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Superconducting Properties of $(\text{Tl}_{1.6}\text{Pb}_{0.4})\text{-2223}$ Substituted by Praseodymium

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Abstract: The lattice parameters, electrical resistivity, ac magnetic susceptibility, magnetoresistance and Hall effect have been measured for a newly developed compound obtained through partial substitution of Ca^{2+} ions by Pr^{3+} ones in $(\text{Tl}_{1.6}\text{Pb}_{0.4})\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10.8}$. For such compound, the prepared samples are almost single phase of (Tl, Pb)-2223 tetragonal unit cell with a space group 14/mmm. An opposite behavior for both lattice parameters a and c was observed as the concentration of Pr-content was increased. Also, metallic tendency was observed from electrical resistivity measurements in the range of $0 \leq x \leq 0.15$ for temperature values above the superconducting transition temperature T_c . On the other hand, a semiconductor-like behavior was noticed for values $x > 0.15$. Suppression in the superconducting transition temperature was found as the Pr-content was increased. This suppression in superconductivity for the system evolved was discussed according to two models point of views; the Cooper-pair breaking and hole-filling models. The transverse magnetoresistance, below the superconducting transition temperature, was measured in a weak magnetic field up to 4.8 kg. Finally, the Hall voltage was measured at different temperature values above that of superconducting transition using five-probe technique. The Hall coefficient, Hall mobility and Hall angle were expressed as functions of both temperature and Pr-content.

Key words: Resistivity, superconducting transition temperature, Hall coefficient, Hall mobility, Hall angle

INTRODUCTION

Chemical substitutions have always been a very effective way to probe the properties of high-temperature superconductors HTSC's, (Wang *et al.*, 2000). The substitutions in the Cu- and Ca-sites provide valuable insight about superconductivity mechanisms. This is because the conduction of charge carriers mainly occurs through the conduction-slab as shown by Xu *et al.* (2000). Moreover, such substitutions were considered the most effective way for creating the pinning centers in HTSC's (Shi *et al.*, 1989; Shakeripour and Akhavan, 2001). This lead to an enhancement of critical current density used in high technology. For Tl-cuprates, most of substitution studies were concerned with Tl-2223 for its high T_c and with Tl-1223 for its potential use in industry. Partial replacement of thallium by rare-earth elements such as Gd, Er and Yb was presented by many researchers (Abou-Aly *et al.*, 2000; Awad, 2002). Through out their studies, it was found that the superconducting transition temperature depressed as the rare-earth element contents were increased. Also, they discovered that the critical concentration, at which the superconductivity disappeared, relies on the magnetic moment of substituted elements.

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Praseodymium experiences the same properties of rare-earth elements group. At high temperatures it is a paramagnetic element showing a strong anisotropy. When the temperature was lowered enough, it turned anti-ferromagnetic. The magnetic moment of Pr^{3+} is $3.58 \mu_B$ and is considered as one of the light rare-earth elements. Many studies have been achieved to explore the influence of Pr-substitution on high-temperature superconductors. Salamati *et al.* (2003) investigated the effect of Pr-substitution on the superconductivity and interlayer-coupling of the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ phase. His results showed that the superconducting transition temperatures enhanced for lower Pr-content but depressed for higher one. The analysis of excess-conductivity, for his samples, showed that the interlayer-coupling constant decreased monotonically with increasing of the Pr-content. (Aloysius *et al.*, 2005) discussed the effect of Pr-substitution on the superconducting properties of (Bi, Pb)-2212 system. They found that the critical current density, J_c , as well as superconducting transition temperature values for Pr-substituted samples were higher than those of non-substituted ones. Distinct variations in grain morphology were noticed in samples containing Pr-content when SEM technique has been used. Moreover, the composition porosity increased as the Pr-content concentration increased, leading to significant reduction in sample density. Awad and Mohammed (2004) investigated the effect of Pr^{3+} ions on Tl-1223 system. The phase structure of Tl-1223 sustained its composition when replacing Tl^{3+} ions with those of Pr^{3+} . Also, solubility limit of Pr was found to be at $x_s = 0.15$. The superconducting transition temperature depressed as the Pr-content increased. The critical content, at which the superconductivity disappears, was found to be at $x_c = 0.29$. The depression in T_c was interpreted in terms of hole-filling and Cooper pairs breaking mechanisms.

