Study of Electric Resistance and Magnetoresistance for $Ag_x(Tl_{0.8}Pb_{0.1}Bi_{0.1})Ba_2Ca_2Cu_3O_{9-\delta}$ Superconductors

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Superconducting samples of the type $Ag_x(Tl_{0.8}Pb_{0.1}Bi_{0.1})\ Ba_2Ca_2O_{9-\delta}$ with ($0 \leq x \leq 0.4$) are prepared from the high purity oxides Tl_2O_3, PbO, Bi_2O_3, BaO_2, CaO, CuO, and Ag metal at 870°C for 4h. The characterization, electric resistance and magnetoresistance for the prepared samples are performed. Ag addition to $(Tl_{0.8}Pb_{0.1}Bi_{0.1})Ba_2Ca_2Cu_3O_{9-\delta}$ shows a reduction in both lattice parameters "a" and "c" while the grain size is improved till about x=0.2. Resistance data show that the superconducting critical temperature "T_c" has little variation with increasing silver addition. Oxygen annealing of these samples causes an improvement in zero resistance temperature "T_0". The excess conductivity studies show that there are three-dimension fluctuations (3D) in the mean field region for either pure sample or samples containing Ag. As the silver addition increases the transverse magentoresistance decreases till x=0.2 and then increases. Transverse magnetoresistance vs. applied magnetic field at different temperatures shows a decrease as the temperature reaches the superconducting critical temperature. The driving current has an effect in the value of magnetoresistance near the zero resistance temperature but it does not affect the value of the magnetoresistance near the superconducting critical temperature.

Introduction

Since the discovery of superconductivity in the Tl-Ba-Ca-Cu-O system [1], many attempts have been made to synthesize monophasic of Tl-1223 [2-3]. Tl-1223 phase is represented as an interesting phase of the Tl-Ba-Ca-Cu-O system, because it has high superconducting critical temperature "T_c" and a high critical current density "J_C". So Tl-1223 phase is one of the most promising materials for application in high magnetic field at liquid nitrogen temperature. Generally, Tl-1223 phase was reported for the two compositions $TlBa_2Ca_2Cu_3O_{9-\delta}$[2] and $(Tl_{0.5}Pb_{0.5})Sr_2Ca_2Cu_3O_{9-\delta}$ [4]. The substitutions of Hg, Y [5,6] and In [7] into Tl-sites in the $TlBa_2Ca_2Cu_3O_{9-\delta}$ were studied. They found that a small addition of Hg enhances the transition temperature from 122 K to 131 K whereas the addition of In reduces the transition temperature from 122 K to 100 K. Also, $(Tl_{0.5}Pb_{0.5})$ $Sr_2Ca_2Cu_3O_{9-\delta}$ can be improved by partial substitution of Ba^{+2} into Sr^{+2} sites and Tl by Pb and Bi [8,9]. These substitutions enhance the formation of Tl-1223 and improve the superconducting properties. Triscone et al. [10] found that the $[(Tl_{0.6}Pb_{0.2}Bi_{0.2})$ $(Sr_{0.81}Ba_{0.1}Ca_{0.09})_2$ $(Ca_{0.94}Tl_{0.06})_2$ $Cu_3]$ $O_{9-\delta}$ has a transition temperature around 115 K.

The serious problems up to now, in the high temperature superconducting materials are the weak links between the grains and the brittleness of these materials. These problems make difficulty in large-scale applications such as long wires or tapes with high intergrain "J_C" in high magnetic fields. Silver addition is used to improve the grain size, mechanical properties of high-T_c ceramic superconductors [11,12] as well as to make ductile superconducting tapes and wires. Also, the advantage of using silver is that it enhances the critical current density and the pinning of the vortices. Khan et al. [13] reported the addition of silver in Hg-1201 compound and they found that 2 at.% silver addition increases the intergrain current from $0.3x10^6$ A/cm^2 to $0.6x10^6$ A/cm^2.

In this report we describe the synthesis and characterization of $Ag_x(Tl_{0.8}Pb_{0.1}Bi_{0.1})Ba_2Ca_2Cu_3O_{9-\delta}$ with ($0 \leq x \leq 0.4$) superconducting samples. The transverse magnetoresistance of these samples is measured over a field range from 0 to 4.8 kG at different applied driving currents (1, 10 and 100 mA) and at different temperatures ranging between onset temperature and zero resistance temperature.

