Synthesis of $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$ superconductors with partial substitution of Tl by Hg

Physics

KEYWORDS : TBCCO System, Superconductor, microstructure properties,Tl-2223 superconducting

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ABSTRACT

Bulk polycrystalline samples of high temperature superconductor of the type 2223 have been prepared by a two–step solid state reaction method. Substituting Hg with x=0.4 for $Tl_{2-x}Hg_xBa_2Ca_2Cu_3O_{10+\delta}$ give the best values of Tc =133K. The x-ray data of all the superconducting samples showed a tetragonal structure with a high ratio of Tl-2223 superconducting phase.

Introduction

The discovery of superconductivity in ceramic marked the beginning of high temperature superconductor (HTSc).

One of the high temperature superconducting cuprates (HTSc) which have an inherent potential for practical application, especially at temperatures close to 120 K, is the Tl bilayer phase $Tl_2Ba_2Ca_2Cu_3O_{10}$ (TBCCO) in the form Tl-2223. In bulk form, this superconducting cuprate has a transition temperature (T_c) as high as 125K and in thin film form, it has been found to be superconducting up to 122K[25]..Doping with various elements has been found to be useful and effective in improving its properties[3].

Two superconducting phases, $Tl_2Ca_2Ba_2Cu_3O_{10+\delta}$ and $Tl_2Ca_1Ba_2Cu_2O_{8+\delta}$ were identified in their samples by Hazan et al[4]. They found that these phases have a pseudo tetragonal until cell (5.40×5.40×36.25) Å , correspond to a, b and c lattice dimensions, respectively.

Dou et [5]studied superconducting properties of Tl-Ba-Ca-Cu-O ceramics prepared by a solid state reaction under optimum conditions (880°C sintering in flowing O_2 for 3h). The resistivity, AC susceptibility, and Meissner effect were measured. The temperature-dependent resistivity of $Tl_2Ba_2Ca_2Cu_3O_{10+y}$ showed a degradation in $T_c$after increasing the sintering time from 3h to 6h and the temperature dependent AC susceptibility showed that at zero applied field the superconducting transition is reasonably sharp with T_c=110K.

Partial substitution of Tl by Pb and Ca by Pr in $TlSr_2CaCu_2$ oxide system has been studied by Liu et al (1989)[39]. The x-ray diffraction analysis showed that the samples have a single phase, tetragonal structure with a=3.82 and c=12.00 . **A** T_c(zero) was found to be 78,96,106,98,45 and 25K for $Tl_{0.5}Ca_{1-x}Pr_xSr_2Cu_2$ oxide sample with x=0,0.1,0.2,0.5 and 0.7, respectively and for x=1, the sample exhibited a semiconductor behavior.

Agrawaland Narlikar (1994)[61] investigated the conductivity variation of TBCCO i.e. $Tl_2Ba_2Ca_2Cu_{3-x}A_xO_y$ compound with substitution for (A=Fe,Zn) of Cu ions The substitutional compensation of Zn and Fe in place of Cu-site at different concentrations (x=0-0.5) showed that T_c decreases slowly with increasing substitution percentage of both Zn and Fe.

Single crystals of TBCCO (Tl-2212) have been grown from a stoichiometric mixture of Tl_2O_3 and a precursor $Ba_2CaCu_2O_x$ prepared by Chowdharyet al (2002)[9]. They observed that the transition temperature T_c=105K and the transition width ΔT_c was around 5K, as determined from the temperature-dependent magnetization (MT) measurements, at a field of 10G.

Kareem and Tariq[10] have investigated the effect of simultaneous substitution of strontium at the barium site of $Tl0.6Pb_{0.4}Ba_{2-x}Sr_xCa_2Cu_3O_{9-\delta}$ Superconductors and found that $T_{c\,(off)}$= 113K for $Tl_{0.6}Pb_{0.4}Ba_{1.5}Sr_{0.5}Ca_2Cu_3O_{9-\delta}$. Khan et al[11] have studied the enhanced Inter-grain Connectivity in $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_3O_{10-\delta}$ Superconductors.

