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Effects of γ-Irradiation in $YBa_2Cu_3O_7$ Thick Films and $Tl_2Ba_2Ca_2Cu_3O_{10}$ Tapes

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Abstract

We have investigated the effect different doses of γ-rays on the behavior of the critical current density, J_c in $YBa_2Cu_3O_7$ thick films and $Tl_2Ba_2Ca_2Cu_3O_{10}$ tapes at high temperatures. All samples were irradiated at room temperature by a ^{60}Co γ-ray source at a dose rate of *0.5 MR/h*. In the $YBa_2Cu_3O_7$ thick films, J_c was found to be enhanced almost linearly with increasing γ-irradiation dose up to *50 MR*. In the $Tl_2Ba_2Ca_2Cu_3O_{10}$ tapes, small γ-irradiation doses were found to significantly enhance J_c at most temperature values. As the dose was increased J_c was found to have a slight further enhancement and nearly reaches saturation at the highest doses applied. At the highest temperature values used, γ-irradiation was found to have insignificant effect on J_c in the $Tl_2Ba_2Ca_2Cu_3O_{10}$ tapes. These results are discussed and explained in terms of the roles of several mechanisms created by γ-rays in the regions of the grain boundaries.

Keywords: Superconductors, grain boundaries, dose, γ-rays, critical current.

Introduction

Optimizing the critical current density J_c, in high-temperature superconductors (HTSCs) is very necessary for their electric power applications. When a transport current density, $\boldsymbol{J}$ is applied to a HTSC in the mixed state it will produce a Lorentz force, $\boldsymbol{F}_L$ on the vortices trying to move them. The Lorentz force is given by; $\boldsymbol{F}_L = (1/c)\, \boldsymbol{J} \times \boldsymbol{\Phi}_o$ where $\boldsymbol{\Phi}_o$ is the quantized magnetic flux per vortex. If the Lorentz force is not opposed by pinning forces, an electric field, $\boldsymbol{E} = (-1/c)\, \boldsymbol{B} \times \boldsymbol{v}$ will be induced in the sample causing energy to be dissipated, were **v** is the velocity of vortices transverse direction to both $\boldsymbol{J}$ and $\boldsymbol{B}$, and $\boldsymbol{B}$ is the applied magnetic field. Unfortunately, the pinning of vortices in HTSCs is fairly weak, especially at high temperatures [1]. The vortices can be effectively pinned by any structural inhomogeneities of the material. These structural inhomogeneities arise from impurities, grain boundaries, voids, etc. Defects created in the HTSCs using several types of energetic radiations were found to be very successful in increasing the vortex pinning forces [2-8]. Understanding the influence of crystal-lattice disorder on the superconducting properties of HTSCs is important from technological and fundamental points of view.

In polycrystalline HTSC materials, two types of disorder can appear; microscopic and macroscopic. The microscopic disorder such as impurities, vacancies is associated with perturbation of crystal lattice on the atomic scale. The macroscopic disorder is associated with structural inhomogeneities such as grain boundaries. The polycrystalline HTSCs are generally composed of superconducting grains separated by highly disordered non-superconducting or even dielectric grain boundaries [9]. Because the regions of grain boundary are highly disordered, the influence of both mechanisms in them is very significant compared with the less disordered grains. Superconducting coherence can be established between the grains by Josephson coupling leading to enhancement of the transport properties. Also the regions of the grain boundaries are effective vortex pinning forces, preventing vortex motion perpendicular to the grains orientation [10] resulting in larger J_c values. At the same time the grain boundaries may act as weak links, thus impeding the current flow between the superconducting grains resulting in lower J_c values. Improving the grain connectivity will result in higher J_c values. Because the regions of grain boundaries and nearby regions are depleted from charge carriers, they are expected to be strongly sensitive to γ-rays.

