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EFFECT OF Ti NANOPARTICLES ON (Bi,Pb)-2223 SUPERCONDUCTING THIN FILMS

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Thin films of $Bi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3O_{10+d}$ with the addition of 0.2, 0.4, and 0.6% nanoparticles TiO_2 were prepared by the deposition on silicon (111) substrates using pulsed laser deposition technique. As -deposited samples annealed at 800 ^{O} C for two hours in oxygen environment, The properties of thin films are analyzed structurally by X-ray diffraction and morphologically by two techniques: scanning electron and atomic force microscopy. The analyses showed an orthorhombic structure with two phases for thin films: a high-2223 and a low-2212 phases. Four point probe method was used to study the electrical properties of the samples and all of them showed superconductivity behavior. Highest critical temperature T_C found at 116.5 K in 0.2% Ti concentration.

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1. Introduction

Superconductivity is a phenomenon of great complexity and diversity. It is one of the most interesting and challenging subfields of condensed matter physics. For instance, the mechanism of high temperature (high-T_C) superconductors remains unresolved despite of enormous efforts by scientists in the world to study it. On the other hand, tremendous progress has been made in the practical applications of superconductivity, as for the high T_C superconductors they were used as thin films to design electronic devices, microwave integrated circuits and resonators.

Among the high T_C superconductors, BiSrCaCuO (BSCCO) compounds have gained wide applications bulk or thin films due to their interesting electrical and magnetic properties with an enhancement in the chemical and thermal stability [1,2,3,4,5,6].

Different techniques have been employed to grow thin films of superconducting materials.

The Pulse laser deposition (PLD) has the advantage of producing a more controlled high-T_C materials thin film with uniformity, a relative high quality, and less contamination. Moreover, it’s reported that films prepared by PLD would have improved superconductor properties compared with those deposited by other methods [7, 8, 9, 10].

The microstructure of the Bi compound thin films was closely related to the deposition process and its specific chemical and physical characteristics. All the reported thin film growth techniques have required a post annealing or a heating of the substrate during the deposition. Films annealed in situ or ex situ oxygen pressures did strength the links and increased the contact areas between the grains thus enhance the growth of a particular phase [10, 11, 13, 14, and 15].

On other side the existence of Pb in the started materials promotes the stabilization of the high-T_C 2223 phase but its content depends on the annealing conditions [16].

It is found that multiphases formed in the film, which had influence on the electrical property of the film [17].

Takahira *et al.* [18] fabricated (Bi, Pb) 2223 epitaxial thin films on $SrTiO_3$ (100) or $LaAlO_3$ (100) single crystalline substrates by Nd:YAG pulsed laser deposition. They found that after the post-annealing Bi-2223 became the main phase due to the Pb introduction into the films. Moreover the films exhibited T_C zero of about 100 K.

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It was established that modifying the grains texture of super-conductor structure improves the indicated characteristics. Recently, researchers trying to synthesis Bi-based compounds doped or added with nanoparticles [19] to enhance their properties.

In nonstructural materials, grain boundaries may play a larger role in the overall conductivity response. This may be ascribed to amount of doping in the samples filling the intergrain space which provided effective flux pinning centers and resulted in significant enhancement of current density ; Furthermore, resulting in better grain growth and causing larger grains which leads to the improve of the strength of the connection between grains in the superconductors [17,20,21,22,23].

Oboudi *et al.* [24, 25] achieved the growth of epitaxial $M_xBi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3O_{10+d}$ thin films on Si (111) substrates, where M are Ag or Au nanoparticles and x=0.0 to 1.0%, by using PLD system and post-deposition oxygen annealing. The results showed improvement in T_C by increasing both kind nanoparticles to 1.0 wt% which had a maximum T_C value for all prepared films.

However, more investigations are needed to study the physical properties of Bi-2223 superconductors synthesized as thin film with nanoparticles addition. Thus in this work we're trying to prepare and investigate the properties of $Bi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3O_{10+d}$ thin films with the addition of TiO_2 nanoparticles, as to our knowledge, no such studies have been reported.

2. Experimental

$Ti_xBi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3O_{10+d}$ pellets with (x=0.2, 0.4 and 0.6 Ti with size 50 nm) used as a target to grow a thin films on a Single crystal silicon Si n-type substrates with ordination (111). Before the deposition process the substrate were heated to a temperature of 300 ºC.