Masoon et al [12] investigated the effect of sintering temperature and timeon the transition temperature for $Tl_2Ba_2Ca_2Cu_3O_{10}$ and $Tl_2Sr_2Ca_2Cu_3O_{10+\delta}$ systems type 2223 and studied the effect of these parameters on the structural and electrical resistivity of both prepared systems. They found that for Tl-2223 system, the T_c increased and decreased with increasing sintering temperature and time respectively.

Discoveries of Tl-based system have not only set new T_c records with zero resistance up to 125K, but also have provided a new insight into the mechanism of high-T_c oxide superconductivity[1].

In this paper we investigated the effect of Bi doping on the electrical and structural properties of $Tl_2Ba_2Ca_2Cu_3O_{10}$system type 2223..

Experimental

$Tl_{2-x}Hg_xBa_2Ca_2Cu_3O_{10+\delta}$ with (0≤x≤1) were prepared by a two step method as follows: precursors $Ba_2Ca_2Cu_3O_{10+\delta}$ was first prepared using high purity powders of $BaCO_3$, CaO and CuO as starting materials. Then, Tl_2O_3 was added to the mixture and grinding them in agate mortar for about 30 min to obtain a very fine and optimum homogenous powder. The mixtures were pressed into a pellet of (0.2-0.3)cm in thickness and 1.3cm in diameter, under a pressure of about 3 ton/cm^2 . The samples were sintered in air atmosphere of 860°C for 3h.

The resistivity measurements were carried out by the four probe method with 30mA current .The structure of the prepared samples was obtained by using x- ray diffractometer (XRD) type Philips having the following features (source : $Cu_{K\alpha}$,voltage : 40kv , current : 20 mA, wavelength : 1.5405 Å) . Magnetization data have been taken in an automatic magnetometer (VSM 3001). For magnetization measurements, 50 Oe magnetic field has been applied

Results and discussion

We have investigated the effect of Hg doping in the Tl-2223 superconductor by preparing a series of samples (at 860ºC for 3h) with complete stoichiomerty $Tl_{2-x}Hg_xBa_2Ca_2Cu_3O_{10+\delta}$ with x ranging from 0 to 1.

The superconducting properties of the samples have been examined by electrical measurements and DC magnetization measurements. Fig.(4.7) shows the temperature dependence of the electrical resistivity for a $Tl_{2-x}Hg_xBa_2Ca_2Cu_3O_{10+\delta}$ sample with 0≤x≤1.

However, a point of interest is that there are obvious phase fluctuation as well as in T_c in these data and as we will see in all the coming results, this we believe the main characteristics of cuprateHTSc at least of the types we are studying. The reason, in our openion is that the variation in the Cu-O bonds and c-parameter are due here to the variation of δ. The later lead to variation in CuO_2 layer thickness .Obviously here for all the doping range the fluctuation in T_c is within the HTSc regime. The highest transport mechanism is at x=0.4. This means higher resonant tunneling occurs between CuO2 layers through localized centers across the c-parameter.

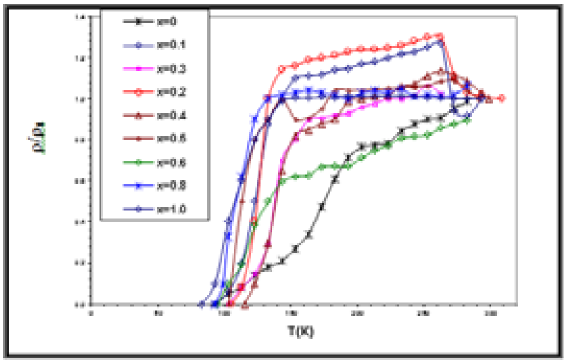


Fig.(4.7): Temperature dependence of normalizedresistivity for $Tl_{2-x}Hg_xBa_2Ca_2Cu_3O_{10+\delta}$ samplesfor different Hg content