In the present paper, we report on the influence of γ-irradiation doses on the behavior of the critical current density in $YBa_2Cu_3O_7$ thick films and $Tl_2Ba_2Ca_2Cu_3O_{10}$ tapes. The results are discussed and explained by the possible mechanisms of defects created by γ-irradiation in the regions of the grain boundaries.

Experimental Procedure

The $YBa_2Cu_3O_7$ thick films were prepared by solid-state reaction technique from stoichiometrically mixed high purity (*99.99*) powders of $BaCO_3$, Y_2O_3, and CuO. The powders were thoroughly mixed and ground in agate mortar for *2 h*. The powders were then filled in a ceramic crucible, pressed into loose compacts and then calcinated in air at $900\ ^oC$ for *10 h*. After slow cooling to room temperature, the compacts were crushed in the mortar after which they were ball milled in acetone for *2 h*. Then they were dried pressed into *1 cm* diameter pellets at a pressure of *5 MPa* and fired to $950\ ^oC$ in air for *10 h*. The sample was then cooled down at a rate of *5 K/min* to $450\ ^oC$ at which it was sintered and annealed in flowing oxygen for *5 h*. The sample was finally cooled down to room temperature. Thick films of YBCO were prepared using the screen printing technique [11]. Fine grained precursor powder of YBCO was thoroughly mixed with an organic binder “ethylene glycol (HOCH2CH3OH)” to form a paste of appropriate viscosity. The prepared YBCO paste was then screen printed on a polished MgO single crystal (*100*) substrate with dimensions (*10 mm ×10 mm ×1mm*) using a *200* mesh fine nylon screen. The substrate was masked using a nylon masking tape. The screen-printed film was first dried at *200 °C* for *2 h* to remove the organic binder. The dried film was then heat treated in air at *950 °C* for *10 h*, cooled at a rate of *5 K/min*, followed by oxygen flow at *450 °C* for *5 h* and cooled down to room temperature.

In preparing the $Tl_2Ba_2Ca_2Cu_3O_{10}$ tape appropriate amounts of high purity Tl_2O_3, BaO_2, CaO and CuO were thoroughly mixed and ground for *2 h*. The mixed powder was then rapped in a gold foil and subjected to a three-step heat treatment, each of *5 h* at *890 ^oC* in air with intermediate grinding and pressing between steps. The resulting powder was pressed into pellets and annealed at *910 ^oC* in flowing oxygen. The powder was then packed into a silver tube with *4 mm* outer and *2.4 mm* inner diameters. The tube was then rolled to produce a *0.2 mm* thick and *4 mm* wide tape. The tape was then annealed at *850 ^oC* in air for *5 h* followed by a uniaxial pressing at *500 MPa*. The annealing cycle was repeated to produce a nearly single phase and textured $Tl_2Ba_2Ca_2Cu_3O_{10}$ tape.

The voltage-current (*V-I)* characteristics were measured using the standard *dc* four-probe method. The probes were fixed to the samples with silver paste. The sample was cooled using a closed cycle helium cryostat (Leybold *320*) under a vacuum of *10^{-2} mbar*. The voltage was measured using a sensitive nano-voltmeter (Keithley *2182*) with accuracy better than *10 nV*. The current was measured using a programmable constant current source (Keithley *2245*) with accuracy of *1 μA*. The current density used for electrical resistance measurement was *1 mA/cm^2*. The temperature was measured with a calibrated platinum resistance sensor with an accuracy of ± *0.1 K*. The critical current densities were determined using an electric field criterion of *1 μV/cm*. In all measurements, the sample was first zero-field cooled down to *77 K* and then the *V-I* characteristics were measured.