Experimental

Bulk samples of the type $Ag_x(T_{0.8}Pb_{0.1}Bi_{0.1})Ba_2Ca_2Cu_3O_{9-\delta}$ with ($0 \leq x \leq$) 0.4) were synthesized by a soild-state reaction. Appropriate amounts of high purity powders, Tl_2O_3, PbO, Bi_2O_3, BaO_2, CaO, CuO and Ag-metal, were weighed in the correct stoichiometric proportions and ground in an agate mortar. The ground powders were sieved using 75 μm sieving. All operations were carefully carried out in a dry glove box filled with argon to reduce the reaction of CaO with CO_2 and moisture. The powder pressed in the form of a disc of diameter 1.5cm and thickness 0.3cm. Then, the sample was wrapped in a silver foil to reduce possible volatilization of T1, Bi and Pb that could eventually react with the small quartz tube furnace. The sample was heated at a rate 240 ^oC/h to 870 ^oC maintained at this temperature for 3.5 h and then cooled to room temperature by a cooling rate 300 ^oC/h. The prepared samples are annealed in oxygen flow at 580^oC for 6h. Subsequently, the furnace was cooled down to room temperature by a cooling rate of 2 ^oC/min.

X-ray diffraction (XRD) analysis was performed using Philips PW-1729 powder diffracotmeter using Cu-Kα radiation, λ = 1.5418 Å. The microcrystallity of the samples were examined using a Joel scanning electron microscope JSM-5300, operated at 15 kV, with resolution power of 4 nm.

The electrical resistance was measured using a standard four point probe method in the temperature range from room temperature down to zero resistance temperature. The samples used for resistance measurements have dimensions of about 1.5x0.3x0.1 cm^3 and fine copper wires made the electrical contacts to the sample with a conductive silver paint. The applied driving current is 1 mA. The transverse magnetoresistance was measured in a low magnetic field ranging from 0 kG to 4.8 kG at three driving currents 1, 10 and 100 mA. The measurements were performed at three temperatures lie between onset temperature and zero resistance temperature. The external magnetic field was applied perpendicular to the driving current and it was produced from an electromagnet. The temperature was measured using Kp-Au 0.07 at.% Fe thermocouple and stabilized with the aid of a temperature controller within $\pm$ 0.1 K.

Results and Discussion

The refined lattice parameters "a" and "c", determined from x-ray diffraction patterns, for the tetragonal phase in Ag_x $(Tl_{0.8}Pb_{0.1}Bi_{0.1})$ $Ba_2Ca_2Cu_3O_{9-\delta}$ are shown in Fig. (1). The data in this figure show that the addition of Ag to $(TI_{0.8}Pb_{0.1}Bi_{0.1})$ Ba_2 Ca_2 Cu_3 $O_{9-\delta}$ has a very small reduction in both ,,a, and ,,c,, This means that the silver addition does not change the lattice

structure but about 0.96% reduction in cell volume is observed when we add 0.4 Ag. These results are in agreement with the data of Khan et a1. [13], who found that 0.02 Ag additions in Hg-1201 reduce the volume of the cell by 1 %.