With x=0 shows a behavior typical of undoped Tl-2223:a critical transition temperature (T_c) at 120K and zero resistance at (T_{coff})90K. The Hg doped samples have a higher resistivity than the undoped samples. The sample with x= 0.4 shows a higher transition temperature (T_c=133K), while the sample with x=0.5 display a T_c at 115K. Fig.(4.8) shows the variation of T_c'swithHg doped Tl-2223 versusHg concentration.The data indicate a maximum T_c near x=0.4 and for x≥0.5, T_ca gainfor higher percentage $T_c$decreases (see Table 4.2).

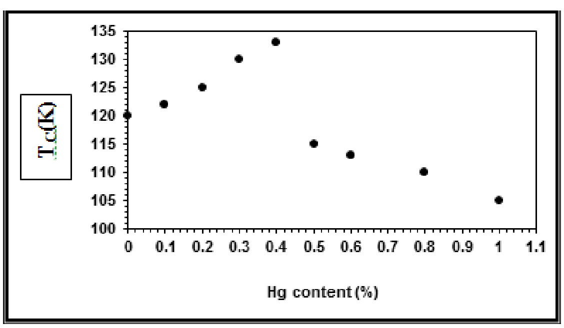
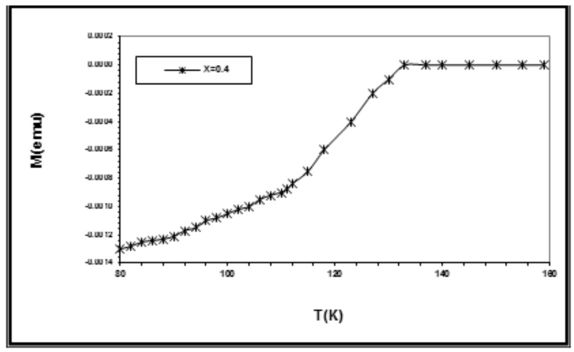


Fig (4.8): Transition temperature of$Tl_{2-x}Hg_xBa_2Ca_2Cu_3O_{10+\delta}$ samples as a function of Hg content

We now briefly discuss the stabilization of the Tl-2223 by a moderate Hg doping.Pure Tl-2223 with ideal stoichiometery would have all the copper atoms in the 2+ state,making it an insulatorat room temperature.The higher Cu valence state ,which is necessary for a cuprate to be superconducting, is created via the substitution of $Tl^{3+}$by Hg^{2+}leavingCa^{3+}vacancies at cation sites,and the internal redox reaction is $Tl^{3+}+Cu^{2+}=Tl^{(3-\delta)+}+Cu^{(2+\delta)+}$ [53]. These complex internal mechanisms are difficult to control during synthesis. Partialsubstitution of Tl^{3+} with Hg^{2+} in $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$ produces a stable Tl-2223 phase with the highest possible T_c and increases the effective Cu valence and brings the system from an under doped regime to an optimally doped regime and thus promotes the formation of the 2223 phase.

As the Hg concentration increases further (beyond x=0.5), the system goes from the optimally doped regime to an over doped regime, resulting in the observed decrease of T_c. Similar results have been reported by Jiaand Zettl[53]for$Tl_{2-x}Hg_xBa_2Ca_2Cu_3O_{10+\delta}$ system, prepared under vacuum in quartz tube and the quartz ampoules were sintered at 830°C to 870°C and slow cooled to room temperature.

Fig.(4.9) shows the temperature dependence of low field magnetization (50O_e) for a $Tl_{1.6}Hg_{0.4}Ba_2Ca_2Cu_3O_{10+\delta}$ sample.For these measurements, the bulk samples have been finely powdered in order to avoid problems due to intergrain coupling. As Fig.(4.9) shows, the magnetically determined critical transition temperature (T_c) is ~133K.This result is consistent with the electrical resistivity measurements.The interaction of the samples with magnetic field under superconducting state reveals that the state of the art of the superconductors, inthat they allow normal and superconductor states to coexist under the magnetic field and they process separately and keep their properties a part.



