Effect of Hg on superconducting and microstructure properties of $Tl_{2-x}Hg_xSr_2Ca_2Cu_3O_{10+\delta}$ systems

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Abstract

Bulk polycrystalline samples have been prepared by the two-step solid state reaction process. It has been observed that as grown Tl2-xHgxSr2Ca2Cu3O10+δ (with x = 0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.8, 1) corresponds to the 2223 phase. It has been found that Tc varies with Hg content .The optimum Tc is about 120K for the composition Tl1.6Hg0.4Sr2Ca2Cu3O10+δ.The microstructure for Tl1.6Hg0.4Sr2Ca2Cu3O10+δ observed to be most dense and this phase exhibits the highest stability.

Keywords
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تأثير الزئبق على خصائص التوصيلية الفائقة و الخصائص التركيبية للنظام $Tl_{2-x}Hg_xSr_2Ca_2Cu_3O_{10+\delta}$

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الخلاصة:

حضرت الماذج المتعددة التبلور بطريقة تفاعل الحالة الصلبة وعلى مرحلتين. لوحظ نمو الطور 2223 في المركب $Tl_{2-x}Hg_xSr_2Ca_2Cu_3O_{10+\delta}$ولقيم (x=0,0.1,0.2,0.3,0.4,0.5,0.6,0.8,1) . ووجد بأن T_c تتغير بتغيرمحتوى ال Hg وان اعلى T_c حصلنا عليها كانت بحدود 120K العائد للمركب $Tl_{1.6}Hg_{0.4}Sr_2Ca_2Cu_3O_{10+\delta}$. بينت فحوصات البنية التركيبية بأن المركب $Tl_{1.6}Hg_{0.4}Sr_2Ca_2Cu_3O_{10+\delta}$ هو الاكثر كثافة والاعلى استقرارية.

Introduction

Soon after the discovery of the Bi-Sr-Ca-Cu-O system, Sheng et al[1,2] have reported a superconducting transition in a Tl-Ca-Ba-Cu oxide composite with onset temperature up to 120K and zero resistance below 107K. A number of studies has been made to prepare new compounds in the Tl system and to understand the relationship between their superconducting properties and crystal structure.

Dou et al [3] studied superconducting properties of Tl-Ba-Ca-Cu-O ceramics prepared by a solid state reaction. The temperature dependent AC susceptibility showed that at zero applied field the superconducting transition is reasonably sharp with T_c=110K. Zhenhang et al [4] investigated two superconducting phases with T_c at 120 and 90K for Tl-Ba-Ca-Cu oxide system. Scanning electron microscopy shows that the grains are well crystalline and uniformly distributed.

Tl-based in the form of $Tl_2Sr_2Ca_2Cu_3O_{10+\delta}$ which has a quite high critical temperature (T_c=120K) doping with various elements has been found to be useful and effective in improving its properties[5,6].

Partial substitution of Tl by Pb and Ca by Pr in $TlSr_2CaCu_2$ oxide system has been studied by Liu et al[7]. The x-ray diffraction analysis showed that the samples have a single phase, tetragonal structure with a=3.82 and c=12.00A. 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. Jia et al[8,9] reported on the effect of substitution of Hg at the Tl sites in the oxygen deficient TlO_δ layer of $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$ cuprate superconductor. They prepared the samples by the two-step reaction process and they found that partial substitution of $Tl^{3+}$with Hg^{2+}in $Tl_2Ba_2Ca_2Cu_3O_{10}$ produces a stable Tl-2223 phase with the highest possible T_c and claimed that at room temperature the Hg-doped samples have a higher resistivity than the undoped specimen.

In this paper, the effect of Hg on the formation of the high-T_c phase in the $Tl_2Sr_2Ca_2Cu_3O_{10+\delta}$ system was investigated.