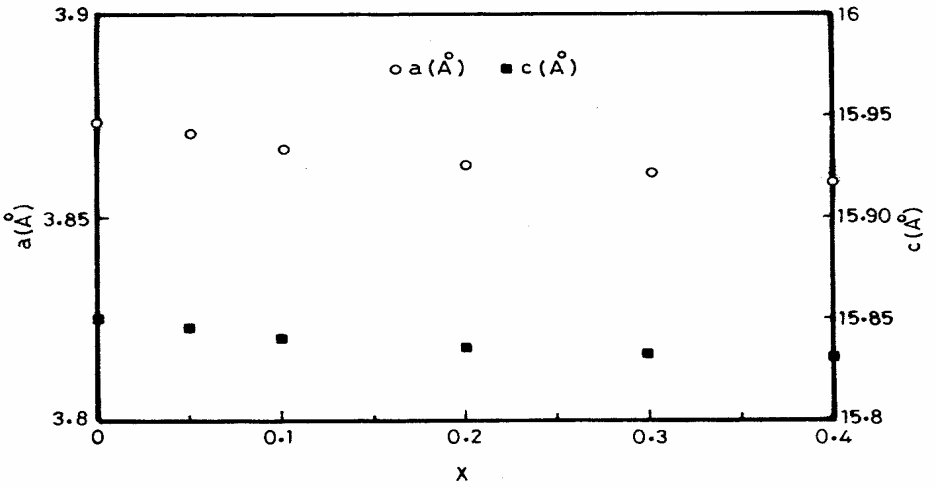

Fig. (1): Variation of lattice parameters "a" and "c" vs. Ag contentx.

In order to evaluate the microstructure features of the $Ag_x(T1_{0.8}Pb_{0.1}Bi_{0.1})Ba_2Ca_2O_{9-\delta}$ synthesized in the present investigation, the samples were subjected to scanning electron microscope (SEM). Typical SEM micrographs are shown in Fig. (2) for Ag_x ($T1_{0.8}$ $Pb_{0.1}$) Ba_2 Ca_2 Cu_3 $O_{9-\delta}$ with x=0, 0.1,0.2 and 0.4, respectively. For x=0, we notice that the needles are predominant. As the silver content increases the granular structure is appeared among the needles, which becomes like rods (Fig. 2b). The granular structure is increased by increasing silver content to x = 0.2 and also the grains size. For x > 0.2, a nearly melting and inhomogeneous samples are appeared (Fig. 2d). So, we can expect that the silver additions improve the samples till x = 0.2 and then they worst the samples.

Figure (3) displays the temperature dependence of the normalized resistance R (T)/R (294) for $Ag_x(T_{10.8}Pb_{0.1}Bi_{0.1})Ba_2Ca_2Cu_3O_{9-\delta}$ with x = 0.1, 0.2 and 0.4 for as-synthesized and as-oxygen annealed samples. All the samples are characterized by a metallic behavior form room temperature down to 200 K. Basically the normal metallic behavior reflects the conduction along the CuO planes. The deviation from the normal state linear behavior, starting above "Tc" to 200 K, indicates the super conducting fluctuation of the electron pairs. Increasing of silver content shows that the residual normalized resistance decreases till about x = 0.2 and then increases. These results indicate that the silver addition

improves the intergrain electrical contacts till x = 0.2. All the samples have a sharp transition to zero resistance temperature except the sample with x = 0.4 which has a tail to reach the zero resistance temperature.

