

## Superconducting Properties of the $(\text{Bi}_{0.8}\text{Pb}_{0.2})_2(\text{Sr}_{0.9}\text{Ba}_{0.1})_2 \text{Ca}_2\text{Cu}_{3-x}\text{Ni}_x\text{O}_{10+\delta}$ System.

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### Abstract:

The effect of substitution of Ni on Cu in  $(\text{Bi}_{0.8}\text{Pb}_{0.2})_2(\text{Sr}_{0.9}\text{Ba}_{0.1})_2 \text{Ca}_2\text{Cu}_{3-x}\text{Ni}_x\text{O}_{10+\delta}$  for  $(x=0,0.1\dots1,2,3)$  superconductor system and sintering time has been investigated. The samples were prepared by solid-state reaction methods.

The results show that the optimum sintering temperature is equal to  $850^\circ\text{C}$ , and the sintering time is equal to 140 h. The highest transition temperature ( $T_c$ ) obtained for  $(\text{Bi}_{0.8}\text{Pb}_{0.2})_2(\text{Sr}_{0.9}\text{Ba}_{0.1})_2 \text{Ca}_2\text{Cu}_{3-x}\text{Ni}_x\text{O}_{10+\delta}$  composition was 113 with  $x=0.8$

Phase analyses of the samples by X-ray diffraction (XRD) analysis showed an orthorhombic structure with a high  $T_c$  phases (2223) as a dominant phase and low  $T_c$  phase (2212) in addition to some impurity phases.

**Key words:** Bi-based superconductor, crystal structure, electrical resistivity.

### Introduction:

Since Maeda et al.[1] discovered superconductivity in the BiSrCaCuO (BSCCO) system, significant research has been focused on the synthesis and the use of various dopants to volume fraction high  $T_c$  phase in multiphase ceramics. This family of superconductors can be expressed by the general formula  $\text{Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+4}$ , where  $(n=1, 2, 3)$ ,  $n$  is CuO layers [2], dependence on  $n$  values, the three phases have different critical temperature, the Bi-2201 has 10K, the Bi-2212 has 80K, and the Bi-2223 has 110K. Kocabas. [1] prepared the samples by used the solid state reaction, The sample  $(\text{Bi}_{1.5}\text{Pb}_{0.4}\text{Sb}_{0.1}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y)$  were sintered at  $850^\circ\text{C}$  for (60, 80 and 100)h in air and then furnace cooled to room temperature. He was found the high  $T_c$  at 108, 110, and 112K, respectively. [3] reported a  $T_c$  of 128K for

$\text{Bi}_{1.95}\text{Tl}_{0.05}\text{Ba}_{1.9}\text{Sr}_{0.1}\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$  system without Pb doping. Halim et al.[4](2010) reported influence of addition of Eu on the structural and superconducting properties of  $(\text{Bi}_{1.6}\text{Pb}_{0.4})\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{Eu}_x\text{O}_{10+\delta}$  ceramics prepared by solid-state reaction methods. X-ray diffraction data shows that the addition of  $x=0.03$  of Eu helped in improving the formation of the high phase. They found that the critical temperature  $T_c$  gradually decreases with increasing  $x$ . In this paper we investigated the effect of substitution Ni on Cu in  $(\text{Bi}_{0.8}\text{Pb}_{0.2})_2(\text{Sr}_{0.9}\text{Ba}_{0.1})_2 \text{Ca}_2\text{Cu}_{3-x}\text{Ni}_x\text{O}_{10+\delta}$  for  $x=0,0.1\dots1,2,3$  on the structural and superconducting properties.

### Materials and Methods:

The samples under study prepared by conventional solid-state reaction.

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Bi<sub>2</sub>O<sub>3</sub>(99.9%), Pb<sub>3</sub>O<sub>4</sub>(99.9%), SrCO<sub>3</sub>(99.9%), BaCO<sub>3</sub>(99.9%), CaO(99.9%), CuO (99.9%) and Ni<sub>2</sub>O<sub>3</sub>(99.9%) powders were used as starting materials. The powders with the molar ratio of [Bi] : [Pb] : [Sr] : [Ba] : [Ca] : [Cu] : [Ni] = 1.6 : 0.4 : 1.8 : 0.2 : 2 : 3(1-x) : (0 ≤ x ≤ 3). The powder of precursor was mixed together by using agate mortar. The mixture homogenization takes place by adding a sufficient quantity of 2-propanol to form a past during the process of grinding from about (1 h). In the second step, the materials were grounded to a fine powder and then calcined in air at 800 °C for (24)h and after the mixture was then pressed into pellets (1.3 cm) in diameter and (0.2) cm thick, using hydraulic type (SPECAC), under pressure of (5 ton). The pellets were sintered in air at 850 °C for (140 h). The structure of the prepared samples obtained by using x-ray diffraction (XRD) type (Philips) with the Cu<sub>Kα</sub> source. A computer program has been used to calculate the lattice parameters, which is based on Cohen's least square method.

