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**Effect of Zn substitution on the superconducting
properties of**

$\text{Bi}_{2-x}\text{Pb}_{0.3}\text{Zn}_x\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ System

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Abstract

High temperature superconductors with nominal composition $\text{Bi}_{2-x}\text{Pb}_{0.3}\text{Zn}_x\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ for $0 \leq x \leq 0.3$ were prepared by solid state reaction in air. The effect of substitution of Zn on the superconducting properties has been investigated. X-ray diffraction (XRD) analysis showed an orthorhombic structure with a high T_c phases (2223), low T_c -phase (2212) and impurity phases. The highest transition temperature obtained for $\text{Bi}_{1.9}\text{Pb}_{0.3}\text{Zn}_{0.1}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ was 110 K.

ملخص البحث

استخدمت طريقة تفاعل الحالة الصلبة في تحضير المركب الفائق التوصيل $\text{Bi}_{2-x}\text{Pb}_{0.3}\text{Zn}_x\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ في الهواء ودرس تأثير التعويض الجزئي للخارصين على الخواص الفائقة التوصيل للمركب. أوضحت تحليلات الأشعة السينية أن التركيب معيني وأظهرت وجود الطور الفائق التوصيل العالي (2223) ، والطور الواطئ (2212) إضافة إلى طور الشوائب. أعلى درجة حرارة انتقال ($T_c=110\text{K}$) كانت للمركب $\text{Bi}_{1.9}\text{Pb}_{0.3}\text{Zn}_{0.1}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$

Introduction

Multilayered high- T_c cuprates, which have more than three CuO_2 planes in a unit cell, exhibit very unique magnetic and SC properties because they include two types of CuO_2 planes. An outer CuO_2 plane ~OP has a pyramidal five-oxygen coordination, whereas an inner plane ~IP has a square four-oxygen one. Nuclear-magnetic-resonance ~NMR experiments revealed that the OP and the IP differ in the doping level [1]. Impurity substitution in the cuprate superconductors has proven to be a

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powerful experimental probe of the basic electronic properties. T_c suppression, the increase of the residual resistivity, the temperature dependence of the penetration depth, the impurity-induced increase of electronic density of states in the low temperature, and impurity-related local bound states seen in STM are just some of the phenomena that have been investigated through impurity studies, and a wealth of information has been garnered [2]. Zn is the most widely substituted impurity in cuprate superconductors. Advantages of using Zn are :Zn mainly substitutes the in-plane Cu(2) sites, thus the effects of planar impurity can be studied, the doping level remains nearly the same when Cu(2) is substituted by Zn, enabling one to look at the effects of disorder at almost the same hole concentration [3,4].

Jeon et al [5] studied the effect of Zn-substitution for Cu in $\text{Bi}_2\text{Sr}_2\text{Ca}(\text{Cu}_{1-x}\text{Zn}_x)_2\text{O}_y$ single crystals ($0 \leq x \leq 0.01$). They found the increase of the out-of-plane conductivity with Zn substitution and the abrupt reduction of the anisotropic parameter γ from 200~250 for the unsubstituted sample to 30~50 for the 0.98% Zn substituted one.

Naqib^[6] investigate the effect of Zn substitution on the hole content, p , and the oxygen deficiency, δ , for a series of high-quality crystalline c -axis oriented thin films and polycrystalline $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2(\text{Cu}_{1-y}\text{Zn}_y)_3\text{O}_{7-\delta}$ compounds over a wide range of composition. He found that annealing under identical conditions makes the Zn-substituted compounds somewhat more oxygen deficient compared to the Zn-free ones.

XRD experiment was conducted on zinc substituted YBCO ceramics in the temperature range 90–270K. In undoped or lightly doped samples, the abrupt change of lattice parameter at several temperatures and the large average temperature derivative of lattice parameter, originating from both thermal expansion and subtle changes of lattice parameter, give evidences for the onset and development of CuO_5 tilting upon cooling. Such titling was suppressed by zinc doping and this effect is discussed in favor of the strong and expanded nature of the disturbance on electronic states created by Zn that is essential for tilting mechanism of CuO_5 pyramids^[7].