Experimental

Samples with nominal compositions $Tl_{2-x}Hg_xSr_2Ca_2Cu_3O_{10+\delta}$ were synthesized by a two-step solid stat reaction method .precursors $Sr_2Ca_2Cu_3O_{10+\delta}$ were first prepared using high purity powders of $Sr(NO_3)_2$,CaO and CuO as starting materials. Then, Tl_2O_3 and Hg_2O were 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 measurement were carried out by the four probe method and the formation of the 2223-phase was systematically checked by X-ray diffraction technique using Phillips diffractometer with source $Cu_{K\alpha}$ radiation. The microstructure of the samples was observed using a scanning electron microscopy (SEM) (JSM-5600).

Results and discussion

The resistivity versus temperature for samples with nominal compositions $Tl_{2-x}Hg_xSr_2Ca_2Cu_3O_{10+\delta}$ are shown in Fig.(1). It is found from this figure that the substitution of 0.1,0.2,0.3 and 0.4 Hg content to the composition raise the transition temperature T_c and more addition (x=0.5) decrease the critical temperature (as observed in Fig. 2 and Table 1), and when x≥0.6 our apparatus could not help us to obtain the value of $T_{c(off)}$ because it is less than the liquid nitrogen temperature.

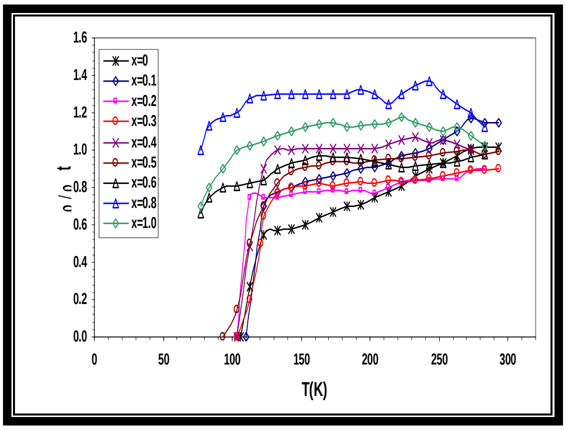


Fig.(1): Temperature dependence of normalized resistivity for $Tl_{2-x}Hg_xSr_2Ca_2Cu_3O_{10+\delta}$ samples for different Hg content

This result indicates that the Hg has catalytic effect on the reaction to form the high-T_c phase (HTP) within the range x=0.1-0.4. However, certain amount of Hg is necessary for the occurrence of this reaction, while excessive Hg substitution promotes another reaction, which is likely to assist the formation of low-T_c phase instead of the high-T_c phase[10,11].

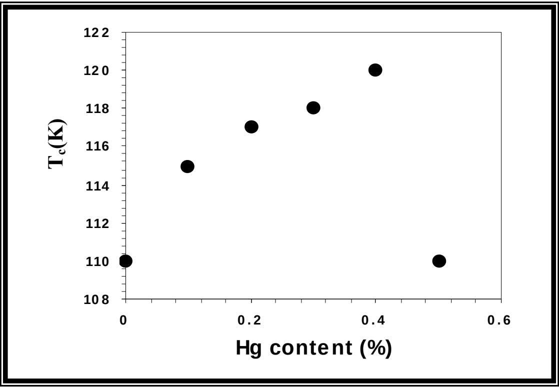
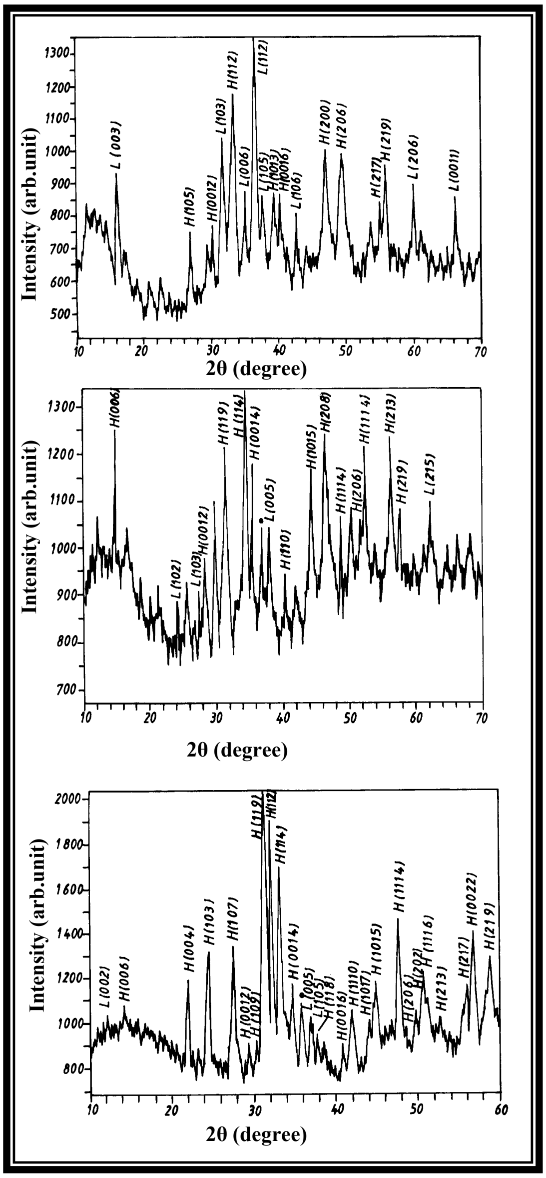