The deposition process is carried out inside a vacuum chamber by using Nd:YAG SHG Q-switching laser with 1064 nm, 500 pulses and pulse energy 700 mJ. The focused beam is incident on the target surface making an angle of 45° with it. The plasma plume condensed on substrate placed at a distance of 2 cm in front of the target.

All the as-deposited $Bi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3Ti_xO_{10+d}$ thin films annealed at a temperature of 800 ºC with an oxygen flow of 1L/min for 2 hours to complete the oxidation process.

The thickness of the film measured by optical interferometer and was determined to be 194 nm.

The structure of the prepared samples was obtained by using X-ray diffractometer type Philips with the Cu-Kα radiation.

The electrical resistivity (ρ) was studied to evaluate the critical temperature T_C.

In order to study the surface morphology and, especially, the effect of the particulate formation on the quality of the thin film surface, atomic force microscopy (AFM) from Angestrion Advanced Inc. USA (AA3000) was used.

Scanning electron microscopy (SEM) from (FEL Inspect S50, Netherlands) used to analyze the surface morphology and nature of the grains for all samples.

Energy dispersive X-ray spectrometric (EDX) type FEI-SEM model Inspect-S50 was used to provide compositional information for elements in $Bi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3Ti_{0.2}O_{10+d}$ thin film.

3. Results and discussion

XRD pattern of all the thin films are shown in Fig. (1). Miller indices indicate that there are two phases: 2223-high and 2212-low, with Bi-2223 being the dominant one. The relative volume fractions (V. F.) of the Bi-2223 and Bi-2212 phases were estimated using formulas in [26] see Table 1.

Thin films show a peak that belongs to Si due to the absorption edge of Si substrate.

There is a consistence variation in the intensity and the position of the peaks indicating the change in phase composition of the samples and the crystalline arraignment degree. Moreover, new peaks have been noticed with increasing Ti nanoparticles added to 0.6 w%. This may attributed to the change in the oxygen amount of the films. Such behavior was also obtained by other researchers in [20, 24].

The lattice constants evaluated from 2θ of major high-T_C phase peaks, which showed the crystal symmetry of all the samples is orthorhombic, as listed in Table 1.

The deformation in the c-axis adjusts the amount of charge transfers from Bi-O layer to Cu-O layer, which will be a driving force to the pairing generation of superconductor holes forming bosons. This behavior, due to the ordered arrangements of the oxygen vacancies and the Ti addition at the same time, which led to the change of the spacing between the CuO layers and that affected the charge transfer to the CuO layers [27].

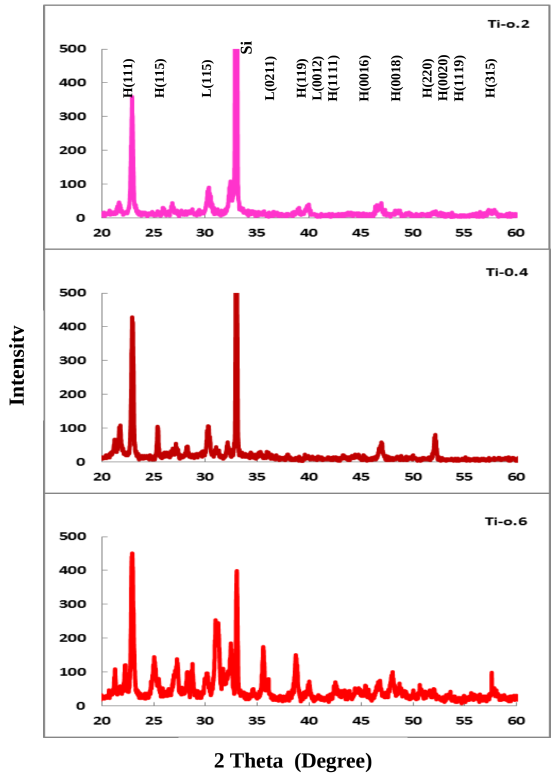

Fig.1. XRD pattern of $Ti_xBi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3O_{10+d}$ thin films

Table 1. The volume fraction of phases% , lattice parameters, and volume cell of $Ti_xBi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3O_{10+d}$ thin films with different concentrations.

x	*V.F. of phases formed (%)*		*a (Å)*	*b (Å)*	*c (Å)*	*V(Å)3*
	2223 phase	*2212 phase*				
0.2	81.818	18.181	5.782	5.489	37.458	1188.819
0.4	75	25	5.368	5.410	37.528	1089.846
0.6	80	20	5.047	5.399	37.121	1011.500

EDX spectra of the $Ti_{0.2}Bi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3O_{10+d}$ film as illustrated in Fig. 2, it confirms that the desired elements in the chemical composition of the synthesized samples are present and practically do not differ from the target composition. Overlapping of some peaks such as Bi and Pb at the same value of energy was observed indicating that Bi ions were partially replaced by Pb ions in the system.