Results and Discussion

The *V-I* characteristic curves were obtained at all temperatures at zero applied magnetic field before and after irradiation. Figure 1 shows the *V-I* characteristic curves for the $YBa_2Cu_3O_7$ thick films at *77 K* and for several doses of γ-rays. From these *V-I* curves we obtained the critical current density at each applied field and temperature using an electric field criterion of *1 μV/cm*. Similar *V-I* curves were obtained for the $Tl_2Ba_2Ca_2Cu_3O_{10}$ tapes but not presented here. Figure 2 displays the behavior of J_c in the $YBa_2Cu_3O_7$ thick films at *77 K* as a function of γ-irradiation dose at no applied fields. It is clear that J_c increases almost linearly with the irradiation dose up to the maximum dose used (*50 MR*). Figure 3 shows J_c in the $Tl_2Ba_2Ca_2Cu_3O_{10}$ tapes as a function of γ-irradiation dose at several temperatures. We can see that J_c values were enhanced after irradiation dose of *100 MR* followed by another small enhancement after irradiation dose of *200 MR*. After irradiation dose of *400 MR*, no significant further enhancement in J_c was observed. We can also see that the enhancement in J_c values after irradiation becomes very slight at high temperatures.

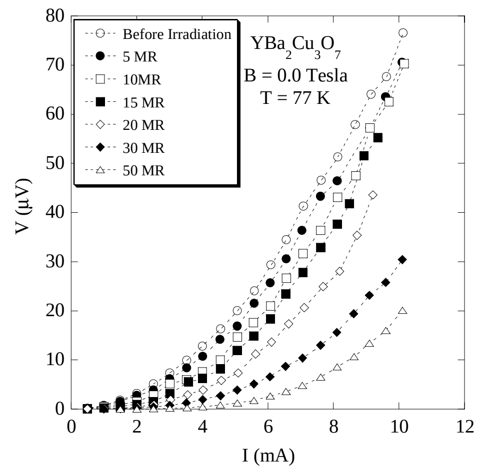

Figure 1: The *V-I* characteristic curves for the $YBa_2Cu_3O_7$ thick films at *77 K* at zero applied magnetic field for several doses of γ-rays.

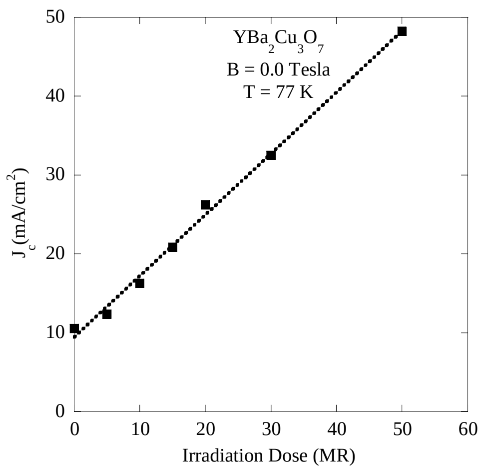

Figure 2: The behavior of the critical current density, J_c in the $YBa_2Cu_3O_7$ thick films at *77 K* as a function of γ-irradiation dose at zero applied magnetic field.

Three main results on the behavior of J_c were obtained. The first is the noticeable enhancement of J_c values at small doses of γ-irradiation in both types of samples. The second result is the slow rate of increase (almost saturated at high fields) of J_c values observed in the $Tl_2Ba_2Ca_2Cu_3O_{10}$ tapes as γ-irradiation dose is increased. The third result is the insignificant effect of γ-irradiation doses on J_c values in the $Tl_2Ba_2Ca_2Cu_3O_{10}$ tapes at high temperatures.

In order to explain these results, we need to understand the effects caused in the material after it is irradiated with γ-rays. The interaction of the energetic γ-rays with the HTSC material causes two major effects; atomic ionization [12] and atomic disorder [13].

