Pinning mechanism in (Bi,Pb)-2223 polycrystalline samples prepared with Al$_2$O$_3$ nano-particles

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Abstract. Nano-size Al$_2$O$_3$ (40 nm) addition effect on the polycrystalline (Bi,Pb)-2223 system properties was studied by XRD, SEM, TEM/EDX and electrical measurements. Samples with 0.0 and 0.2 wt% Al$_2$O$_3$ are synthesized in air by solid state reaction. Nanometer Al$_2$O$_3$ particles were added during the final sintering cycle of a multi-step preparation process. Analysis of magnetoresistivity has shown that the dissipation phenomenon follows the thermally activated flux creep model. Both onset temperature of dissipation, effective activation energy $U$, critical current density $J_c$ behaviour in applied magnetic field and volume pinning force density $F_p$ are enhanced by addition of 0.2wt.% Al$_2$O$_3$. Enhancement of the vortices flux pinning in doped samples is mainly originated from the surface normal-like pinning centers.

1. Introduction

Among the currently known high-Tc (HTc) superconductors, (Bi,Pb)-2223 appears to be the most promising candidate for the application at liquid-nitrogen temperature. However, their electrical transport properties drawback with increasing sample temperature and applied magnetic due to their low flux pinning energy. One of the possible solutions to enhance the flux pinning properties of HTc superconductors is by chemical doping and addition of nanosized particles [1-4]. Chemical dopants and preparation method play an important role on the microstructure and electrical properties of the superconducting material. Nano-sized particles added may be able to modify the crystalline structure and generate defects in superconducting matrix. Among the nanoparticles addition, only few ones are reported to react with the high-Tc superconductor materials during processing to become therefore second-phase [5]. It was found that Al$_2$O$_3$ nanosized particles addition into HTc superconductors results in enhanced of the pinning properties. Recently, we have studied systematically the effect of Al$_2$O$_3$ nanoparticles addition in YBCO compounds [5] and we have shown that the alumina react with YBCO to form nanometric inhomogeneities, containing aluminum, intergrowth within the superconducting matrix. The sub grain boundaries appeared in YBCO matrix are found to be efficient pinning centers. For bismuth based superconductors (BSCCO), the improvement of the $J_c$(H) of Bi-2212 has been demonstrated by the presence of Al-containing phases imbedded in the superconducting matrix [3]. We have also studies systematically the effect of nanometer Al$_2$O$_3$ addition on the (Bi,Pb)-2223 superconductor compounds. We have reported that the addition of a small amount (0.2wt.%) of Al$_2$O$_3$ during the final processing of (Bi,Pb)-2223 superconductors, did not affect its formation but increased the $J_c$ and improved the strength of pinning [6].

The broad resistivity transition of high-Tc superconductors in a magnetic field has been a subject of great interest. This effect has been widely considered to be the result of the thermally activated flux
motion of vortices and could be used to provide rich information about the properties of HTc superconductors at lower temperatures regime. With this goal in mind we extend our study on the effect of nanometer Al$_2$O$_3$ addition on the (Bi,Pb)-2223 polycrystalline superconductors by analyzing the resistivity as a function of temperature for different applied magnetic fields. The magnetoresistivity dependence on the temperature measurements were performed in free and 0.2wt.% Al$_2$O$_3$ addition samples. The effective pinning energy was estimated.

2. Experimental

Polycrystalline samples of (Bi,Pb)-2223 was elaborated by the solid-state synthesis route through a two-cycle annealing process. Details of the sample preparation have been described previously in Ref. [6], and herein we give a brief description. Nanometer Al$_2$O$_3$ with 40 nm in diameter was added during the second thermal cycle to the BSCCO precursor powder by mixing and hand grinding powders. In this work, the additional amount of Al$_2$O$_3$ represents 0 and 0.2% of the total mass of the sample. Then, powder mixtures were pressed into pellets, 0.3 mm thick and 5 mm in diameter, under a uniaxial pressure of 1GPa. These Pellets were sintered at 830 °C for 72 h in air. The structure and phase purity of the powder sample ground from sintered pellets were examined by powder XRD using a Philips 1710 diffractometer with CoKα radiation. The microstructure of samples was characterized using a scanning electron microscope (SEM) JEOL JEM-5510 and transmission electron microscope (TEM) FEI Tecnai G2 operating at 200 kV with a LaB$_6$ filament. The critical current density dependence on the applied magnetic field $J_c(H)$ was determined from $V-J$ characteristics using a criterion of 5μV/cm. The pellets were carefully cut into bars shaped samples with active cross section of the current flow of 0.3 mm$^2$. The temperature dependence of the electrical resistivity for different values of applied magnetic field, $T$, $J$ ($\mu$V/cm). The applied magnetic field was applied always perpendicular to the thickness and to the excitation current, that was injected along the major length of samples (noted wide sample surface). The acquisition of the data close to the transition was obtained with the temperature varying in rates of 0.4 K/min.

