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ARTICLE

Partial Substitution Effects on the Structure and Electrical Properties of the High Temperature Superconducting System $Bi_{2-x}\ Tl_x\ Ba_{2-y}\ Sr_y\ Ca_2\ Cu_3\ O_{10+\delta}$

A. El Ali (Al-Dairy)[a], **A. D. Alazawi**[b], **Y. A. Hamam**[a] and **K. Khasawinah**[a]

[a] *Physics Department, Yarmouk University, Irbid, Jordan*
[b] *Department of Physics, University of Mousl, Iraq*

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Abstract: The partial substitution effects on the structure and electrical properties of the high temperature $Bi_{2-x}\ Tl_x\ Ba_{2-y}\ Sr_y\ Ca_2\ Cu_3\ O_{10+\delta}$ superconducting compounds have been investigated with x=0.00, 0.05, 0.10, 0.20 and y= 0.0, 0.1, 0.2, 0.3. X-ray diffraction studies showed that the structure was tetragonal with a=b=5.43Å, c=34.13Å for x=0, y=0. The structure almost remained tetragonal for all x values with y=0, and the c-parameter increased only for x=0.05 and then a decrease in the c-parameter with increasing x value was observed. On the other hand, for x=0 and y= 0.1, 0.2, 0.3 the structure was tetragonal with a slight decrease in the lattice parameters with increasing y. There was an observed change to orthorhombic phase with increasing both x and y at the same time.
The critical temperature was very sensitive to the thallium content x, in the absence of strontium, as it increased to 128K for small x but decreased noticeably with increasing x value. A similar behavior was observed for Sr content. However a large enhancement in the critical temperature was observed in the presence of lower concentrations of both thallium and strontium. Finally, the increase in the volume of the lattice or the oxygen content is associated with higher critical temperature values.
Key Words: HTSC, Partial Substitution, Crystal Structure, BSCCO, TBCCO.

Introduction

The discovery of superconductivity above 77K in Bi-Sr-Cu-O compound has been reported in [1,2]. The discovery of the high temperature superconducting system, Tl-Ba-Cu-O, was initially overshadowed by the discovery of the Bi-Ca-Sr-Cu-O system [3,4,5]. Extensive studies by several groups were carried out to study the structural, electrical, and magnetic properties of the Bi-based or Tl-based superconducting systems [6-12]. Some other workers studied the effects of doping these compounds with certain elements.[13-20]. However, many scientists concentrated on the Bi-based superconducting system since Tl is poisonous and needs special careful treatment. It was reported that doping the superconducting phases $Tl_2Ba_2CaCu_2O_8$ (Tl-2212) and $Tl_2Ba_2Ca_2Cu_3O_{10}$ (Tl-2223) with Pb or Li improved only their critical currents, and when the superconducting phases $Bi_2Sr_2CaCu_2O_x$ (Bi-2212) and $Bi_2Sr_2Ca_2Cu_3O_x$ (Bi-2223) were doped with Pb or Li, their critical temperatures and critical currents were improved [19-22]. It was also reported that the addition of specific small amounts of boron (B) to Tl-based superconductors increased the superconducting phase Tl-2223 and improved the critical temperature of the samples, while higher amounts of B eliminated Tl-2223 phase and reduced the Tl-2212 phase [23]. Furthermore, it was reported that fluorine addition to Tl-based compounds affected their properties drastically [24].

Corresponding Author: A. El Ali (Al-Dairy). Email: abedali@yu.edu.jo

Very few researchers worked on systems containing both Bi and Tl elements at the same time [21,22]. Epitaxial films of Tl-Bi-Sr-Ba-Ca-Cu-O were prepared using laser ablation and showed critical temperatures between 106-110K and critical currents at 77K of about 10^5 A/cm^2. Due to their high superconducting transition temperatures and high irreversible magnetic fields, Tl-based and Bi-based superconductors are very promising materials for making magnetic tapes above 77K.

This work aims at studying further superconducting samples containing both Bi and Tl. We have doped $Bi_2Ba_2Ca_2Cu_3O_{10}$ (Bi-2223 phase) with different amounts of Tl and Sr. The effect of this partial substitution of Tl and Sr on the structure and the electrical properties of the superconducting system $Bi_{2-x}\,Tl_x\,Ba_{2-y}\,Sr_y\,Ca_2\,Cu_3\,O_{10+\delta}$, with x=0, 0.05, 0.1, 0.2, and y= 0, 0.1, 0.2, 0.3 will be investigated in this paper.