This study aims to investigate the influence of introducing Pr^{3+} ions into (Tl, Pb)-2223. Through out this research, superconducting samples of type $(\text{Tl}_{1.6}\text{Pb}_{0.4})\text{Ba}_2\text{Ca}_{2-x}\text{Pr}_x\text{Cu}_3\text{O}_{10-\delta}$ with $0 \leq x \leq 0.3$ have been prepared using solid-state reaction technique and had been investigated using x-ray powder diffraction. The electrical resistivity, ac magnetic susceptibility, magnetoresistance and Hall effect have been measured for the samples under investigation.

MATERIALS AND METHODS

Samples of the concerned specimens with different doping levels $0 \leq x \leq 0.3$ were prepared by a single-step of solid-state reaction technique. Amounts of Tl_2O_3 , PbO_2 , BaO_2 , CaO , Pr_6O_{11} and CuO were used to prepare one sample of about 2.3 g with nominal composition as 1.8:0.4:2:2-x:x:3. Then, the samples were grinded in an agate mortar and then sifted using a $135 \mu\text{m}$ sieve. After that, the powder was pressed in a disc having dimensions of 1.5 cm diameter and about 3.0 mm thickness. This disc was wrapped in a silver foil in order to reduce the thallium evaporation during the preparation. All these operations were carried out in a glove box under argon atmosphere to prevent the absorption of moisture and CO_2 . The heat treatment for preparing the sample was carried out in four successive steps. Firstly, the samples were heated in a sealed quartz tube at a rate of 4°C min^{-1} to 760°C . Secondly, heating was continued with different rate of 2°C min^{-1} till 860°C was reached. After that, the temperature was kept fixed for 6 h. Finally, the samples were cooled at a rate of 6°C min^{-1} till the room temperature value was reached.

The samples were characterized by x-ray powder diffraction (XRD) using an X'Pert's Philips with CuK_α radiation ($\lambda = 1.5418 \text{ \AA}$) in the range $2^\circ \leq 2\theta \leq 70^\circ$. The variation of electrical resistivity with temperature was recorded using the standard four-probe technique in a closed cryogenic system using helium gas. The samples were cut into rectangular plates of dimensions about $1.5 \times 0.2 \times 0.3 \text{ cm}^3$. Four copper probes were attached to the samples using a conductive silver paint.

The transverse magnetoresistance and Hall voltage were measured using a five-probe technique. These measurements were carried out at different temperature values. A magnetic field having a normal

direction to that of the driving current was applied. The value of such field was up to 4.8 KG and was generated from an electro-magnet.

A Lakeshore ac susceptometer model series 7000 with helium cryostat was used to measure both real and imaginary parts of ac magnetic susceptibility, χ' and χ'' , respectively. These measurements were taken for different applied ac magnetic field values from 1 G up to 12 G and temperature range from 140 K down to liquid nitrogen temperature. The ac-magnetic susceptibilities were measured on powder samples of typical masses of 200 mg and a frequency of about 110 Hz.

RESULTS AND DISCUSSION

It is clear from Fig. 1, that as the Pr-content increases the lattice parameter a shows a slight elongation; on the contrary; for lattice parameter c which contracts as x increases till it reaches a value of 0.2. The elongation in a can be interpreted as a result of increasing the Cu-O bond strength more than normal. The contraction in c may be due to the replacement of Ca^{2+} ions (6-coordinate, octahedral $r_i = 114 \text{ \AA}$) with a smaller ionic radius Pr-ions (6-coordinate, octahedral $r_i = 113 \text{ \AA}$). Also, partial replacement of Ca^{2+} ions with Pr^{3+} ones increases the oxygen-contents and produces an increase in the average copper-oxidation state (Liu *et al.*, 1991). This leads to a smaller Cu-O distance within the CuO_2 -planes. Saturation in the lattice parameter c is observed for $x \geq 0.2$, indicating that the specimens solubility limit x_s of this substitution is about 0.2.

The variation of electrical resistivity with temperature for the superconducting specimens with $0 \leq x \leq 0.3$ is shown in Fig. 2a-c, respectively.

Electrical resistivity measurements within the limits $0 \leq x \leq 0.15$ reflect metallic behavior in temperature range from T_c up to 300 K. This behavior can be interpreted through applying non-Fermi liquid model. Also, it reflects the spin charge separation in CuO_2 -planes resulting in longitudinal transport relaxation rate $1/\tau \sim T$ (Anderson, 1991). For all metallic samples, a small curvature is clearly observed for temperature values above T_c . This is an indication for the superconducting thermodynamic fluctuations (Abou-Aly *et al.*, 2002). This curvature may be also explained as the opening of spin-gap that appears in the high-temperature superconductivity due to magnetic impurities substitutions (Koo and Cho, 2003). Figure 2b and c shows the relation between electrical resistivity and temperature for samples with x values ranging between 0.2 and 0.3. A reverse relationship between electrical resistivity and temperature (semiconductor-like behavior) is seen clearly on Fig. 2a and b.