Fig(4.9):Variation in magnetization with temperature for $Tl_{2-x}Hg_xBa_2Ca_2Cu_3O_{10+\delta}$system

The merit of the above case is that the superconductor electrons protect themselves from the magnetic field through certain arrangements, most probably in an orthogonal structure that leads to pairing in the lowest energy state, and create a polarization around and consequently yielding a narrow energy gap.

Through such an energy gap and the orthogonal structure, we expect the transport of the super electrons, in the pairing state-will be three dimensional, firstly in Cu-O plane and secondly tunneling through the narrow energy gap by excitation through Tl-O plane or its substitute.

A point of interest in the magnetization measurements is its reliability over the electrical one it is simpler and direct ,we mentioned this because it reflected clearly the type II and consequently a second phase transformation.

The powder x-ray diffraction patterns of these samples are consistent with a 2223-type structure. Figs.(4.10,11)show the x-ray patterns of Hg doped Tl-2223 with x=0.2,0.4,0.6,0.8,1.0. All the major peaks correspond to the (Tl/Hg)-2223 phase[92], the minor impurity phases detected include $BaCuO_2$ and Ca_2CuO_3[53]. Also we have seen an improvement in the structure properties and thehigher rate of phase (Tl/Hg-2223) that appears with increasing Hg contentup to x=0.4. At this ratio, the oxygen factor(δ) gets a maximum value see Table(4.2). The increase of Hg content (x>0.5) leads to the emergence of another peaks,which is corresponded to some unknown impurity and the crystallinity becomes less, which may be attributed to the substitution of Hgcause more cuprate vacancies that the HTSc need, to a high scattering effect of supper electrons in crystalline structure.

Such results may also be due to the difference between the ionic radii for both of Hg and Tl, where the ionic radii Hg^{2+}(1.11Å) is longer than of Tl^{3+}(0.95Å) which render c-parameter to be longer or to get deformeddrastically. The lattice parameters of Hg doped Tl-2223 are reported in Table (4.2), which shows that as the Hg doping level increases,the oxygen content (δ) decreases giving the evidence of the serious role of oxygen in the superconducting mechanism.Althoughthe "a"lattice parameter changes little, but the "c" lattice parameter slightly increases with increasing Hg concentration (see Fig.4.12). Such tiny changes may not have a significant role in the transport processes as compared with the role of (δ).This point is another factor in the transport mechanism model of our work.

For the x=1 sample, a secondary superconducting phase, Tl-1223, has also been detected in addition to the above mentioned impurity phases.

Table (4.2) Variation in T_c values, lattice parameters and oxygen content for different compositions of$Tl_{2-x}Hg_xBa_2Ca_2Cu_3O_{10+\delta}$sintered at 860°C for 3 hours.

x	a (Å)	c (Å)	T_C (K)	δ
0.2	3.846	35.740	125	0.293
0.4	3.843	35.754	133	0.322
0.6	3.841	35.816	113	0.212
0.8	3.840	35.868	110	0.197
1	3.838	35.872	105	-0.129