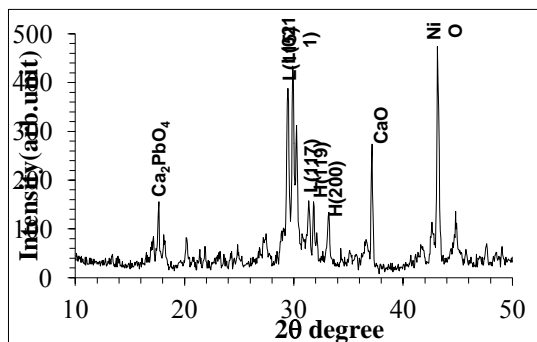
### Results and Discussion:

X-ray diffraction patterns for the (Bi<sub>0.8</sub>Pb<sub>0.2</sub>)<sub>2</sub> (Sr<sub>0.9</sub>Ba<sub>0.1</sub>)<sub>2</sub> Ca<sub>2</sub>Cu<sub>3-x</sub>Ni<sub>x</sub>O<sub>10+δ</sub> system with (x=0,0.1...1,2,3) prepared with sintered at 850 °C for 140h are shown in Fig. (1-2). It could be seen from the spectra that there were two main phases in samples of the Bi-base systems, high-T<sub>c</sub> phase (2223), low-T<sub>c</sub> phase(2212) and a small amount of impurity phases of Sr<sub>2</sub>Ca<sub>2</sub>Cu<sub>7</sub>O<sub>8</sub>, Ca<sub>2</sub>PbO<sub>4</sub>, CaO and NiO. An analysis of the number of corresponding peaks and their intensities indicating the dominant high-T<sub>c</sub> phase with increasing Ni while

have low-T<sub>c</sub> increased with increasing more than 1. Also it is noticed that the main peaks of the samples are attributed to the 2223- phase, this result indicate that the substitution by Ni may well enhances crystallinity and volume fraction of the 2223 phase and this leads to increases in the T<sub>c</sub> value while in contrast, that of the 2212 phase raises up when x=1,2 and 3. Fig.(2) shows x-ray diffraction patterns for the values x=2,3 show that there were two main phases in samples of the Bi-base systems : (2212) phase, a Low-T<sub>c</sub> phase and amount of impurity phases Ca<sub>2</sub>PbO<sub>4</sub>, CaO and NiO. Indeed substitution of Ni (more than 1) cause more cuprate vacancies that the HTS need to a high scattering effect of super electrons in crystalline structure.

The structural calculations of the lattice parameters were carried with the help of a computer program. The values of lattice parameters, c/a and T<sub>c</sub> values for the samples are listed in Table(1) show an orthorhombic structure. It is found from this Table an increase in the a-axis lattice and decrease in the c-axis lattice with increases Ni content for the samples consist This could be explained by the increase of the oxygen content in the unit cell by the replacement of Cu<sup>+2</sup> by Ni<sup>+3</sup> in the structure. It was speculated that the excess of oxygen goes into the bismuth oxide layers causing an increase in the lattice parameter a and consequently a decrease in the lattice parameter c as was confirmed by Yilmazlar et al[5]. Another reason is due to the ionic radii of Ni<sup>+3</sup> (0.78 Å) is shorter than of Cu<sup>+2</sup>(0.96Å) which render c-parameter to be shorter or get deformed[6].





$(\text{Bi}_{0.8}\text{Pb}_{0.2})_2(\text{Sr}_{0.9}\text{Ba}_{0.1})_2 \text{Ca}_2\text{Cu}_{3-x} \text{Ni}_x\text{O}_{10+\delta}$