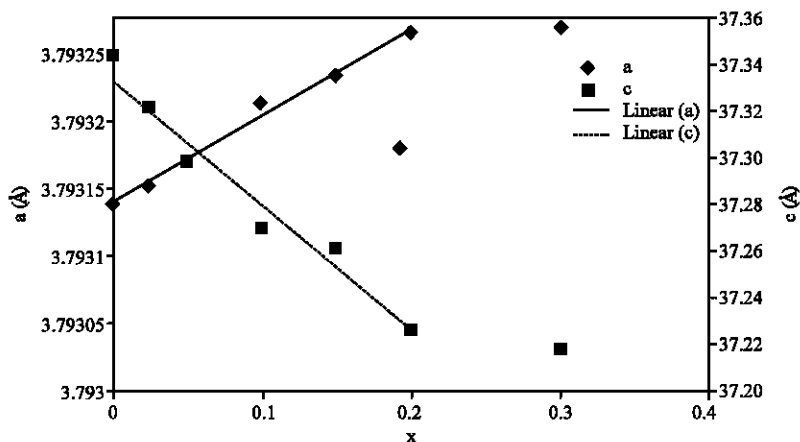


Fig. 1: Variation of lattice parameters a and c with Pr-content

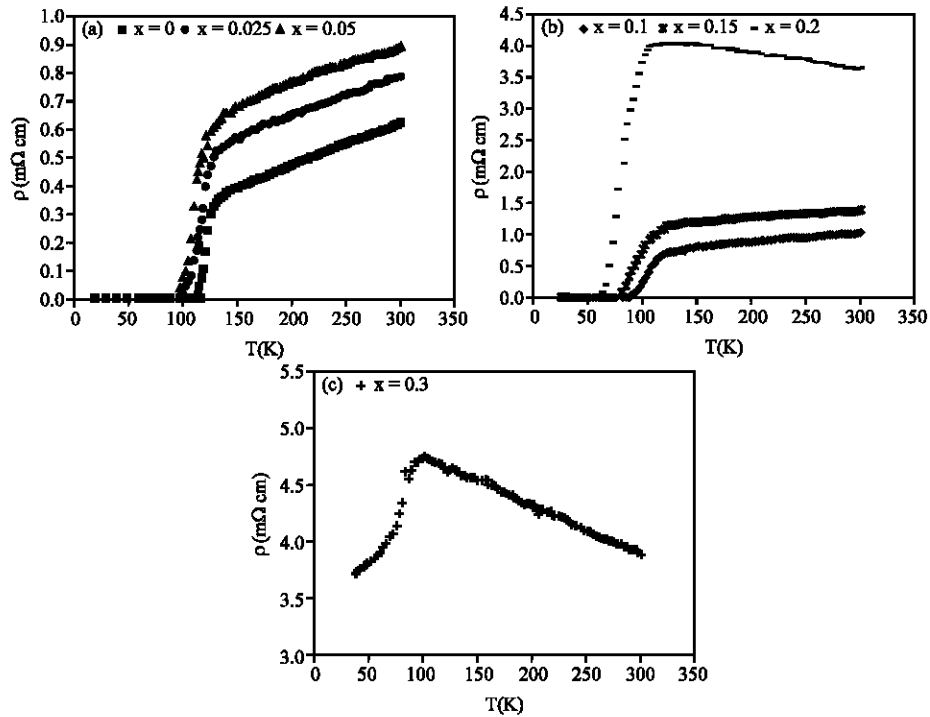


Fig. 2: Temperature dependence of electrical resistivity for $(\text{Tl}_{1.6}\text{Pb}_{0.4})\text{Ba}_2\text{Ca}_{2-x}\text{Pr}_x\text{Cu}_3\text{O}_{10.8}$ for different values of x , (a) $x = 0, 0.025$ and 0.05 , (b) $x = 0.1, 0.15$ and 0.2 and (c) $x = 0.3$

This proportionality was followed by a superconducting transition as the temperature was lowered. The zero-resistivity temperature was not reached for the sample with $x = 0.3$ till the temperature reaches 24 K. At this point, it contains multiphase, indicating the poor electrical contact between its grains. These results coincide with those obtained from the x-ray measurements. The semiconductor-like behavior is supported by a theoretical discussion based on Anderson impurity model (Eskes and Sawatzky, 1988). From this discussion it was concluded that the electronic states near the Fermi surface are inerrant for large Cu 3d and O 2p wave functions overlap. Also, these states become localized when this overlap is reduced.