3. Results and discussions

To explore the effect of Al$_2$O$_3$ doping on the flux-pinning behaviour in the polycrystalline (Bi,Pb)-2223 superconductors. Two samples sintered with 0.0 and 0.2wt.%Al$_2$O$_3$, having same phase compositions and granular structure (see table. 1), were considered. Phase compositions were estimated by comparing the peak intensity of XRD powder.

| Samples x wt. % Al$_2$O$_3$ | Phase compositions (% vol) | $J_c$(A/cm$^2$) |
|-----------------------------|-----------------------------|-----------------|
| (Bi,Pb)-2223 | Bi-2212 | Ca$_3$PbO$_4$ |                     |
| 0.0 | 90 | 9 | 1 | 215 |
| 0.2 | 89 | 10 | 1 | 235 |

Measurements of the resistivity dependence on the temperature $\rho(T,0)$ for 0.0 and 0.2wt.% Al$_2$O$_3$ samples are shown respectively in inset of Fig. 1a and b. Both curves exhibit a transition to the superconducting state below onset superconducting temperature $T_{c}^{onset}$ from where the transition to zero resistance occurs. The temperature in which the zero resistance state, $T_{co}$, is 99 and 98 K for samples sintered respectively with 0.0 and 0.2wt.% Al$_2$O$_3$. At higher temperatures, both samples
exhibited linear temperature dependence, characteristics of metallic behaviour at normal state. The magnitude of the normal state resistivity decreases with addition of Al$_2$O$_3$.

The normalized temperature dependent resistivity $\rho(T)/\rho(120\text{K})$ in various magnetic fields for free and 0.2wt.% Al$_2$O$_3$ added samples are shown respectively in Fig 1a and b, where $\rho(120\text{K})$ is the resistivity at temperature 120 K. A broadening of the resistive transition with an increase of the applied magnetic field is obviously observed. Such behavior is a remarkable feature of granular superconductors [7,8]. In the presence of the magnetic field, the $\rho(T,H)$ curves are characterized by two distinctive sections; at high temperature a branch which reveals the intragranular transition and at lower temperature a broad foot structure associated with the grain-boundaries network or intergranular effects, which are considered to be weak Josephson type links [9]. The Broadening of 0.2 wt.% Al$_2$O$_3$ added sample is apparently reduced as compared to free one. The temperature at which the $\rho(T,H)$ curves start to departure each other for different applied magnetic fields is, $T_{\text{off}}$, is well accepted in the literature that is correspond to the final transition of the grains in granular superconductors and the resistivity at this temperature is residual intergranular resistivity. In the presence of magnetic field this temperature resulted to be the same, for 0.2wt% Al$_2$O$_3$ added sample, $T_{\text{off}}=103.5\text{K}$. Moreover, $T_{\text{off}}$ decreases in free sample with an increase of the external applied magnetic field over 500 Gauss. Similar features of the $\rho(T,H)$ curves were reported for the foamed Bismuth based superconductor polycrystalline [7].

![Figure 1](image-url)

**Figure 1.** $\rho(T)/\rho(120\text{K})$ curves in the transition region for different applied magnetic fields of free sample (a) and 0.2 wt% Al$_2$O$_3$ added sample (b). Inset: $\rho(T)/\rho(120\text{K})$ in zero applied magnetic field.

In the $\rho(T,H)$ curves, we define the zero resistance transition temperature under the action of magnetic field as the onset of the dissipation temperature $T_{\text{off}}$. The $T_{\text{off}}(H)$ curves for both samples are represented in Fig. 2a. It can be observed that at fixed applied magnetic field, $T_{\text{off}}$ increases with Al$_2$O$_3$ addition. This result displays clearly the efficient effect of Al$_2$O$_3$ addition to delay the onset of dissipation phenomena. Numerous works have demonstrated that the onset of the dissipation in these high-Tc ceramics starts at the weak-links regions, which are zones of weak superconductivity [10]. These weak-links limit the supercurrent flow, via Josephson effect, to narrow channels of width lower than the coherent length [11].