Fig.(2): Transition temperature (T_c) of $Tl_{2-x}Hg_xSr_2Ca_2Cu_3O_{10+\delta}$ samples as a function of Hg content.

In order to explain such a high T_c in these materials, various explanations have been put forward; firstly, the substitute may lead to a change in the carrier density of states[12], indicating various magnetic moments, secondly the c-axis is slightly elongates, and the Cu-O flat layers are still not buckled, hence, ordering of super electrons coupling, is enhanced in the materials, due to even a tiny inhomogeneity.

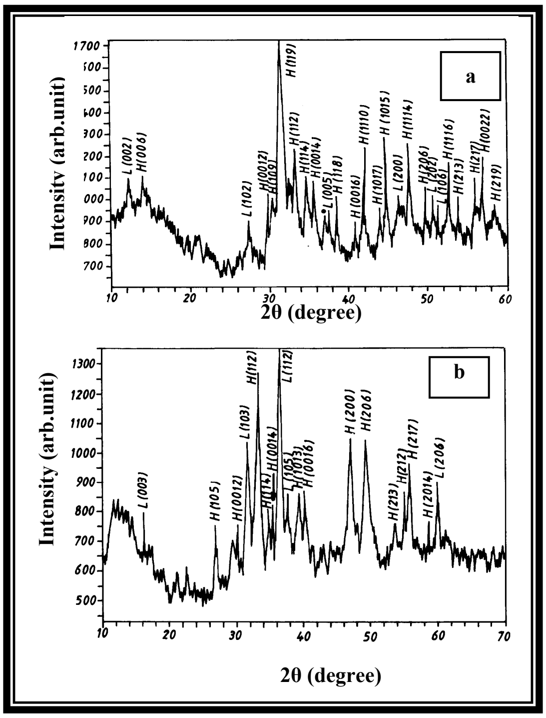
The x-ray diffraction pattern for the Hg-doped samples have exhibited a decrease of the low-T_c phase and an enhancement of the peaks due to the high-T_c phase (Figs.3 and 4). However, the above results suggest that the growth of the high-T_c phase is promoted by Hg-doping and for the x≥0.4 samples a secondary superconducting phase Tl-1223 was detected.

Table (1) and Fig.(5) show an increase of the c-axis lattice constant for Hg-doped samples as compared with the undoped samples , the reason is due to the substitution of Hg for Tl where the ionic radii of Hg^{2+}(1.11Å) is longer than that of Tl^{3+}(0.95Å) as mentioned previously which renders c-parameter to be longer or get deformed .Indeed, the deformation in the c-axis as a result of substitution or deficiency of some atoms ,adjusts the amount of charge transfer from Tl layer to Cu layer. This will be a driving force to the pairing generation of superconductor holes forming bosons[13] which are the current carriers in our superconductor.