In Fig. 3, most of $\chi'(T)$ curves exhibit a typical two drops behavior as expected in polycrystalline high-temperature superconductor samples. The first drop in χ' is usually correlated to the onset of the intragrain superconductivity coexisting with grain-boundary normal zones. While the second drop, appearing at lower temperature was due to the onset of intergrain superconductivity. Also, it is clear that the transition from normal state to superconducting state was very sharp for all samples, except at $x = 0.15$, indicating the high purity of these specimens. The transition width for such value of x is very broad which may be due to the higher contents of Pr-substitution reduce the number of pinning centers. This prevents the flux trapping in the bulk grains and causes a flux flow. Thus, no saturation in the intergranular field is attained and no plateau is obtained.

From the Table 1, it could be shown that the temperature values obtained using magnetic susceptibility measurements are lower than those determined from electrical resistivity ones. This is because any tiny part of material going on superconductive transformation loses its resistance and

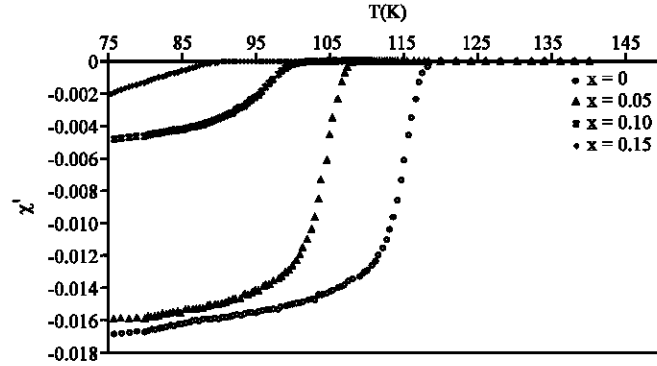


Fig. 3: Variation of real part of ac magnetic susceptibility with the temperature for $(\text{Tl}_{1.6}\text{Pb}_{0.4})\text{Ba}_2\text{Ca}_{2-x}\text{Pr}_x\text{Cu}_3\text{O}_{10.8}$ $x = 0, 0.05, 0.1$ and 0.15

Table 1: The superconducting transition temperature extracted from resistivity and ac magnetic susceptibility measurements versus praseodymium content for $(\text{Tl}_{1.6}\text{Pb}_{0.4})\text{Ba}_2\text{Ca}_{2-x}\text{Pr}_x\text{Cu}_3\text{O}_{10.8}$

X	$T_c(\text{K})$ resistivity	$T_c(\text{K})$ susceptibility
0	122.50 ± 0.61	119.3 ± 1.10
0.025	120.75 ± 0.60	116.5 ± 1.10
0.05	111.00 ± 0.60	107.8 ± 1.10
0.1	103.50 ± 0.55	99.8 ± 0.98
0.15	87.80 ± 0.51	85.5 ± 0.86
0.2	81.80 ± 0.40	78.4 ± 0.78

hence $R = 0$, when one or more continuous superconducting paths are in place between the measuring voltage electrodes. In contrast, diamagnetic measurements depend on macroscopic current loops to shield the magnetic field B from an appreciable fraction of the sample. This happens when full superconducting current paths become available. Therefore, filamentary paths can produce sharp pronounced drops in diamagnetism, which also require extensive regions of superconductivity. A suppression in T_c is observed as Pr-content increases and the rate of suppression is about $dT_c/dx = 2.04 \text{ K/at.}\%$. This suppression in T_c was also observed by Awad and Mohammed, (2004) for Tl-1223 substituted by Pr. Various mechanisms for interpreting T_c suppression in high-temperature superconductors have been proposed. These mechanisms include the Cooper pair breaking effect arising from magnetic impurities (Xiao *et al.*, 1990; Williams *et al.*, 1995) the strong potential scattering effect (Fehrenbacher, 1996) and the carriers localization effect (Xu *et al.*, 1997; Agarwal *et al.*, 1991). Since the change of T_c with the Pr-content is linear over the wide range of Pr-content, the Cooper pair breaking is one of the factors that suppress T_c . Also, the hole-filling mechanism plays a major role for T_c suppression.