Figs.(3 and 4) and Table (1) show the effect of Ni on Cu in the  $(\text{Bi}_{0.8}\text{Pb}_{0.2})_2 (\text{Sr}_{0.9}\text{Ba}_{0.1}) \text{Ca}_2\text{Cu}_{1-x} \text{Ni}_x\text{O}_{10+\delta}$  with  $x$  ranging ( $x=0,0.1, \dots, 1,2,3$ ) by preparing a samples at 850 °C for 140h. It is found from Fig(3) that the resistivity for all the samples decrease with decreasing temperature as though is some cases a complete zero-resistance could not be found .The critical temperature for sample has no Ni content ( $x=0$ ) is equal to 100 K. The sample with  $x= 0.8$  shows a higher critical temperature 113K, and the resistivity went down nearly sharp as shown in Fig. (4) .The reason may be attributed to the existence of the high  $T_c$ - phase as referred in the x-ray analysis, while the sample with  $x=0.7$  display a  $T_c$  at 103K. Increasing Ni to 0.9,1,2 and 3 decreases the critical temperature to  $< 77\text{K}$  ,as we expect (if we use liquid He for cooling) ,the decrease in  $T_c$  are further evidence that the presence of the increasing amounts of Ni content in Bi-2223 phase stabilizes and promotes the growth of Bi-2212 phase at the expense of Bi-2223 [4]. While that for  $x=3$  the compound becomes insulator , this unique behavior might be related to nothingness found element Cu that be layer of Cu-O<sub>2</sub>. Suggests that increase Ni the cationic substituting for Cu tend to inflict disorder in Cu-O networks which at a higher dopant level may show superconducting-like resistance behavior but don't reach to

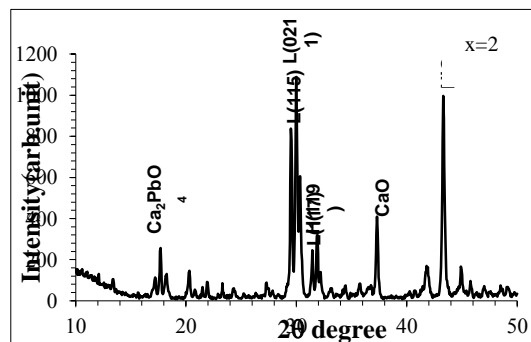


Fig.(2):X-ray diffraction patterns for the samples for  $x=2$  and 3.

zero resistivity. The reason of that may be ascribed to two effects:(a)The changes of the carrier concentration and microstructure induced by the replacement. (b)The decrease of  $T_c$  should be mainly attribute to the magnetic scattering induced by Ni ions but its valence can fluctuate (Ni+2/Ni+3) in CuO plane [7].

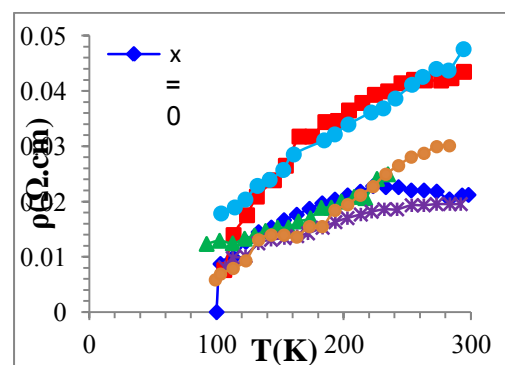


Fig.(3): Temperature dependence of resistivity for  $(\text{Bi}_{0.8}\text{Pb}_{0.2})_2(\text{Sr}_{0.9}\text{Ba}_{0.1})_2 \text{Ca}_2\text{Cu}_{3-x} \text{Ni}_x \text{O}_{10+\delta}$ , for different nominal composition.

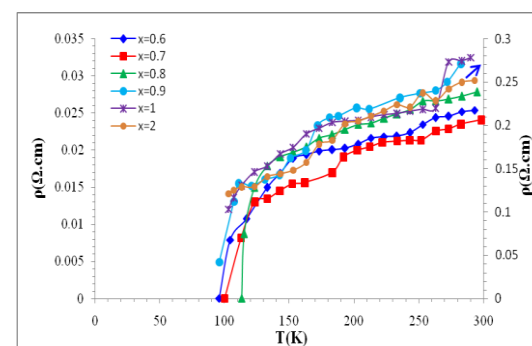
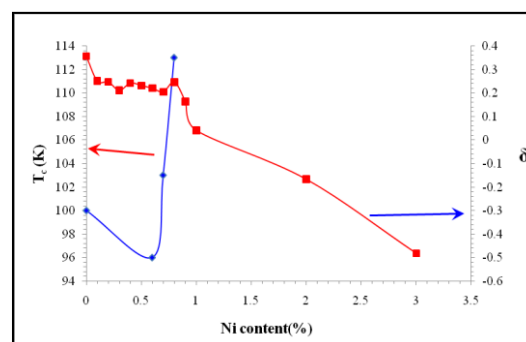


Fig.(4): Temperature dependence of resistivity for  $(\text{Bi}_{0.8}\text{Pb}_{0.2})_2(\text{Sr}_{0.9}\text{Ba}_{0.1})_2 \text{Ca}_2\text{Cu}_{3-x} \text{Ni}_x \text{O}_{10+\delta}$ , for different nominal composition.

