تأثير استبدال البيئة الحيوية للنحاس باللانثنيوم علي خصائص النقل للموصل الفائق YBa2(Cu1-x Lax)3O7-6 د. سناء مسعود عبد القادر ، أ. نجاح ابراهيم الحراري ، د. سعد سالم سعد قسم الفيزياء- جامعة غريان – كلية التربية – ككلة , المعهد العالي للعلوم والتقنيات الطبية – ابوسليم

الملخص العربي:

 $YBa_2(Cu_{1-x} La_x)_3O_{7-\delta}$  الموصل الفائق النحاس في لنكن قادرين علي انتاج مادة فائقة التوصيل عالية الجودة , يجب فهم استبدال إشارت الدراسة لخصائص النقل في هدا العمل باستخدام قياسات درجة ( X = 0.0.66 ) بتركيز La باللانثنيوم الحرارة الي انه يمكن الحصول علي افضل خصائص للتوصيل الفائق باستبدال تركيز النحاس باللانثنيوم بتركيز معي المعن الحصول علي افضل خصائص للتوصيل الفائق العربي الفائق المعن الحصول علي افضل خصائص للتوصيل الفائق الفائق النوم العربي علي المعائر المعل باستخدام قياسات درجة ( X = 0.0.66 ) بتركيز ( X = 0.0.66 ) باللانثنيوم الحرارة الي انه يمكن الحصول علي افضل خصائص للتوصيل الفائق المعن المعائم المعائر المعائر المعائر المعائر الفائق المعائر المعا

X (X = . ) (X = . )

وأدني ( $ho=9.2\mu\Omega.
m{cm}$ ) تتوافق ضمن التركيز ( $ho=9.2\mu\Omega.
m{cm}$ ) وأدني مقاومة كهربائية

فوق هذا النطاق X( لوحظ خفض وانحلال الموصل الفائق مع زيادة تجاوز التنشيط الزائد للانثنيوم لهذا المدي يؤدي إلى فصل قوي للسلاسل والشبيكة (الهيكلية),  $(x \le 0.06) = x$ ) طريقا بديلا لتلدن الأكسجين على نطاق واسع. $(0.04) \ge x \ge 0.02)$  بينما يمكن اعتماد أفضل تنشيط للانثنيوم ضمن المدي

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# EFFECT OF REPLACEMENT OF THE Cu VITAL ENVIRONMANT FOR La ON THE TRANSPORT PROPERTIES OF YBa<sub>2</sub>(Cu<sub>1-x</sub> La<sub>x</sub>)<sub>3</sub>O<sub>7</sub>.

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### Abstract

To be able to produce high quality superconducting material, the effect of substitution of copper in  $YBa_2(Cu_{1-x} La_x)_3O_{7-\delta}$ superconductor by La content ( x=0-0.06 ) have to be understood. The study of transport properties of YBa<sub>2</sub>(Cu<sub>1-x</sub>  $La_x$ <sub>3</sub> O<sub>7- $\delta$ </sub> in this work using temperature measurements indicated that the best superconducting properties can be obtained for substitution of Cu by La concentration within the range (x=0.02-0.04). The highest  $T_c$  ( $\approx$ 90K), sharpest transition to superconductivity ( $\Delta$ =5 K) and lowest electrical resistivity ( $\rho$ = 9.2 $\mu$ \Omega.cm) are corresponding to the substitution samples of x=0.02 and 0.04. Depression and degradation of the superconducting state as (x) increases above this rang is observed. Excess dopant of La ( $x \le 0.06$ ) results in a strong decoupling of chains and planes whereas optimal La doping  $(0.02 \le x \le 0.04)$  can be adopted an alternate route to extensive oxygen anneal.

## **1.Introduction**

The search for high-temperature superconductivity and novel superconducting mechanisms

Is one of the most challenging tasks of condensed-matter physicists as well as material scientists

[1-2]. The studies on various substitutions in oxide superconducting systems have proven to be of great importance since changes in the critical transition temperature  $(T_c)$  are usually observed. For

Example, the effects of non-isovalent substitutions for Y in YBCO superconductors have attracted a great deal of attention in the past[3,4]. Results show that, basically, such doping can vary the hole concentration in a controlled manner influencing the superconducting properties of the material obtained.

Doping of different ions at the copper sites in YBCO superconductors serves as a useful diagnostic probe to investigate the role of different copper sites in the occurrence of superconductivity in these superconductors[5-6]. In almost all such cases, the destabilization of YBCO superconducting phases and consequently the degradation of superconductivity in these compounds have been determined at low substitutional level. Also, the superconducting properties severely degraded by the substitution of transition elements for copper[7,8]. This Suggesting that the principal component for superconductivity involves the Cu-O bonding.

In this work, we present a systematic study of the effect of Cu substitution by La on the transport properties of  $YBa_2(Cu_{1-x} La_x)_3O_{7-\delta}$ .