Figure 4a and b display the variation of real part and imaginary part of ac magnetic susceptibility as a function of temperature for $(\text{Tl}_{1.6}\text{Pb}_{0.4})\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10.8}$. Curves of χ' data indicate that the superconducting transition temperature was not affected by increasing the applied ac magnetic fields although a small broadening was observed as the ac magnetic fields increase. Also, the Meissner volume diamagnetism shifted to a lower value as the applied ac magnetic field was increased. A minimum value was shown for χ'' at high-temperature values and maximum at peak temperature T_p that shifts to lower temperatures with increasing the external magnetic fields. Usually, the temperature dependence of χ'' for high-temperature superconductors contains two peaks; the first was correlated to the first drop in the real part of χ' and was interpreted as the intragrain superconductor contribution to the susceptibility. Meanwhile, the second one was related to the coupling matrix (intergrain superconductor). The second peak is more affected by the external magnetic field since the intergranular

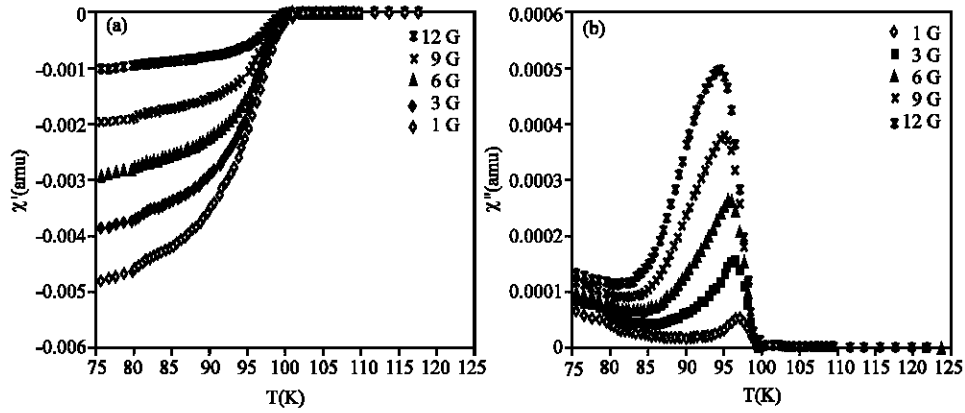


Fig. 4: Variation of (a) real part and (b) imaginary part of ac magnetic susceptibility as a function of temperature for $(\text{Tl}_{1.6}\text{Pb}_{0.4})\text{Ba}_2\text{Ca}_{1.9}\text{Pr}_{0.1}\text{Cu}_3\text{O}_{10.8}$.

pinning force is rather weak, unlike the intragrain peak that acquire a stronger pinning of vortices in the superconducting grains. The χ'' -T curves of our samples have only one peak, indicating that these samples have no or small separation between the intragrain and the dissipative intergrain transition. This means that the applied magnetic field is not sufficient to penetrate the intragrain superconductors, implying good electrical contacts between the superconducting grains. The maximum in χ'' occurs when the superconducting volume is penetrated by the applied magnetic field. That is, when shielding current is equal to the critical current density of this volume. In this case the flux pinning is the only mechanism controlling the flux dynamics. When the applied field just reached the center of the sample, the critical current density J_c at the temperature (T_p) corresponding to the maximum value of χ'' is related to the full penetration field B_a according to the equation (Bean, 1964):

$$J_c = \frac{B_a}{\mu_0 R} \quad (1)$$

where, R is the average radius of grain estimated from SEM measurements.

The dependence of J_c on $(1 - \frac{T_p}{T_c})$ is shown in Fig. 5 for $(\text{Tl}_{1.6}\text{Pb}_{0.4})\text{Ba}_2\text{Ca}_{2-x}\text{Pr}_x\text{Cu}_3\text{O}_{10.8}$ with $x = 0, 0.05$ and 0.1 . The critical current density data are well fitted according to the empirical scaling relation (Pavard *et al.*, 1999):

$$J_c(T) = J_c(0) \left(1 - \frac{T_p}{T_c}\right)^\gamma \quad (2)$$

where, $J_c(0)$ is the critical current density at 0 K and γ is the critical exponent.

The values of $J_c(0)$ and γ obtained from the best fitting according to Eq. 2 are shown in Table 2. Data, in Table 2, shows that the values of γ are around 2. These values are in consistence with other superconductors (Ravi, 2000). However, they are quiet different from what is expected ($\gamma = 1.5$) from Ginzburg-landau theory for $J_c(T)$ of an infinite slab near T_c (Lee *et al.*, 1995). The value of $\gamma = 2$ indicates that the intergrain junctions are SNS type (Chu and McHenry, 2000). Moreover, the critical current density at 0 K enhances till $x = 0.05$ and then it suppresses for $x > 0.05$. This enhancement in $J_c(0)$, at lower Pr-contents, may be due to the lattice defects produced from the partial substitutions

