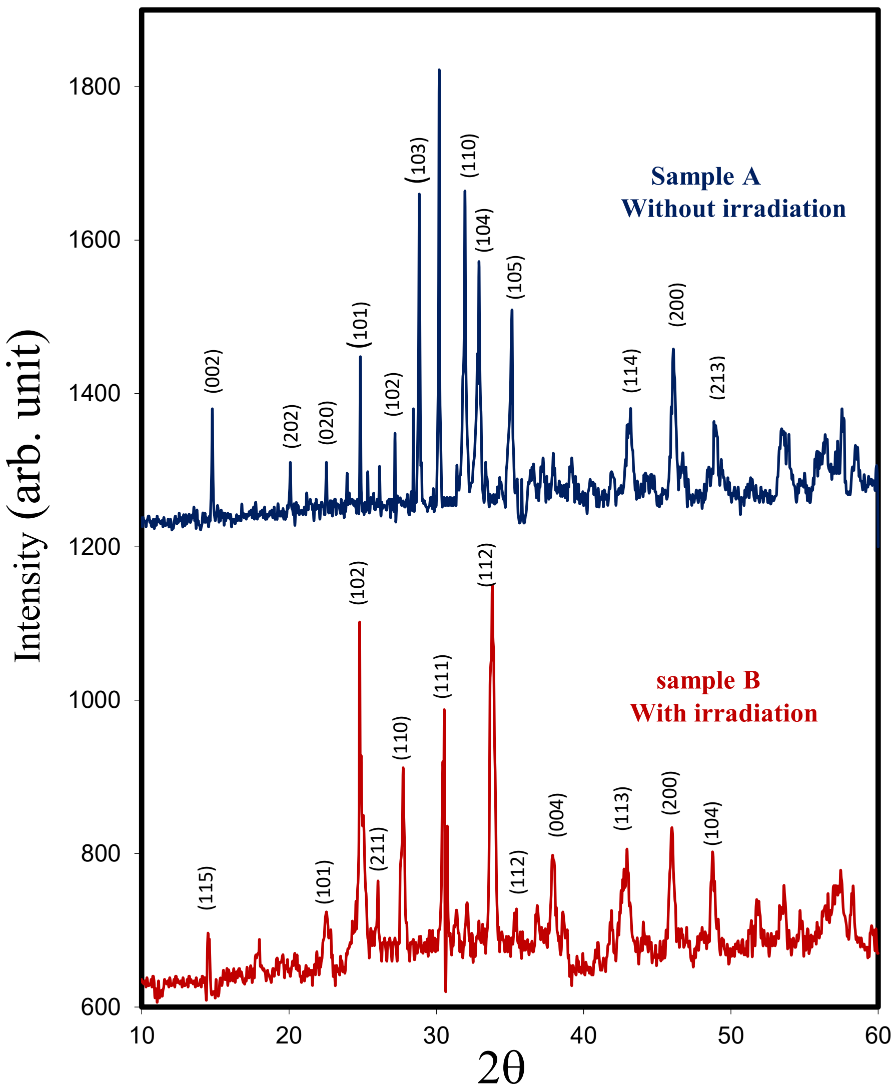

FIGURE 2. XRD patterns for samples (a) before and (b) after irradiation

CONCLUSION

In this manuscript, two samples of the superconducting compound $Tl_{0.5}Pb_{0.5}Ba_2Ca_2Cu_3O_{9-\delta}$ were prepared under ideal conditions by solid-state reaction. One sample was exposed to gamma radiation, while the other remained unaffected. Compared to the unexposed sample, XRD analysis revealed a rectangular structure with a low lattice constant (c). The transition temperature of the as-grown samples was found to be sensitive to gamma rays; a decrease in the temperatures (Tc_{off}) and (Tc_{on}) from 117 to 112 K and from 133 to 129 K was observed. XRD revealed a tetragonal crystal structure in our samples, with a variable c/a ratio.

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