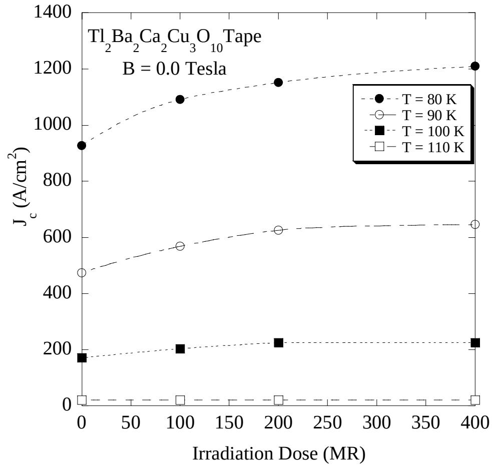

Figure 3: The behavior of the critical current density, J_c in the $Tl_2Ba_2Ca_2Cu_3O_{10}$ tapes at several temperatures as a function of γ-irradiation dose at zero applied magnetic field.

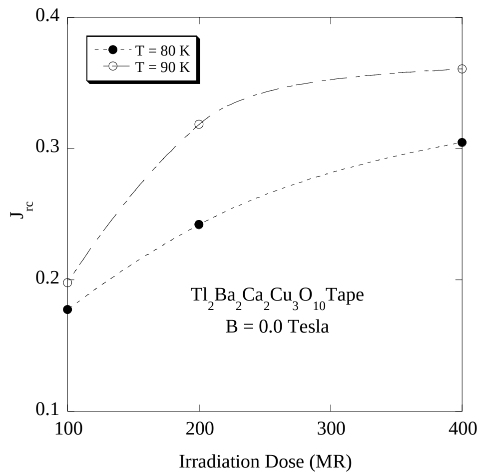

Figure 4: The relative critical current density, J_{rc} as a function of the irradiation dose at *80* and *90 K.*

Atomic ionization enhances the superconducting properties of the whole material especially those with low density of charge carrier. But because the density of charge carrier in the regions of the grains is relatively high [14], the γ-ray fluxes (up to *1000 MR*) produced by monoenergatic radioactive sources will not produce a significant enhancement of the superconducting properties in them. Thus γ-irradiation is not expected to have a positive influence on the transport properties in regions of the grains regardless of the γ-irradiation dose. But because the grain boundaries and the regions around them are strongly depleted from charge carriers [9], the atomic ionizations created by γ-rays well produce a significant effect on enhancing the superconducting properties. Thus the regions of the grain boundaries are very sensitive to γ-rays at all doses causing a net enhancement of the critical current density due to atomic ionizations.

The atomic disorders created by γ-irradiation are mainly of the microscopic type of disorder such as, vacancies and interstitials (point-like defects) of small separations and randomly distributed through the sample. The vortex pinning forces of these point-like defects are not expected to be significant in the region of the grains or the grain boundaries regardless of the γ-irradiation dose. But the regions of grain boundaries are known to produce effective vortex pinning forces, preventing vortex motion perpendicular to the grains orientation [10], and thus leading to an enhancement in the critical current density. At high doses of γ-rays, a more pronounced and opposite effect might take place in the regions of the grain boundaries. The weak links between the regions of the grains might be damaged by the high doses of γ-rays causing a suppression of the phase coherence of the superconducting wave functions (of the superconducting electrons) between grains in polycrystalline HTSCs [15, 16] leading to a suppression of the critical current density. Thus the grain boundaries of the polycrystalline HTSC material are expected to be very sensitive to γ-irradiation doses causing either a net enhancement (at low doses) or a net suppression (at high doses) of the critical current density.

The energetic γ-photons are also known to remove oxygen atoms from the CuO_2 resulting in oxygen disorder [17, 18]. Oxygen deficiencies in the CuO_2 planes also cause depletion of the concentration of hole-carriers which leads to a suppression in the critical current density [19, 20]. This effect is more pronounced in the regions of the grain boundaries since they are usually depleted from oxygen.