#### **2.Experimental Work**

Appropriate amounts of yttrium oxide, barium carbonate and copper oxide with starting composition  $YBa_2Cu_3O_{7-\delta}$  were mixed and ground in a marble mortar for one hour. The mixture was

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calcined in air at 910 °C for 24 hours, with several intermittent grinding followed by oven cooling at 40°C per hour. The powders were reground and then pressed into tablets of ~13 mm diameter and 4 mm thickness using SPECAC press. The tablets were sintered at 910 °C for 24 hours and slow cooled to room temperature  $\cdot$ .

Samples were prepared by using solid state reaction with four starting composition YBa<sub>2</sub>(Cu<sub>1-x</sub> La<sub>x</sub>)<sub>3</sub>O<sub>7- $\delta$ </sub> for x=0.0, 0.02, 0.04, 0.06. Sample were prepared by thoroughly mixing appropriate amounts of high purity ( $\geq$  99.99%) powders of BaCO<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub>, La<sub>2</sub>O<sub>3</sub>, and CuO with starting compositions YBa<sub>2</sub>(Cu<sub>1-x</sub> La<sub>x</sub>)<sub>3</sub>O<sub>7- $\delta$ </sub> for x=0, 0.02, 0.04, and 0.06. These powders were heated for 24 hours at 910 °C with several intermittent grindings and oven cooled. The powders were then pressed into pellets with approximately 13 mm diameter and 4 mm thick and heated at 910 °C in air for another 24 hours followed by furnace cooling to room temperature .

Van der pauw method shown in Figure 3.1 was used to determine the resistivity at room temperature. The sample was fixed on to a sample holder with silver paint. Digital voltmeter and multimeter were used to record the voltage and resistance respectively.



Figure.1 Four-point probe electrical resistivity measurements method, (a) van der

pauw (b) a typical four point Probe The room temperature resistivity *p* was computed by

p =

(1)

Where: *t* is the thickness of the sample,  $V_{CD}$  is the voltage between the points C and D and  $I_{AB}$  is the current through point A to B.

The resistance at various temperature of the samples was measured by four-point probe technique using silver paint contacts in closed cycle refrigerator. The temperature was cooled down at a cooling rate of 40  $^{\circ}$ C/ hour and heated up with the same rate. The resistance and temperature were recorded and plotted as graphs with a normalized resistance versus temperature.

## **3.RESULTS AND DISCUSSION**

To be able to produce high quality superconducting material, the effect of substitution of copper in  $YBa_2(Cu_{1-x} La_x)_3O_{7-\delta}$  superconductor by La content (x=0-0.06) have to be understood. The experimental results obtained by using resistivity measurement techniques are presented in this work to show the effect of Cu substitution by La on the transport properties and T<sub>c</sub> transition.

Figure( 2 )shows the temperature dependence of the electrical resistance for several YBa<sub>2</sub>(Cu<sub>1-x</sub> La<sub>x</sub>)<sub>3</sub>O<sub>7- $\delta$ </sub> samples from 300 K to 4.2 K, with La concentration range from x=0 to 0.06. All the samples within La concentration range (x=0-0.04) show metallic behavior ( $\frac{\partial R}{\partial T} > 0$ )before transition to superconducting state. Increasing La concentration x to 0.06, the sample shows a semiconducting behavior ( $\frac{\partial R}{\partial T} < 0$ ) before transition to superconductivity. The sample x=0.06 shows much higher resistance values and larger broad transition to superconductivity summarized in Table 4.2. Furthermore, highest T<sub>c</sub> ( $\approx$  90 K) and sharpest transition to superconductivity ( $\approx$  5 K) is seen for the doped samples with La within the range x=0.02 and 0.04. Figure(3) shows the concentration dependence of the onset transition temperature.



Figure( 2 ) The temperature dependence of the resistance of YBa<sub>2</sub>(Cu<sub>1-x</sub> La<sub>x</sub>)<sub>3</sub> O<sub>7- $\delta$ </sub> for x=0, 0.02, 0.04 and 0.06

It is observed that the highest  $T_{c \text{ onset}}$  corresponds to the samples of x=0.02 and 0.04. Whereas Figure (4) show the concentration dependence of the critical transition temperature  $T_{c \text{ zero}}$  of YBa<sub>2</sub>(Cu<sub>1-x</sub> La<sub>x</sub>)<sub>3</sub> O<sub>7- $\delta$ </sub>. The highest  $T_c$  is observed in the samples with La concentration in the range of x=0.02 and 0.04, whereas the lowest  $T_c$  value is seen of the sample (x=0.06).Figure (5) shows the variation of the broadening of the superconducting transition (width= $\Delta$ T) with La concentration of YBa<sub>2</sub>(Cu<sub>1-x</sub> La<sub>x</sub>)<sub>3</sub> O<sub>7- $\delta$ </sub> prepared samples . It can be clearly seen that the sharpest transition corresponds to the samples within La concentration range of x=0.02-0.04. Figure (6) show the concentration dependence of electrical resistivity ( $\rho$ ) of the Journal of Faculties of Education **52**  YBa<sub>2</sub>(Cu<sub>1-x</sub> La<sub>x</sub>)<sub>3</sub>O<sub>7- $\delta$ </sub> prepared samples . This figure shows that the lowest resistivity value ( $\rho = 9.2\mu \Omega.cm$ ) corresponds to the sample of La concentration of x=0.02, and the highest value ( $\rho = 4.3x10^{-5} \Omega$  .cm) corresponds to La concentration x=0.06. The best superconducting properties corresponds to the samples of La concentration x=0.02 and 0.04.