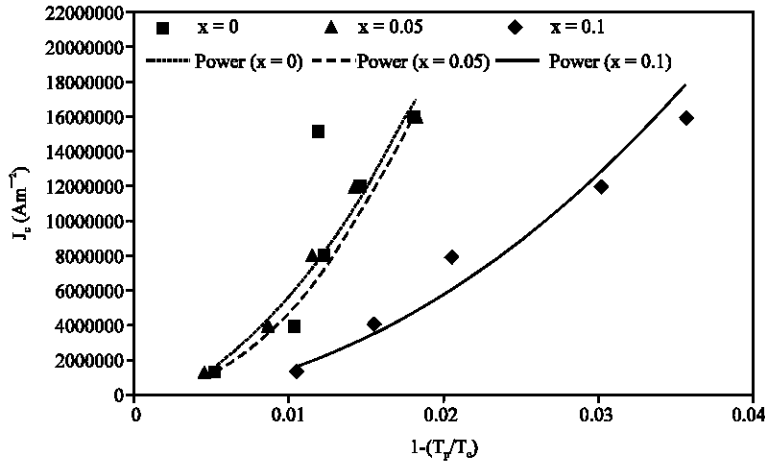


Fig. 5: The dependences of J_c on for $(\text{Tl}_{1.6}\text{Pb}_{0.4})\text{Ba}_2\text{Ca}_{2-x}\text{Pr}_x\text{Cu}_3\text{O}_{10-\delta}$ with $x = 0, 0.05$ and 0.1

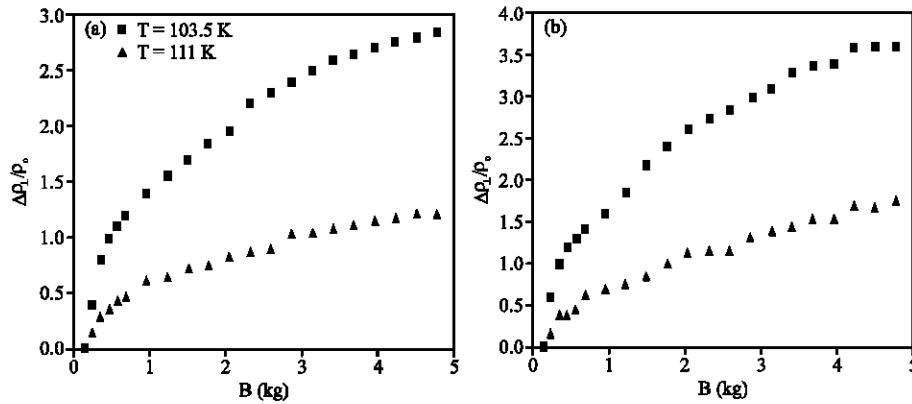


Fig. 6: The variation of the transverse magnetoresistance $\Delta\rho_{\perp}/\rho_0$ with the external magnetic fields for $(\text{Tl}_{1.6}\text{Pb}_{0.4})\text{Ba}_2\text{Ca}_{1.975}\text{Pr}_{0.025}\text{Cu}_3\text{O}_{10-\delta}$ for (a) $x = 0.0$ and (b) $x = 0.025$

Table 2: The variation of $J_c(0)$ and γ with Pr-content

X	$J_c(0) \times 10^6 \text{ A cm}^{-2}$	γ
0	3.0	2.05
0.025	4.5	1.94
0.05	6.0	1.87
0.1	1.0	1.96
0.15	0.6	1.89

of Ca^{2+} by Pr^{3+} ions that enhances the flux pinning. The suppression in $J_c(0)$, at higher Pr-contents, is attributed to the increase of the grain boundaries resistance and the reduction of the flux pinning inside the samples. Also, the formation of secondary phases plays an important role for suppressing J_c . The results of the transverse magnetoresistance $\Delta\rho_{\perp}/\rho_0$ as a function of the external magnetic fields, measured at different temperature values below the superconducting transition temperature, for $(\text{Tl}_{1.6}\text{Pb}_{0.4})\text{Ba}_2\text{Ca}_{2-x}\text{Pr}_x\text{Cu}_3\text{O}_{10-\delta}$ with $x = 0$ and 0.025 are shown in Fig. 6a and b, respectively.