In this paper we studied the effect of Zn substitution on the structure and transition temperature of BSCCO system to bring this superconducting material into more useful technical forms .

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Current application of high temperature superconductors include, medical imaging systems , magnetic shielding devices , infrared sensors and microwave devices ^[8] .

Experimental Part

Solid –state reactions was used to prepare samples of $\text{Bi}_{2-x} \text{Pb}_{0.3} \text{Zn}_x \text{Sr}_2 \text{Ca}_2 \text{Cu}_3 \text{O}_{10+\delta}$ for $x=0,0.1,0.2$ and 0.3 . Mixing appropriate amounts of the oxide, carbonates of Bi,Pb,Zn,Ca,Cu, and Sr develop $\text{Bi}_{2-x} \text{Pb}_{0.3} \text{Zn}_x \text{Sr}_2 \text{Ca}_2 \text{Cu}_3 \text{O}_{10+\delta}$ precursor. The mixture homogenization takes place by adding a sufficient quantity of 2-propanol to form paste, during the process of grinding for about (50-60) min. Measuring the weight of the dried mixture (w_1), and put it in an alumina crucible, calcined in a tube furnace in air that has programmable controller type (Eurptherm818) for 24 hours at 800 °C with a rate of 2°C / min. This mixture was then pressed into pellets 1.3 cm in diameter and (0.2 – 0.3) cm thick, using hydraulic type (SPECAC) under pressure of 0.5GPa. The pellets placing in alumina crucible then heated at a rate of 120 °C/h, at 850 °C. This temperature was kept constant for about 140 hours .After that, the furnace was cooled to room temperature by the same rate

Four probe dc methods at temperature range (80-300) K was used to measure the resistivity (ρ) and to determine the critical temperature (T_c)

The structure of the prepared samples was obtained by using x-ray diffractometer (XRD) (Philips).A computer program was established to calculate the lattice parameters a, b, and c. The program is based on Cohen's least square method ^[9] .

The densities (d_m) of unit cell of the samples were calculated using the following equation ^[10] :

$$d_m = \frac{W_m}{N_a V}$$

Where W_m is the molecular weight, N_a is Avogadro's number and V is the volume of the unit cell.

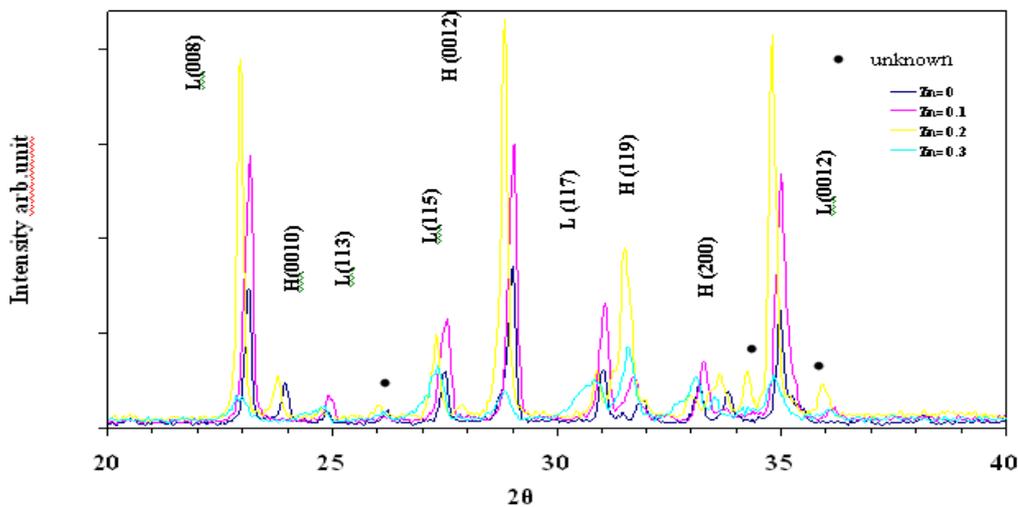
Result and discussions

The series of XRD spectra for BiSrCaCuO samples with Zn varying from 0 to 0.3 are shown in Fig.(1). This Fig. indicate that the samples have polycrystalline orthorhombic structure, with lattice constants a,b,c listed in Table (1) for different Zn content. Two main phases were