The Ni substitution effect on the excess oxygen was also investigated, to determine the optimum value of substitution ratio required to obtain the critical temperature  $T_c$ . The variation of oxygen content ( $\delta$ ) and the critical temperature  $T_c$  as a function of Ni content is shown in Fig.(6) and Table (1). From this figure it can be observed that substituting Ni decreases the amount of oxygen content ( $\delta$ ) in the combination. It has been suggested that the loss amount of oxygen changes the distortion of the Bi–O layers, a large change of hole concentration ( $n_h$ ) in the Cu–O<sub>2</sub> planes would be anticipated. The redistribution of the hole concentration between the Bi–O layers and the Cu–O<sub>2</sub> planes would lead to changes in lattice parameters and the hole concentration in the Cu–O<sub>2</sub> planes, resulting in the large decrease in  $T_c$ [8].

It was found that critical temperature  $T_c$  depends on the number of Cu–O<sub>2</sub> layers and on the oxygen

content in the samples, however the higher  $T_c$  value is found for higher value of oxygen content as shown for samples with  $x=0.8$ . This could be attributed to the presence of excess oxygen atoms in the Cu–O<sub>2</sub> layers will create more holes in the perovskite layers, the creation of holes will shorten the Cu–O<sub>2</sub> bond length and this leads to an improvement of the  $T_c$ .



**Fig.(6):** Variation of the critical temperature  $T_c$  and oxygen content ( $\delta$ ) as a function of Ni content.

**Table (1):** Variation in lattice parameters,  $c/a$  and  $T_c$  values, for different composition of  $(\text{Bi}_{0.8}\text{Pb}_{0.2})_2(\text{Sr}_{0.9}\text{Ba}_{0.1})_2\text{Ca}_2\text{Cu}_{3-x}\text{Ni}_x\text{O}_{10+\delta}$

x	a(Å)	b(Å)	c(Å)	c/a	$\delta$	$T_c$ (K)
0.0	5.391	5.411	37.281	6.915	0.356	100
0.1	5.395	5.36	37.21	6.897	0.251	<77
0.2	---	---	---	---	0.247	<77
0.3	5.399	5.42	37.12	6.875	0.211	<77
0.4	---	---	---	---	0.242	<77
0.5	5.41	5.404	37.044	6.847	0.232	<77
0.6	5.425	5.395	36.955	6.811	0.22	96
0.7	5.431	5.361	36.91	6.796	0.205	103
0.8	5.448	5.266	36.89	6.771	0.246	113
0.9	5.437	5.2	36.881	6.783	0.163	<77
1	5.495	5.416	36.98	6.854	0.041	<77
2	---	---	---	---	-0.167	<77
3	---	---	---	---	-0.482	Insulator

### Conclusion:

This research presents the experimental results of addition of Ni in  $(\text{Bi}_{0.8}\text{Pb}_{0.2})_2(\text{Sr}_{0.9}\text{Ba}_{0.1})_2\text{Ca}_2\text{Cu}_{3-x}\text{Ni}_x\text{O}_{10+\delta}$  superconductor. The measurements showed that critical temperature was maximum value with

$x=0.8$  where  $T_c = 113\text{K}$ . XRD patterns analyses show an orthorhombic crystal structure, and there is a reduction of c-parameter value with the increase of Ni content.

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### الخواص الفائقة التوصيل للنظام $(Bi_{0.8}Pb_{0.2})_2(Sr_{0.9}Ba_{0.1})_2 Ca_2Cu_{3-x} Ni_x O_{10+\delta}$

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#### الخلاصة:

تم دراسة تأثير التعويض الجزئي للنكل بالنحاس للمركب  $(Bi_{0.8}Pb_{0.2})_2(Sr_{0.9}Ba_{0.1})_2 Ca_2Cu_{3-x} Ni_x O_{10+\delta}$  الفائق التوصيل ولقيم  $x = 0, 0.1, 1, 2, 3$  وكذلك تم دراسة تأثير زمن التليد. حضرت النماذج باستخدام طريقة تفاعل الحالة الصلبة. وقد بينت النتائج إن الظروف المثلى للتخصير هي 850 درجة مئوية للتليد وبزمن 140 ساعة. وكانت أعلى درجة حرارة حرجة مساوية إلى 113 كلفن للمركب حيث نسبة النكل مساوية إلى 0.8.

بينت فحوصات الأشعة السينية إن التركيب هو معيني قائم. كما تبين ان الطور العالي (2223) للتوصيل الفائق هو الغالب إضافة إلى الطور الواطئ (2212) كما لوحظ وجود بعض أطوار الشوائب.