The transverse magnetoresistance was produced when the transport current was flown in the presence of an applied magnetic field (Fig. 6a, b). In this situation, the vortices arising from the applied field were involved with the current. Vortex motion and heat dissipation were evolved due to such

interaction generating a resistive term called flux-flow resistance. This was a type of magnetoresistance due which an achievable value for the critical current in many samples was limited. A rapid increase was shown in the magnetoresistance values as the magnetic field was raised up to $B \approx 1.1$ kG. Then, this increase was slowed down (nearly plateau) when a value of 4.4 kG was reached by the magnetic field. This behavior might be due to an increase in the applied magnetic field subjected to the intergranular spaces. This increase lead to a destruction in weakly linked networks. The grain bulk was penetrated by magnetic flux of higher magnetic field values. This flux was trapped in there leaving the intergranular field almost unchanged. A plateau representing the variation of $\Delta\rho_{\perp}/\rho_0$ with B was obtained in this case. The $\Delta\rho_{\perp}/\rho_0$ data decreased as the temperature reached the value of T_c . From these results it was concluded that only a small fraction of the carriers were superconductor near T_c . At lower temperatures more carriers become superconducting, so, the change between the zero-field and the applied field resistivity is greater which hints that the vortex pinning may be unaffected (Gomaa, 1999). Another explanation of this behavior was due to the absence of flux trapped near T_c temperature.

It is clear from Fig. 7, Hall coefficient decreases as the temperature increases. This behavior was quite similar to that obtained in most of high-temperature superconductors (Roa-Rojas *et al.*, 2001). All previous results showed that the Hall coefficient had an inverse proportionality with temperature. Ironically, this was in contrast with the expectations for non-magnetic metal. This anomalous behavior, known as Curie paramagnetic-behavior, was discussed according to skew scattering of carriers by spin fluctuations (Fiory and Grader, 1988) and non Fermi-liquid model (Anderson, 1991). Also, the value of the Hall coefficient increased as Pr-content was increased. This behavior means that the hole-concentrations decreased as the Pr-content were increased. This could be attributed to the partial replacement of Ca^{2+} ions by Pr^{3+} ions enough to reduce the number of holes in CuO_2 -planes through the hole-filling mechanism. The Hall coefficients for all samples were positive, indicating the kind of carriers are holes and our prepared samples are far and away from highly over-doped regime.

From Fig. 8, an inverse proportionality was noticed between Hall mobility and the temperature. This is a usual property in the metallic behavior, where, $\mu_H \propto T^{-2}$ because $\cot \Theta_H = 1/(\mu_H B)$.

From Fig. 9, a direct proportionality was found between the Hall angle and the temperature. This behavior was supported by the non-Fermi liquid model that provided a neutral explanation for

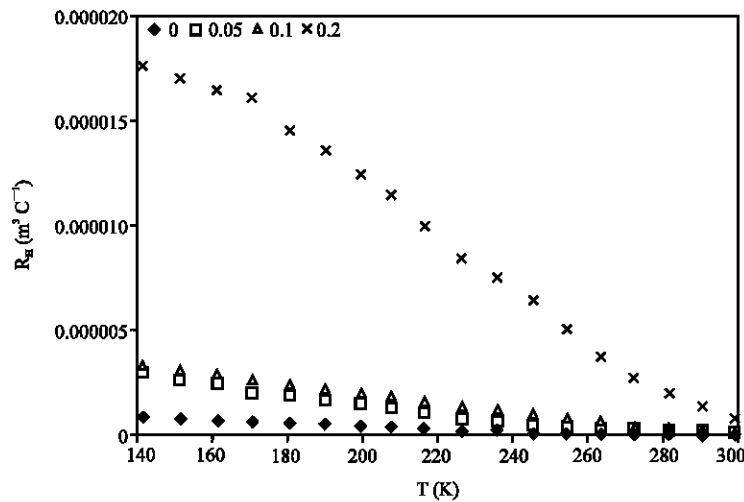


Fig. 7: The variation of the Hall coefficient with the temperatures for $(\text{Tl}_{1.6}\text{Pb}_{0.4})\text{Ba}_2\text{Ca}_{2-x}\text{Pr}_x\text{Cu}_3\text{O}_{10.8}$