The broadening of the resistivity and dissipation phenomenon is usually understood with thermally assisted flux motion theory. In the classical Anderson flux creep model, the resistivity caused by the
motion of vortices is given by the Arrhenius relation with the form 
\[ \rho = \rho_0 \exp(-U/k_B T) \]; where 
\( \rho_0 \) is the pre-exponential factor independent of field and orientation, 
\( k_B \) is the Boltzman constant and 
\( U \) is the real activation energy that generally depends on the temperature \( T \), the current density \( J \) and the applied magnetic field \( H \). Generally, the current density dependence of activation energy is a constant when the measuring current density is low [12]. The plots of \( \log \rho(T)/\rho(120K) \) against \( 1/k_B T \) give \( U \) as the slope value. For free and 0.2wt.% Al\(_2\)O\(_3\) added samples, the reverse temperature dependence of \( \log \rho(T)/\rho(120K) \) exhibit a linear behavior in the low-resistivity part of the \( \rho(T) \) curves at various applied magnetic fields. This indicates that the activation energy is independent of the temperature. The calculated \( U \) of both samples as function of applied magnetic fields \( H \) is plotted in Fig. 2b. In agreement with the observed behavior in the dependencies of the dissipation temperature \( T_{\text{off}} \) as a function of applied magnetic field, the 0.2 wt% Al\(_2\)O\(_3\) added sample exhibits the higher values of intergranular pinning energy in all the range of applied magnetic field.

![Figure 2.](image)

Fig. 2. \( T_{\text{off}} \) (a) and Effective pinning energy, \( U \), (b), as a function of applied magnetic of free and 0.2 wt% Al\(_2\)O\(_3\) added samples. In all curves lines between points are guides for eyes.

Fig. 3 shows the dependence of normalized critical current densities (e.i. \( J_c(H)/J_c(0) \)) on applied magnetic field for free and 0.2 wt% Al\(_2\)O\(_3\) added samples at 77 K. Applied magnetic field is oriented perpendicular to the sample wide surface. To isolate the flux pinning improvements, the measured \( J_c(H) \) values in magnetic field can instead be normalized to their zero field values \( J_c(0) \). Both samples show degradation of the normalized critical current density with increasing the field. The Al\(_2\)O\(_3\) added sample have much slower \( J_c(H)/J_c(0) \) drops than non-added one for applied fields. The above results suggest that the improvement of critical current density behaviour under applied magnetic field is due to flux pinning enhancement, resulting from pinning centers generated by nanosized Al-rich phase introduced in (Bi,Pb)-2223 matrix. More than one pinning mechanism may exists and their effects will add up in HTc superconductors, due to the complexity of these materials.
It is widely accepted that a very fruitful tool which one can investigate the flux pinning properties is the determination of volume pinning force density, $F_p = J_c \times B$ from experimental measurements of critical current density at each applied magnetic field. Fig. 3b shows the plots of, $F_p$ as a function of reduced field, $h = H/H_{irr}$ at 77 K for free and Al$_2$O$_3$ added samples. It is clear that improvement pinning force density strengthen was observed in the sample with 0.2wt.% Al$_2$O$_3$ addition. This is an improvement by about 40 % compared to sample without particles addition. For both samples, the large curves obtained indicate that the pinning properties cannot be explained by only one pinning mechanism over the whole region of the $F_p(h)$ curves, which implies that more than one type of pinning source may have been present at the same time. Moreover we have done a detailed analysis on the scaling of $F_p$ versus applied field based on the Dew-Hughes model [13] and we have found that the dominant pinning mechanism is surface normal-like pinning centers generated by the inclusion of Al-rich phase in the superconducting matrix [6]. This speculation is supported by TEM observations. Indeed, TEM and EDS analyses have shown that alumina reacts with the BSCCO matrix to form nanometric Al–rich phase intergrowth within the superconducting matrix. High resolution TEM image (Fig. 4) shows a sharp interface between Al-rich phase and (Bi,Pb)-2223 phase, which provides effective flux pinning.

4. Conclusion
Nano-size Al$_2$O$_3$ (40 nm) addition effect on the pinning properties of polycrystalline (Bi,Pb)-2223 system was analyzed. Samples with 0.0 and 0.2 wt% Al$_2$O$_3$ are synthesized in air by solid state reaction. Both samples, display same (Bi,Pb)-2223 phase concentration (~ 90 %) and granular structure. We have studied the magnetoresistivity properties of Al$_2$O$_3$ doped (Bi,Pb)-2223 polycrystalline samples in applied magnetic field, H, ranging from 0 to 2 kG. The analysis of resistive transitions in a magnetic field has shown that the dissipation phenomenon follows the thermally activated flux creep model. The Al$_2$O$_3$ doped sample exhibits better transport properties; the onset temperature of dissipation and the effective activation energy increase with an addition of 0.2 wt.% Al$_2$O$_3$. $J_c$ behavior in applied magnetic field and volume pinning force density are also improved. Enhancement of the vortices flux pinning in 0.2wt.% Al$_2$O$_3$ doped samples is mainly originated from the surface normal-like pinning centers.
Figure 3. High resolution image taken along the [001] direction, showing interface between Al-rich phase (upper) and the (Bi,Pb)-2223 phase (below).

5. References
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