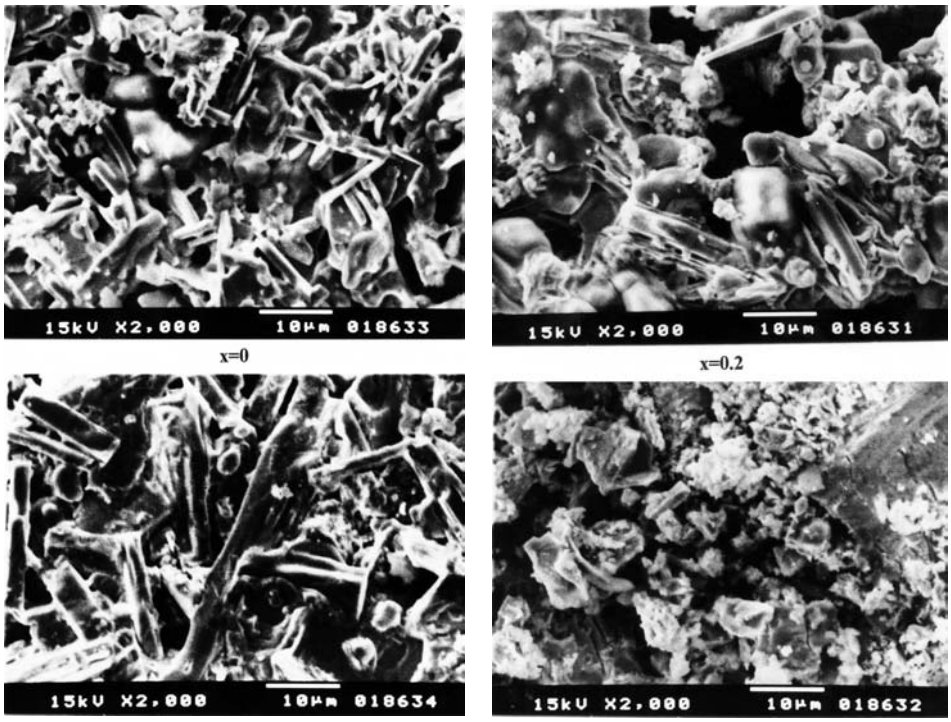

Fig. (2): Typical morphology for $Ag_x(Tl_{0.8}Pb_{0.1}Bi_{0.1})Ba_2Ca_2Cu_3O_{9-\delta}$ (x=0, 0.1, 02 and 0.4).

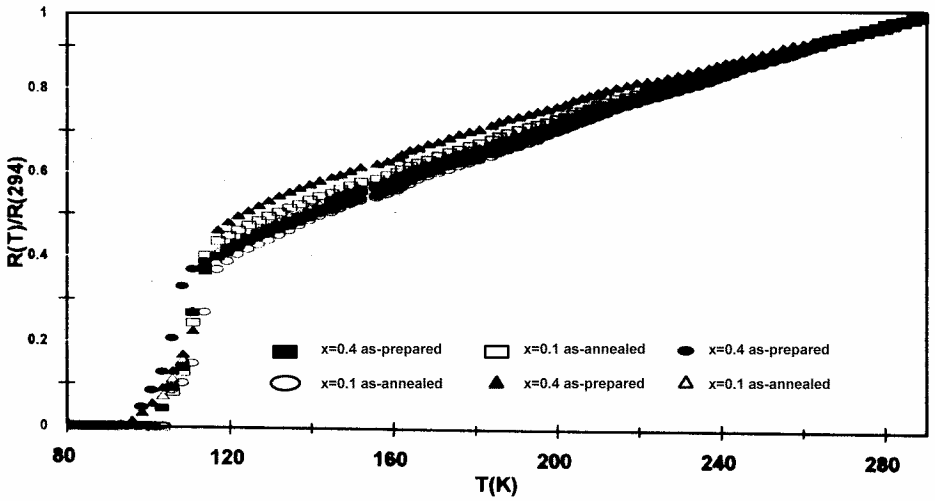

Fig. (3): Normalized electrical resistance for $Ag_x(Tl_{0.8}Pb_{0.1}Bi_{0.1})Ba_2Ca_2Cu_3O_{9-\delta}$ as-synthesized and as-oxygen annealed (x=0, 0.1, 02 and 0.4).

Table (1) gives the super conducting critical temperature “Tc” and zero resistance temperature “To” before and after oxygen annealing vs. silver content. It is clear that the silver addition enhanced the transition temperature form 111 K to 114 K and reduced the transition width “ΔT “ except the sample with x = 0.4. This means that the silver metal can absorb oxygen from (T1, Pb, Bi) Oy 1ayer and can change the sample from over doped regime to nearly optimum doping level.

Table (1): Superconducting critical temperature “T_c” and zero resistance temperature “T_0” for $Ag_x(Tl_{0.8}Pb_{0.1}Bi_{0.1})\ Ba_2Ca_2Cu_3O_{9-\delta}$.

Ag-content, X	**As-synthesized**		**As-oxygen annealed**	
	T_c (K)	**T_0 (K)**	**T_c (K)**	**T_0 (K)**
0.0	111.5	98.5	109.5	101.0
0.05	114.0	103.5	N.M	N.M
0.1	114.0	103.5	115.5	106.0
0.2	114.0	101.0	117.0	103.5
0.3	114.0	101.0	114.0	103.5
0.4	114.0	93.3	114.0	98.0

The largest transition width "ΔT" for x = 0.4 sample is probably due to silver absorbed more oxygen from the rock salt layer leaving oxygen vacancies. Also, we notice that the silver addition has a litter influence on the superconducting critical temperature because the silver does not enter the structure and does not affect the structure of high temperature superconductors [14]. The oxygen annealing causes a reduction in the superconducting critical temperature "Tc" for x = 0, but it has no effect on the “Tc “for x = 0.3 and 0.4. The samples with x = 0.1 and 0.2 are the only samples that have an enhancement in their transition temperatures. Zero resistance for all the samples is improved by the oxygen annealing. This improvement in the zero resistance temperature could be explained as that the oxygen annealing reduces the oxygen vacancies inside the samples.