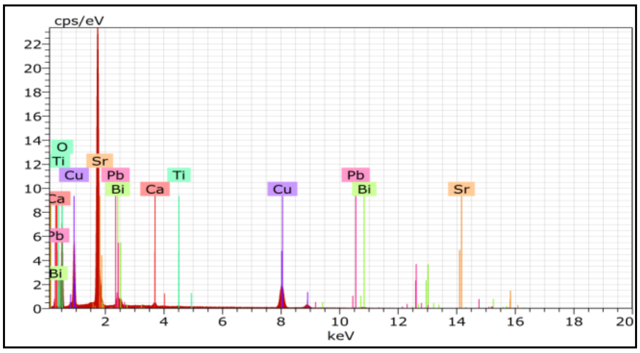

Fig.2: EDX spectra of the $Ti_{0.2}Bi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3O_{10+d}$ *thin film.*

Measurements of electrical resistivity of the thin films are displayed in Fig. 3. It is found that the addition of Ti possess somewhat metallic behavior in the normal state and superconducting transition to the zero resistance with T_C = (116.5, 114 and 112.5) K for (x=0.2, 0.4 and 0.6) respectively. It has been noted that the superconducting transition width, ΔT_C, increases with increase of Ti doping.

Thin films with composition of x=0.2, can be seen as a well-defined one step transition. This behavior can be described by decreasing impurity phases and the 2223 phase existent with a high fraction as referred in the X-ray analysis, as well as to the strong link and to the increase of the contact areas between the grains. While thin film with x = 0.6%, two steps of transition with a small tail was obtained, reflecting the decrease of the intergrain transition, irregularities and presence of impurities mainly distributed at the grain boundaries which lead to the distortion of the bond strength, and consequently degrade the electrical properties.

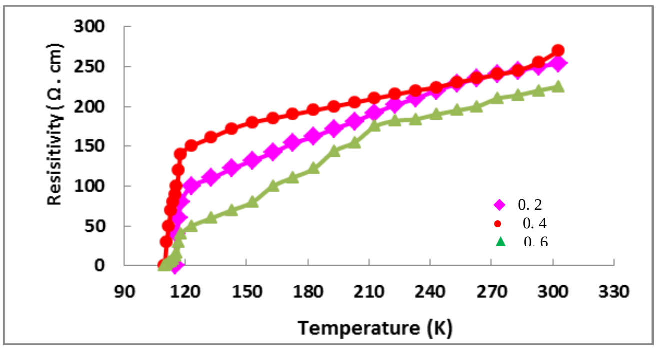

Fig. 3. Electrical resistivity as a function of temperature for $Ti_xBi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3O_{10+d}$ *thin films.*

AFM measurements used to carry out the surface morphology for different Ti concentrations as displayed in Fig. 4. The data collected for the samples from AFM images are listed in Table 2. It can be noticed that the average diameter of grains for samples decreased, while the roughness of the surface increased with the increase of the Ti nanoparticles contents.

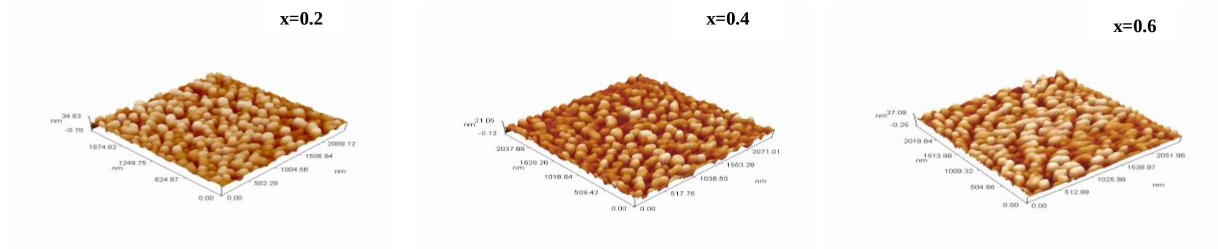

Fig.4: AFM image of $Ti_xBi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3O_{10+d}$ thin films.