Kini et al. (2003) reported [9] that the reduction in copper valence is evident from decrease in resistivity ( $\rho$ ) with increasing x. Any reduction in copper would have resulted in a consequent elongation in the c-axis. Within La concentration x=0.02, it is evident that the dominant charge neutralising mechanism is the intake of oxygen into lattice as reported (Kini et al. 2003). Th rise of La concentration x=0.02 up to 0.06 results in an increase of resistivity values ( $\rho$ ). This increase of the resistivity is in agreement [10] with (Llonca et al. 1995). They reported a model to better understanding of the involved carrier scattering mechanisms for the effect of the partial substitution of Cu by Zn. Their model has been interpreted in the percolative phase separation theory.

It has been reported [11] (Elizabeth et al. 1999) that the dual role of substituting Cu by Co is to enhance  $T_c$  at low concentration and depressing it in higher dopant concentration.

The published result agrees completely with the results of the present in the case of Cu substitution by La. This obtained result can be understood on the basis of the apex oxygen model (Elizabeth et al. 1999) and structural studies [12] (Mark et al. 2000) in doped YBCO indicate that the most sensitive change of crystal structure on doping is related to apical oxygen .Several theoretical models such as (Elizabeth et al. 1999) emphasize the

significance of this oxygen in the occurrence of superconductivity.

Table 3.1. Onset temperature ( $T_{c \text{ onset}}$ ), zero resistance temperature ( $T_{c \text{ zero}}$ ), resistivity ( $\rho$ )

at 300K, transition width ( $\Delta T_c)$  and the normal state of YBa\_2(Cu\_{1-x}\ La\_x)\_3O\_{7-\delta}

X	Tc onset(K)	T <sub>c</sub> zero(K)	$\Delta T_{c}(\mathbf{K})$	ρ= (300K)Ω	Normal state
0	86	76	10	1.1x10 <sup>-5</sup>	metallic
0.02	88	83	5	9.2x10 <sup>-6</sup>	metallic
0.04	90	83	7	1.7x10 <sup>-5</sup>	metallic
0.06	89	62	27	4.3x10 <sup>-5</sup>	semiconductor

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Figure( 3 )The concentration dependence of the onse Figure (3) The concentration dependence of the onset transition temperature  $T_{c \text{ onset}}$  of

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 $YBa_2(Cu_{1-x} La_x)_3 O_{7-\delta}$  prepared samples

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Figure( 4 ) The concentration dependence of the zero transition temperature  $T_{c\ zero}$ 

of the  $YBa_2(Cu_{1-x} La_x)_3O_{7-\delta}$  prepared samples



Figure ( 5 ) The concentration dependence of the transition width ( $\Delta T)$  of the  $YBa_2(Cu_{1\text{-}x}$ 



Figure ( 6 ) Electrical resistivity of  $YBa_2(Cu_{1-x} La_x)_3O_{7-\delta}$  versus La concentration

## CONCLUSION

The study of transport properties of  $YBa_2(Cu_{1-x} La_x)_3 O_{7-\delta}$ in this work using temperature measurements indicated that the best superconducting properties can be obtained for substitution that excess dopant of La (x  $\leq 0.06$ ) results in a strong decoupling of chains and planes whereas optimal La doping ( $0.02 \leq x \leq 0.04$ ) can be adopted an alternate route to extensive oxygen anneaof Cu by La concentration within the range (x=0.02-0.04). The highest T<sub>c</sub> ( $\approx$ 90K), sharpest transition to superconductivity ( $\Delta$ =5 K) and lowest electrical resistivity ( $\rho$ = 9.2 $\mu$ \Omega.cm) are corresponding to the substitution samples of x=0.02 and 0.04. The highest T<sub>c</sub> could

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be related to more holes, more carrier concentration and more cooper pairs. Depression and degradation of the superconducting state as (x) increases above this range  $(0.04 \le x \le 0.06)$  is observed. This depression of T<sub>c</sub> due to increasing of (x) could be explained of chains and planes as a result of the excess of La dopant around the grain, also to the effect of the hole filling within this range  $(0.04 \le x \le 0.06)$ .

Therefore,  $T_c$  enhancement at low dopant La (x) and depressing it due to higher La (x) could be explained by the apex oxygen model and related to apical oxygen.

From this work, one can conclude that excess dopant of La  $(x \le 0.06)$  results in a strong decoupling of chains and planes whereas optimal La doping  $(0.02 \le x \le 0.04)$  can be adopted an alternate route to extensive oxygen anneal and may work as a catalist causes partial melting which enhances all the superconducting properties.

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