In order to study two- dimensions (2D) and three- dimensions (3D) fluctuaton analysis for our samples, a plot of the logarithm of the excess conductivity $\Delta\sigma/\sigma_o$ vs. the logarithm of the reduced temperature $\varepsilon = (T-Tc)/Tc$ For x=0, 0.1 and 0.2 samples is shown in Fig. (4). The excess conductivity above “Tc “, according to the mean field approximation in BCS theory may be written as [15]:

$$\Delta\sigma/\sigma_o = A\in^n, \qquad (1)$$

where σ_o is the normal conductivity at room temperature, A is the temperature independent amplitude and n is the critical exponent and equal – 0.5 and – 1 for 3D and 2D fluctuations, respectively. Excess conductivity analysis shows that, for x = 0, 0.1 and 0.2 samples, the critical exponent n are closed to –0.5 in the regime $-2 < \ln \varepsilon < -3.5$ as predicted by Aslamazov and Larkin (AL theory) [15] for (3D) fluctuation in the mean field region. Also, these data are expected for systems with large anisotropy and its Ginzburg-Landau (GL) coherence length (ξ_c) is smaller than any separation between the Cu-O planes [16].

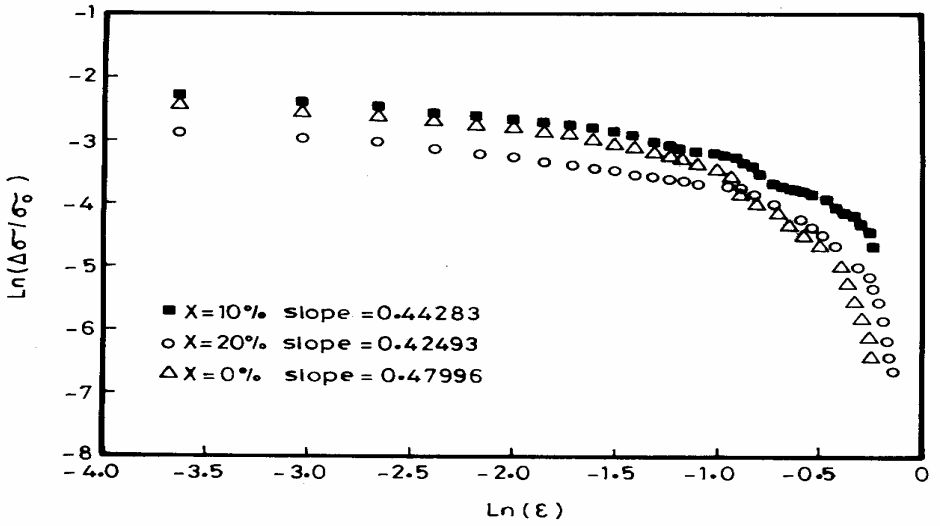

Fig. (4): Log-log plot of the normalized excess conductivity vs. reduced temperature for $Ag_x(Tl_{0.8}Pb_{0.1}Bi_{0.1})Ba_2Ca_2Cu_3O_{9-\delta}$.

Transverse magnetoresistance $\Delta\rho_\perp/\rho_o$ (ρ_o is the resistivity at zero applied magnetic field) for $Ag_x(Tl_{0.8}Pb_{0.1}Bi_{0.1})Ba_2Ca_2Cu_3O_{9-\delta}$ with ($0 \leq x \leq 0.4$) vs. external magnetic field, as synthesized, is shown in Fig. (5) at the same reduced temperature. It is clear that the value of transverse magnetoresistance decreases with the increasing of silver content until around x=0.2 and then increases. We notice that the samples with x = 0.1 and 0.2 reach the plateau with increasing of magnetic field faster then the other samples. Also it is clear that the values of magnetoresistance at fields below 1 kG do not depend on silver contents. These results mean that the silver addition could improve the flux pinning of vortices. These results are consistent with those obtained by Mogilko et al. [17], who found that the addition of silver to Bi-2223 and Bi-2212 produced two main types of pinning centers. The first one is strong pinning and almost independent on the Ag-contents. The second one is weak pinning allowing fluxon transport across the grain boundaries. The Ag presence increases the strength of these pinning sites.