Table 3. The average grains diameter and roughness of $Ti_xBi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3O_{10+d}$ thin films for different values of x.

x	Average Roughness	Average Grain diameter (nm)
0.2	2.81	120.00
0.4	4.53	91.86
0.6	5.10	94.00

The SEM micrographs of all films are shown in Fig. 5, the morphology of the films changes depending on the added Ti concentrations. Sheets (platelets) which occupy a large area of the film are probably the high-T_C phase observed for all samples. Moreover, flaky, needles, and bulky grains noticed. Voids tend to be produced among the grains.

However, film with Ti 0.2% is composed of homogenous crystallites which yield a smoother and denser surface. Increasing Ti to 0.4 % leads to change the surfaces morphology, flower like and bulky grains noticed. While the morphology of film with Ti 0.6% was more complicated, SEM showed an aggregation of nanoparticles. The formation of big area due to the diffusion of nanoparticles is observed. In our opinion, existence of Ti in the grain boundaries (appears as whit areas) leads to coalescence, thereafter it forms a thin layer on a small area of the plate like grains as shown in Fig. (4 - 0.6). Thus, the increase of Ti doping caused two different cases as confirmed by XRD and SEM measurements [28]: first case substitution and it became a part of the structure, the second case it enriched and distributed uniformly on the grain boundaries in great amount. This means the growth of the superconducting phase on the account of its surrounding phase, which supports our explanation for the higher T_C. Together with the XRD results, it can be deduced that the doping with Ti nanoparticles affected the structure of samples in different manners. Similar conclusion has been noticed by Mohammed [29].

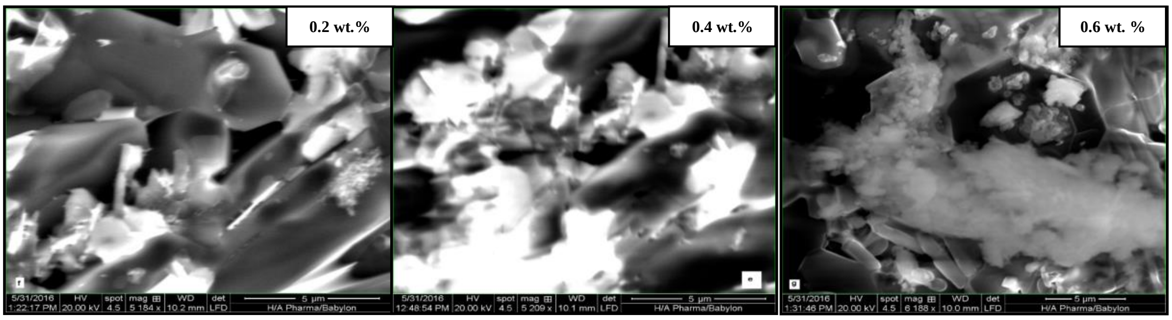

Fig. 5. SEM of $Ti_xBi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3O_{10+d}$ thin films.

However, from the view of the whole results evidently, addition Ti raised T_C of Bi-2223, the highest value found at 0.2%, although increasing Ti concentration to 0.6 degraded the granular quality of the sample but does not inhibit the formation of (BiPb)-2223 phase, meanwhile

T_C is still high at 112.5 K. This could interpret as: adding Ti nanoparticles to Bi-2223 combination with the heat treatment of their thin films will increase the contacts of the grains which provide paths for the super pairs through the strong links.

4. Conclusions

Bi(Pb)-2223 with addition of Ti nanoparticles were prepared as thin film on a Si(111) substrate . The XRD patterns of the samples showed that Bi-2223 phase is the major one independently of Ti content. Increasing Ti doping caused two different cases: first case substitution and it became a part of the structure, the second case it enriched and distributed uniformly on the grain boundaries in great amount, as confirmed by XRD and SEM measurements.

Doping with Ti altered the electronic structure of thin films which influenced the superconducting T_C, however the highest T_C value has been found at 116.5 K of 0.2%.

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