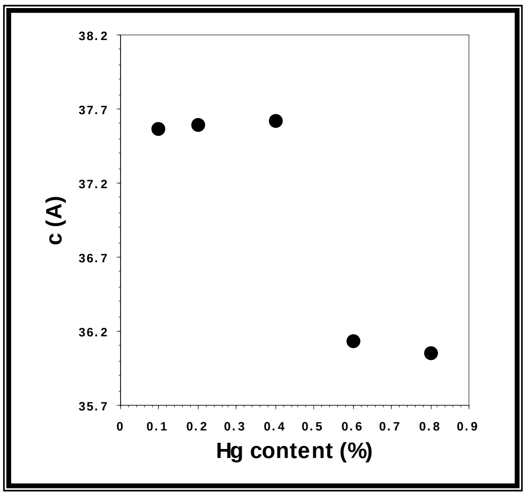


Fig(3): X-ray diffraction patterns for the $Tl_{2-x}Hg_xSr_2Ca_2Cu_3O_{10+\delta}$ samples (a)x=0.1,

b)x=0.2, (c)x=0.4. H-High T_c phase, L-low T_c phase, .-impurity phase ($Sr_2Ca_2Cu_7O_8$).



Fig(4): X-ray diffraction patterns for the $Tl_{2-x}Hg_xSr_2Ca_2Cu_3O_{10+\delta}$ samples (a)x=0.6 (b)x=0.8. H-HighT_c phase, L-low T_c phase, .-impurity phase($Sr_2Ca_2Cu_7O_8$).



Fig(5): Relation of the c-axis lattice constant with Hg content for $Tl_{2-x}Hg_xSr_2Ca_2Cu_3O_{10+\delta}$ system.

Table (1) The Variation in T_c values, lattice parameters and oxygen content for different composition of $Tl_{2-x}Hg_xSr_2Ca_2Cu_3O_{10+\delta}$ system sintered at 860°C for 3h

x	a(A°)	c (A°)	T_C (K)
0.1	3.88	37.565	115
0.2	3.782	37.591	117
0.4	3. 721	37.616	120
0.6	3.877	36.136	<77
0.8	3.925	36.050	<77

The scanning electron micrographs taken for the samples are shown in Figs. (6&7). Fig.(6) is a section of $Tl_{2-0.2}Hg_{0.2}Sr_2Ca_2Cu_3O_{10+\delta}$ structure showing irregular sizes of stacks of plate-like structure which belongs to the high-T_c phase each of these stacks compose of thousands of layers, apparently each grain (group of layers) grows in random directions. Some of the plates have grown one through the other, in different directions, giving rise to bigger grains.

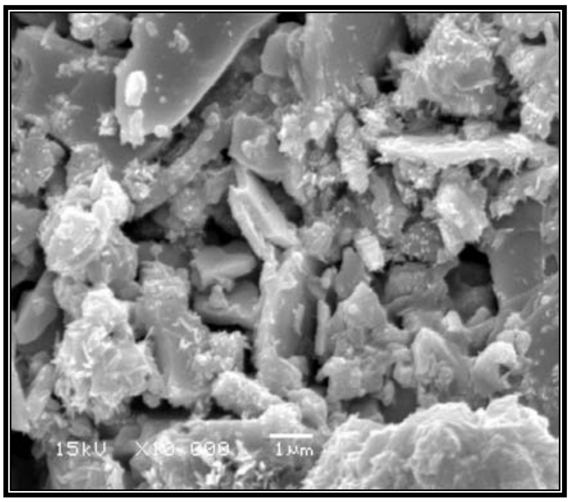
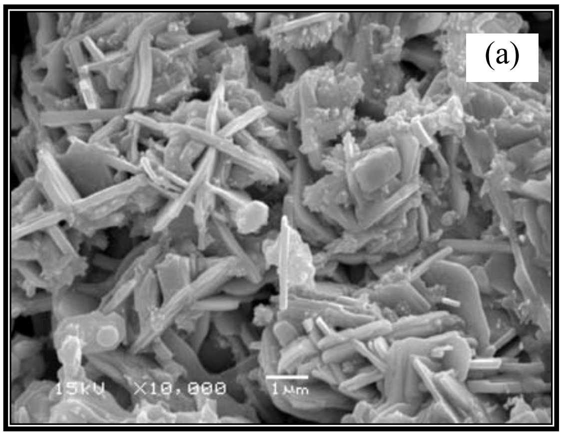
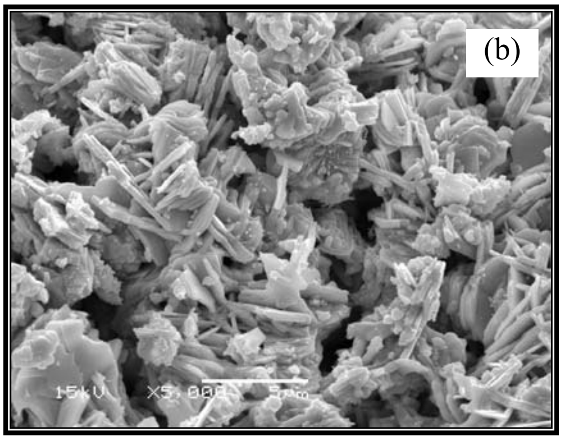