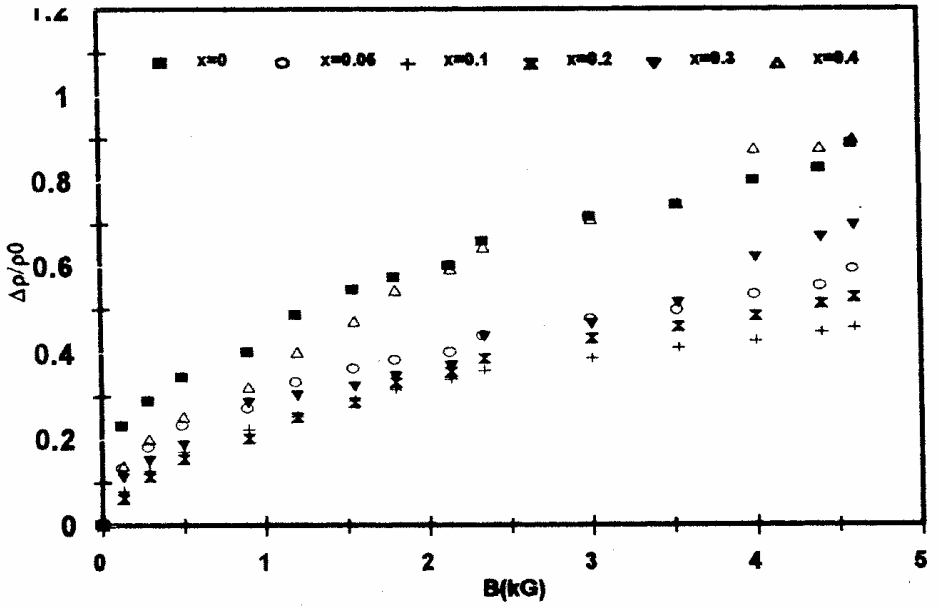

Fig. (5): The transverse Magnetoresistance vs. applied magnetic field for Ag_x $(Tl_{0.8}Pb_{0.1}Bi_{0.1})$ $Ba_2Ca_2Cu_3O_{9-\delta}$ (x=0, 0.1, 01 and 0.2).

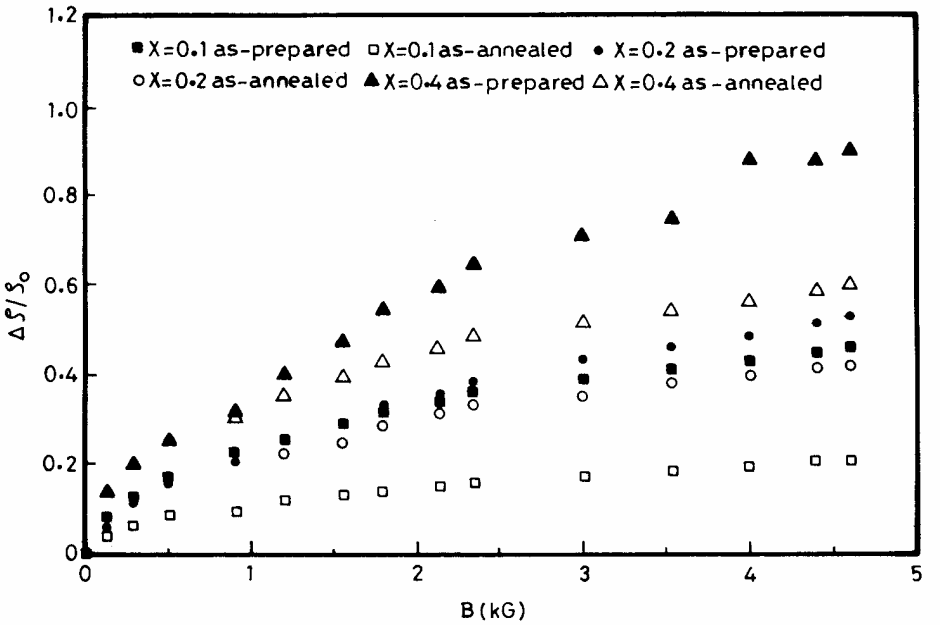

Fig. (6): The transverse magnetoresistance vs. applied magnetic field for Ag_x $(Tl_{0.8}Pb_{0.1}Bi_{0.1})$ $Ba_2Ca_2Cu_3O_{9-\delta}$ as-synthesized and as-oxygen annealed at 111 K.