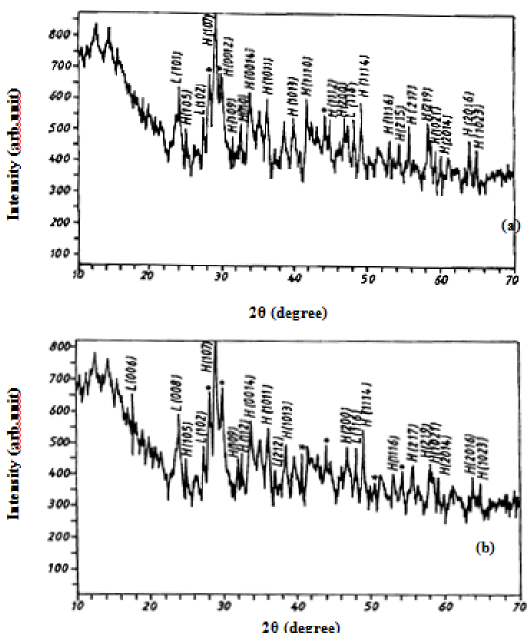


Fig.(4.10): X-ray diffraction patterns for the Tl2-xHgxBa2Ca2Cu3O10+δsamples (a)x=0.2, (b)x=0.4. H-HighTc phase, L-low Tc phase, .-impurity phaseBaCuO2, #-impurity phaseCa2CuO3 , *-unknown impurity.

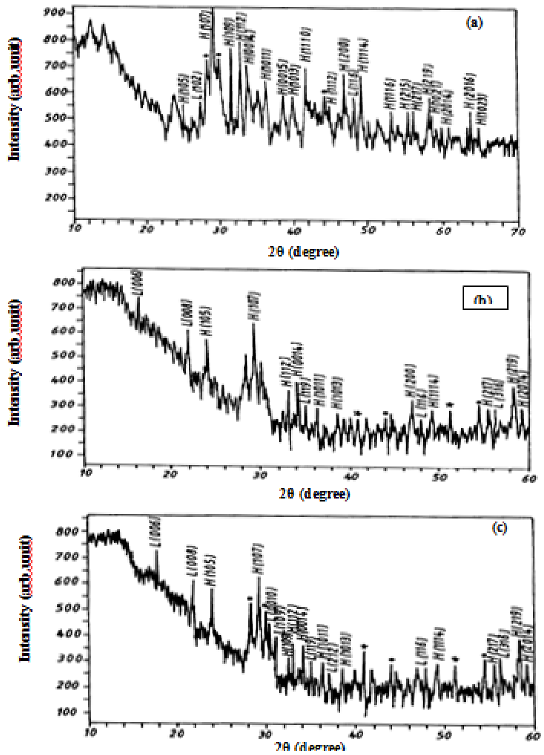
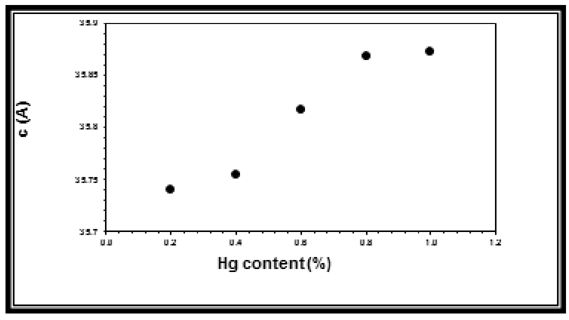


Fig.(4.11): X-ray diffraction patterns for the $Tl_{2-x}Hg_xBa_2Ca_2Cu_3O_{10+\delta}$samples (a)x=0.6, (b)x=0.8, (c)x=1.0 . H-HighT_c phase, L-low T_c phase, .-impurity phase$BaCuO_2$,#-impurity phasend Ca_2CuO_3 , *-unknown impurity.



2Θ (degree)

Fig.(4.12): Variation of the c-axis lattice constant with Hg content for $Tl_{2-x}Hg_xBa_2Ca_2Cu_3O_{10+\delta}$ system.

Conclusions:

The outcome foundation of above data is the following:

- The substitution of Hg in Tl for the compounds $Tl_{2-x}Hg_xBa_2Ca_2Cu_3O_{10+\delta}$ has exhibited a maximum value of T_c(133K) at x=0.4.
- A partial substitution of Tl by Hg was found to promote the formation and grain growth of Tl-2223 phase and then enhance the T_c value.
- The magnetization examination confirmed the electrical investigation and showed a second order transition as an evidence that our 2223 HTSc is of type II.
- XRD pattern analyses have shown a tetragonal structure, and there were at least two superconducting phases.

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