Fig.(6): SEM micrographs of the fracture surfaces of $Tl_{1.8}Hg_{0.2}Sr_2Ca_2Cu_3O_{10+\delta}$ sample.

Figs.(7a,b and c) illustrate how the particles appear at different magnifications for the same specimen. It can be inferred from these three figures, that the structure is composed of two kinds of particle features namely needle-like (the needles here are more obvious) and irregular large grain sizes. These different features reveal the fact that the structure is of two phases. As this specimen is HTSc we can definitely say that the needle like structure represents the HTSc phase while the other is the normal phase simply because the needles spread over most of the specimen, due to this point, we have chosen an area in the specimen which most of it is of irregular size particles see Fig.(7c) even these particles are floating over, seemingly larger areas of needle-like particles; however, here are most of them are oriented normally appearing as points.





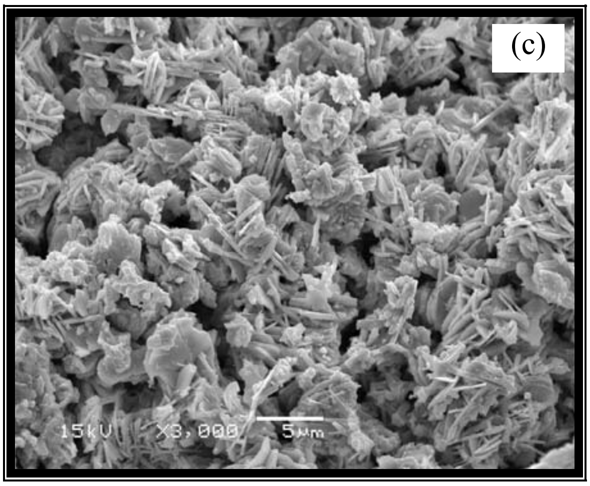


Fig.(7): SEM micrographs of the fracture surfaces of $Tl_{1.7}Hg_{0.3}Sr_2Ca_2Cu_3O_{10+\delta}$ sample,(a) X=10000 (b) X=5000 (c) X=3000

Hg doping leads to enhance the formation needle like particles of uniform grain growth. This is a possible brought about improved nucleation and growth kinetics induced by highly mobile Hg particles. From the formal theory of nucleation and growth kinetics: mercury particles act as nucleation centers and once the nucleation is complete the growth starts, Hg atoms are presumably, as a substituted, sit at Tl sites of the Tl-2223 lattice and come out with the growing superconducting grain.

Conclusions :

[1] The substitution of Hg in Tl for the compounds $Tl_{2-x}Hg_xSr_2Ca_2Cu_3O_{10+\delta}$ has exhibited a maximum value of T_c(120K) at x=0.4.

[2] XRD pattern analyses have shown a tetragonal structure, and there are at least two superconducting phases.

[3] Scanning electron microscopy has shown that 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

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