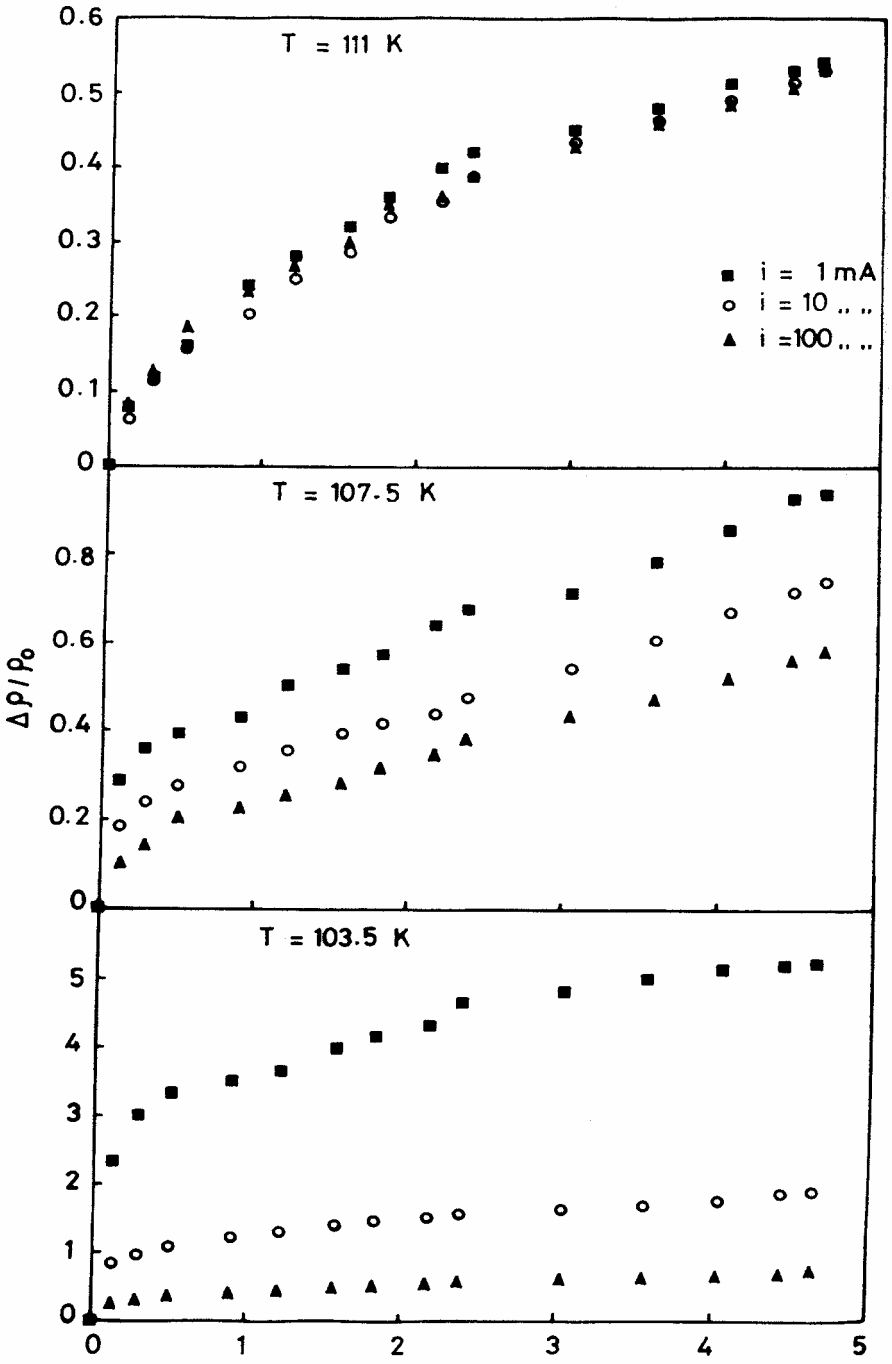

Fig. (7): The transverse magnetoresistance vs. applied magnetic field for Ag_x $(Tl_{0.8}Pb_{0.1}Bi_{0.1})$ $Ba_2Ca_2Cu_3O_{9-\delta}$ at different applied currents temperatures.