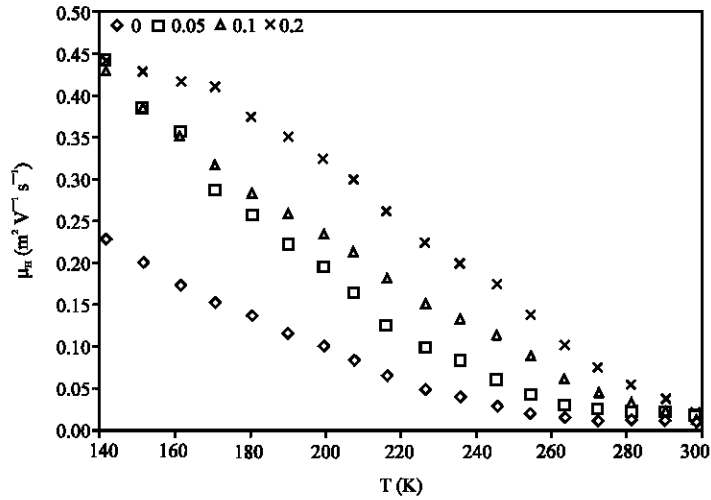


Fig. 8: The variation of the Hall mobility with the temperatures for $(\text{Tl}_{1.6}\text{Pb}_{0.4})\text{Ba}_2\text{Ca}_{2-x}\text{Pr}_x\text{Cu}_3\text{O}_{10.8}$

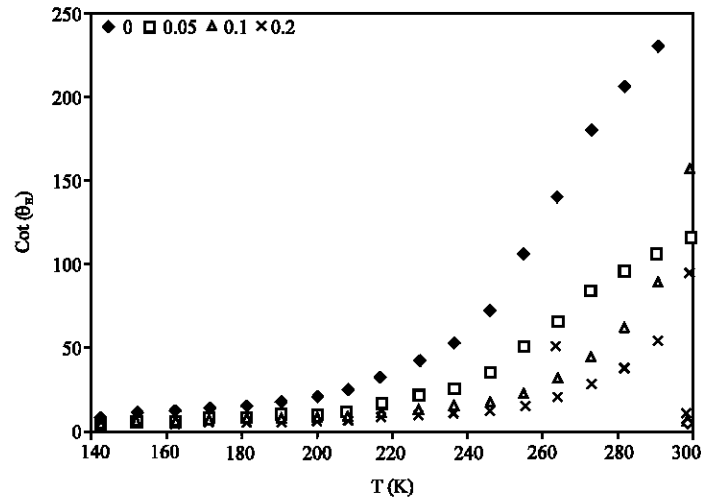


Fig. 9: The variation of the Hall angle with the temperatures for $(\text{Tl}_{1.6}\text{Pb}_{0.4})\text{Ba}_2\text{Ca}_{2-x}\text{Pr}_x\text{Cu}_3\text{O}_{10.8}$

anomalous features of transport data in several high-temperature superconductors (Anderson, 1991). Two different rates were due to the spin charge separation in CuO_2 -planes. The first one is longitudinal (the transport relaxation rate $1/\tau \sim T$), whereas the second is transverse (Hall relaxation rate $1/\tau_H \sim T^2$). The linear resistivity was presented by the first rate, whereas the Hall angle dependence $\text{Cot } \Theta_H \sim T^2$ or $T^2 \sim 1/\tau_H$ was presented by the second one. The values of n_H and μ_H for all samples, were calculated at the room temperature where $n = 10^{26} \text{ m}^{-3}$ and $\mu = 0.05 \text{ m}^2 \text{ V s}^{-1}$ so, it was assured that they lied within the band of normal metal ($n = 10^{28} \text{ m}^{-3}$ and $\mu = 0.0025 \text{ m}^2 \text{ V s}^{-1}$) and semiconductor material one ($n = 10^{22} \text{ m}^{-3}$ and $\mu = 100 \text{ m}^2 \text{ V s}^{-1}$).

CONCLUSION

Series of superconducting samples of type $(\text{Tl}_{1.6}\text{Pb}_{0.4})\text{Ba}_2\text{Ca}_{2-x}\text{Pr}_x\text{Cu}_3\text{O}_{10.8}$ have been successfully prepared via a solid-state reaction technique. The partial replacement of Ca^{2+} ions by Pr^{3+} ions did not

influence the tetragonal unit cell structure of (Tl, Pb)-2223. The solubility limit of this replacement was found to be around $x = 0.2$. The superconducting transition temperature was suppressed as Pr-content increased and this suppression was attributed to both Cooper pair breaking and hole-filling mechanisms. There was no separation between the intragrain and the dissipative intergrain transition in the ac magnetic susceptibility, implying good electrical contacts between the superconducting grains. The critical current density was increased as the Pr-content had done till $x = 0.05$ and then it decreased with further increase in x . Data of transverse magnetoresistance, below the superconducting transition temperature, showed a decrease in their values as the temperature reached the superconducting transition temperature. The Hall coefficient decreased as the temperature increased and this anomalous behavior was discussed according to skew scattering of carriers by spin fluctuations and a non Fermi-liquid model. The hole concentrations and mobility values at room temperature for these samples were between the values of normal metals and semiconductors.

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