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observed in the XRD spectra: high T_c -phase and low T_c -phase; Although the intensity of the $\text{Bi}_{1.8} \text{Pb}_{0.3} \text{Zn}_{0.2} \text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ sample is much more than the other but the existence of impurity phase reduces the transition temperature as shown in coming item. The unknown phases could interpret to creation of the stacking faults which leads to deform the structure. Indeed a shifting of peak position for $\text{Bi}_{1.8} \text{Pb}_{0.3} \text{Zn}_{0.2} \text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ and $\text{Bi}_{1.7} \text{Pb}_{0.3} \text{Zn}_{0.3} \text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ in compression with pure sample was observed, this may attributed to the substitution of Zn ions in Bi ions.



Fig(1): X-ray diffraction patterns for $\text{Bi}_{2-x}\text{Pb}_{0.3}\text{Zn}_x\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ System

The ratio $c/a, c/b$ and density of unit cell were calculated for all samples. It is shown from Fig.(2) and Table(1) the $\text{Bi}_{1.9} \text{Pb}_{0.3} \text{Zn}_{0.1} \text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ system has c/a ratio more than the other samples which indicates an isotropic crystal of this system. Also the same system has

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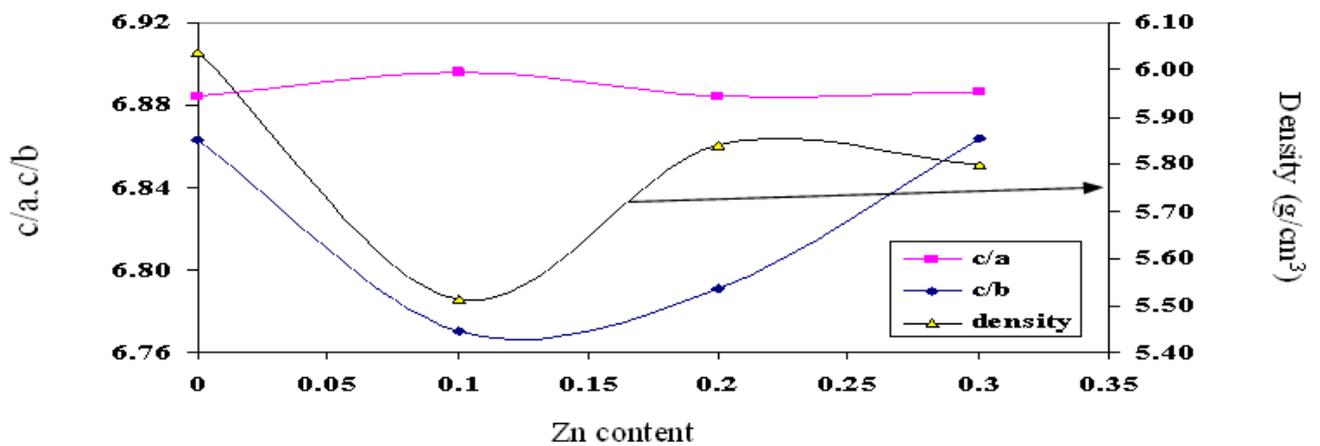