Figure (6) displays the variation of the transverse magnetoresistance $\Delta\rho_{\perp}/\rho_o$ vs. external magnetic field at 111 K for $Ag_x(Tl_{0.8}Pb_{0.1}Bi_{0.1})$ $Ba_2Ca_2Cu_3O_{9-\delta}$ with x = 0.1, 0.2 and 0.4 as-synthesized and as-oxygen annealed samples. The value of magnetoresistance for oxygen-annealed samples is less than that for synthesized samples. The plateau at high external magnetic field is more noticed in the oxygen-annealed samples more than in the synthesized samples. It is well known that from the resistivity measurements, the oxygen annealing improved the zero resistance temperature and intergrain connection between the grains. This improvement in zero resistance temperature could reduce the magnetoresistance resulting from the flux flow effect.

Figure (7) displays the variation of transverse magentoresistance $\Delta\rho_{\perp}/\rho_o$ vs. applied magnetic field for $Ag_x(Tl_{0.8}Pb_{0.1}Bi_{0.1})$ $Ba_2Ca_2Cu_3O_{9-\delta}$ at different driving currents 1, 10 and 100 mA The measurements were carried out at 103.5, 107 and 111 K. The value of transverse magnetoresistance is more affected by the driving current at 103.5 K than that at 107 K. We found that $(\Delta\rho_{\perp}/\rho_o)_{I=1mA}/(\Delta\rho_{\perp}/\rho_o)_{I=10mA}$ is nearly equal to $(\Delta\rho_{\perp}/\rho_o)_{I=10mA}/(\Delta\rho_{\perp}/\rho_o)_{I=100mA}$ with 4.6 kG at both 103.5 K and 107 K. Three curves of transverse magnetoresistance at 111K are overlapping and do not vary with increasing driving currents. This means that the effect of driving current decreases as the temperature reaches the superconducting critical temperature. From these results we can conclude that the decoupling temperature "T_d" is less than 111 K. The overlapping of magnetoresistance curves at 111 K is due to the absence of flux trapped (above the knee), which hints that vortex pinning may be ineffective at elevated temperatures. These results are consistent with our previous results for (Hg, T1)-1223 [6], (T1, In)-1223 [7] and (Tl, In)-1212 [18] which showed that both driving currents and magnetic field are ineffective at the first stage of transition and enlarged the transition width in the second stage of transition. Also, these data could contain some information concerning the special distribution of the "strong" superconducting grains and "weak" superconducting boundaries.

Conclusions

We have successfully synthesized $Ag_x(Tl_{0.8}Pb_{0.1}Bi_{0.1})Ba_2Ca_2Cu_3O_{9-\delta}$ superconductors with ($0 \leq x \leq 0.4$) using a first step of solid-state reaction starting from the high purity oxides. The silver addition does not change the tetragonal structure of T1-1223 phase but the lattice parameters "a" and "c" are reduced. As the silver addition increases, the grain size is improved until x= 0.2 and then nearly diffused samples are formed. The superconducting critical temperature "T_c" enhances from 111.5 K to 114 K by the silver addition and the resistance approaches zero at nearly equal temperatures except the sample with x = 0.4. Oxygen annealing suppresses the "T_c" from 111.5 to 109.5 K for x = 0 whereas it enhances "T_c" from 114 to 117 K for x = 0.2. The zero resistance

temperature "T_0" is improved for all the samples by oxygen annealing, indicating that the Ag-metal produced oxygen vacancies. The data of the excess conductivity above "T_c" show that the silver addition does not change the 3D fluctuation in Tl-1223 phase. The transverse magnetoresistance data as a function of temperature showed a decrease as the temperature reaches "T_c". The driving current has an effect on the value of magnetoresistance only at temperatures below the decoupling temperature "T_d". As the silver addition increases the transverse magnetoresistance decreases up to x = 0.2 and then increases.

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