From this discussion we can see that regions of the grains are not as sensitive to γ-irradiation as the regions of the grain boundaries. This result has been confirmed by experiments on polycrystalline HTSCs [15]. The insignificant role of γ-irradiation on the superconducting properties of the grains has been also confirmed by experiments on HTCS crystals [21, 22]. Thus we can conclude that the major changes to the critical current density observed in our results are mainly due to the nature of the regions of the grain boundaries combined with their high sensitivity to γ-rays. The regions of the grain boundaries act as an enhancing factor of J_c because they have large vortex pining forces, impeding the motion of vortices between the grains. They act as a suppressing factor of J_c because they act as weak links between the grains. On the other hand, in the regions of the grain boundaries, γ-rays have two competing and opposite factors affecting J_c; the enhancing factor at low doses, and the suppressing mechanism at high doses. The enhancing factor of J_c is due to the atomic ionizations. The suppressing factor is due to oxygen disorder that leads to low carrier density and also due to the damage of the weak links between the grains which might occur at high doses.

We can explain the observed results in this study by considering this simultaneous existence of both factors in the grain boundaries and their varying influences with the dose level. At low γ-irradiation doses, the enhancing mechanism is dominant, leading to the noticeable enhancement of J_c in both types of samples. As the γ-irradiation dose increases the role of the suppressing mechanism due to damage of weak links grows up leading to the slow rate of increase of J_c shown in figure 3 for the $Tl_2Ba_2Ca_2Cu_3O_{10}$. At certain dose value, the role of the suppressing mechanism becomes comparable to the role the enhancing mechanism leading to the plateau of J_c shown in figure 3. It is expected that at higher dose than those used in this study, the role of the suppressing mechanism might become larger than that of the enhancing mechanism and leads to a decrease in J_c. The observed very small values of J_c and the insignificant effect of γ-irradiation on it in the $Tl_2Ba_2Ca_2Cu_3O_{10}$ tapes at the highest temperature might be attributed to the large vortex thermal energy which causes the effective pinning forces in the sample to become less significant in impeding the vortex motion resulting in a significant flow of vortices. This large flow of vortices causes a large suppression of J_c such that the enhancing factors caused by γ-rays are not large enough to cause a noticeable effect. It is also interesting to note that in the $Tl_2Ba_2Ca_2Cu_3O_{10}$ tapes, the changes in J_c as the dose of irradiation is increased are nonmonatonic with temperature. Figure 4 shows the relative critical current density, J_{rc} as a function of the irradiation dose at *80* and *90 K*. J_{rc} is defined by: $J_{rc} = \left(J_c^{irr} - J_c^o\right)/ J_c^o$, where J_c^{irr} is the value of J_c after irradiation with a certain dose and J_c^o is the value of J_c before irradiation. It is clearly seen that J_{rc} at *90 K* is always larger than that at *80 K*. But the difference in the enhancement at these two temperatures increases with the dose. This shows the nonmonatonic behavior of J_{rc} with the dose value and also indicates that the enhancing and the suppression mechanisms have a nonmonatonic dependence on temperature.

We believe that the specific doses at which the enhancing mechanisms take over and those at which the suppressing mechanisms take over depend on the specific granular structure of the material. This competing effect of γ-irradiation dose on the superconducting properties of HTSCs has been previously reported [16, 17].

Conclusion

We have investigated the effect different doses of γ-rays on the behavior of the critical current density, J_c in $YBa_2Cu_3O_7$ thick films and $Tl_2Ba_2Ca_2Cu_3O_{10}$ tapes at high temperatures. With increasing γ-irradiation dose up to *50 MR*, J_c was found to be significantly enhanced in the $YBa_2Cu_3O_7$ thick films. In the $Tl_2Ba_2Ca_2Cu_3O_{10}$ tapes, J_c was found to be significantly enhanced at γ-irradiation doses of *100 MR* at most temperature values. At higher doses, J_c was found to have a slight further enhancement and nearly reaches a plateau at the highest doses used. At the highest temperature values used, γ-irradiation was found to have insignificant effect on J_c in the $Tl_2Ba_2Ca_2Cu_3O_{10}$ tapes. These results were attributed to the competing roles of opposite mechanisms of γ-rays in the regions of the grain boundaries and also to the nature of the grain boundaries.

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