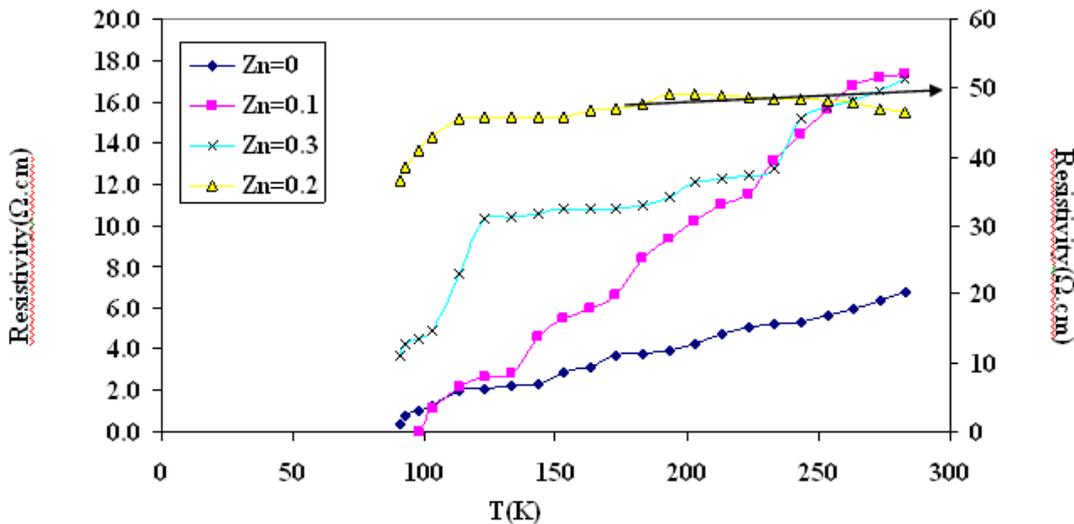
Fig.(2) $c/a, c/b$ and density of unit cell as a function of Zn content

less value of density; the reason may be due to the increase of the unit cell volume.

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The temperature dependence of the electrical resistivity (ρ) for Zn free samples and samples with different Zn contents of ($x=0.1-0.3$) in $\text{Bi}_{2-x}\text{Pb}_{0.3}\text{Zn}_x\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ are shown in Fig.(3) .Metallic behavior was observed for all samples above the onset temperature ,although in some cases($x=0.2$ and $x=0.3$) a complete zero-resistance could not be observed .In addition to that the superconducting transition were not sharp and a double step resistive transition which is an indication of weak links was



Fig(3): Resistivity versus temperature for $\text{Bi}_{2-x}\text{Pb}_{0.3}\text{Zn}_x\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ System

observed. The above results may be related to the variation of oxygen content or to the existence of more than two phases as denoted in X-ray diffraction analyses.

It is found from Table (1) that addition of 0.1 Zn content to the composition enhances the critical temperature to 110 K, while excessive Zn additive 0.2 and 0.3 will reduces the transition temperature to less than the boiling point of liquid nitrogen <77K. The reason may be interpreted as the destruction of the low phase by small amount of Zn =0.1 at the early stage may enhance the formation of high T_c -phase , whereas more addition of Zn will convert the high phase into low phase by producing a depletion layer of PbO which converts BPSCCO to BSCCO and destabilize the high T_c -phase, high T_c -phase due to the

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decrease of the number of CuO₂ layers and hence the disappearance of the 2223 type phase^[11] .

Table(1):Values of lattice constants, c/a, c/b ,Volume ,transition temperature, and density of unit cell.

	0.3	0.2	0.1	0	Zn
	5.3926	5.3857	5.3952	5.393	a(Å)
	5.4104	5.4603	5.4954	5.4098	b(Å)
	37.1362	37.0795	37.2068	37.1294	c(Å)
	6.886511	6.884806	6.896278	6.884739	c/a
	6.863855	6.790744	6.770535	6.863359	c/b
	1083.49	1090.417	1103.136	1083.252	V(Å) ³
	<77	<77	110	93	T _c (K)
	5.798	5.841	5.512	6.039	Density(g/cm ³)

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Conclusions:

An addition of 0.1 Zn content to the $\text{Bi}_{2-x}\text{Pb}_{0.3}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ composition enhances the critical temperature to 110K, while more excessive Zn addition 0.2 and 0.3 will reduce the transition temperature to less than the boiling point of liquid nitrogen < 77K. X-ray diffraction analysis showed polycrystalline orthorhombic structure and there is an isotropic crystal of $\text{Bi}_{2-x}\text{Pb}_{0.3}\text{Zn}_{0.1}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$.

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