Experimental Technique

The solid state reaction technique was used to prepare the superconducting samples $Bi_{2-x}Tl_xBa_{2-}Sr_yCa_2Cu_3O_{10+\delta}$ from appropriate amounts of the high purity oxides Bi_2O_3, Tl_2O_3, $BaCO_3$, $CaCO_3$, $Sr(NO_3)_2$, and CuO as starting materials. The high purity powders were mixed well and loaded in an alumina crucible which was then placed in a furnace. The temperature of the furnace was raised to 850 ^{o}C at a rate of 120 ^{o}C/hr. This material was sintered at 850 ^{o}C for 12 hrs then cooled down at a rate of 30 ^{o}C/hr in air to room temperature. The same heating process was repeated but with a flow of oxygen. The resulting material was then ground and pressed into 1gm pellets of approximately 12mm diameter and 1.2mm thickness. Fig.1 shows the annealing process carried on these pressed pellets in oxygen environment.

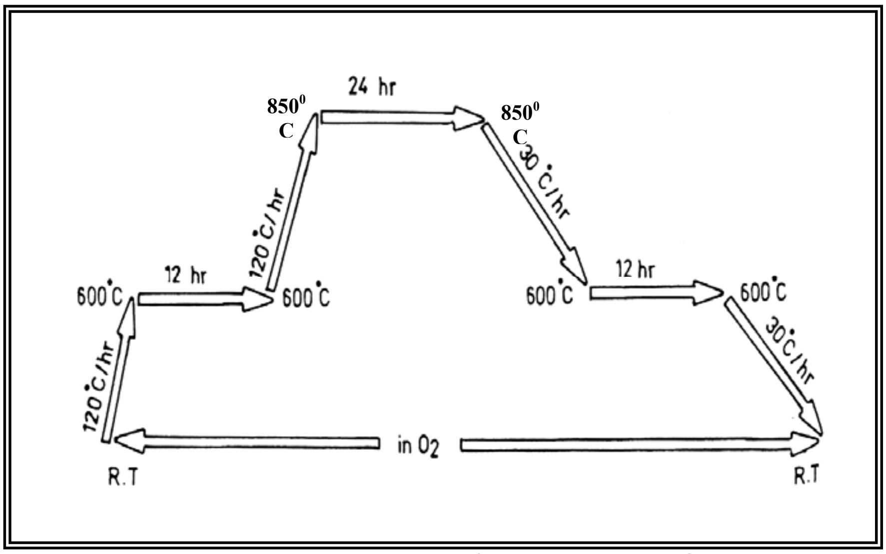

Fig. 1: Annealing process of pressed pellets in O_2

Standard four-probe-method was used to measure the resistivity (ρ) versus temperature (T). For each sample, we plotted resistivity (ρ) versus temperature (T) and the critical temperature (T_c) was determined. We have used the 50% rule to find T_c and not the onset value nor the T_c value for zero resistance. The oxygen content in each sample was measured by using iodometric titration method. Detailed experimental work for the determination of (T_c) and the oxygen content are described in references [12,15].

X-ray diffraction patterns for the $Bi_{2-x}Tl_xBa_{2-}Sr_yCa_2Cu_3O_{10+\delta}$ samples at room temperature were obtained using Phillips X-ray diffractometer with CuK_α source and λ=1.5418 Å. Using these patterns and a computer program the lattice parameters were then calculated.

Results and Discussion

Using the X-ray data obtained, the structures of $Bi_{2-x}\,Tl_x\,Ba_{2-y}\,Sr_y\,Ca_2\,Cu_3\,O_{10+\delta}$, high temperature superconducting samples prepared at 850^{0}C were studied. The structural calculations of the lattice parameters were carried with the help of a computer program written in BASIC. Fig.2 shows x-ray diffraction pattern of the Bi-2223 phase (x=0, y=0). It shows the existence of a single tetragonal phase with lattice parameters a=b=5.43Å and c=34.13Å.

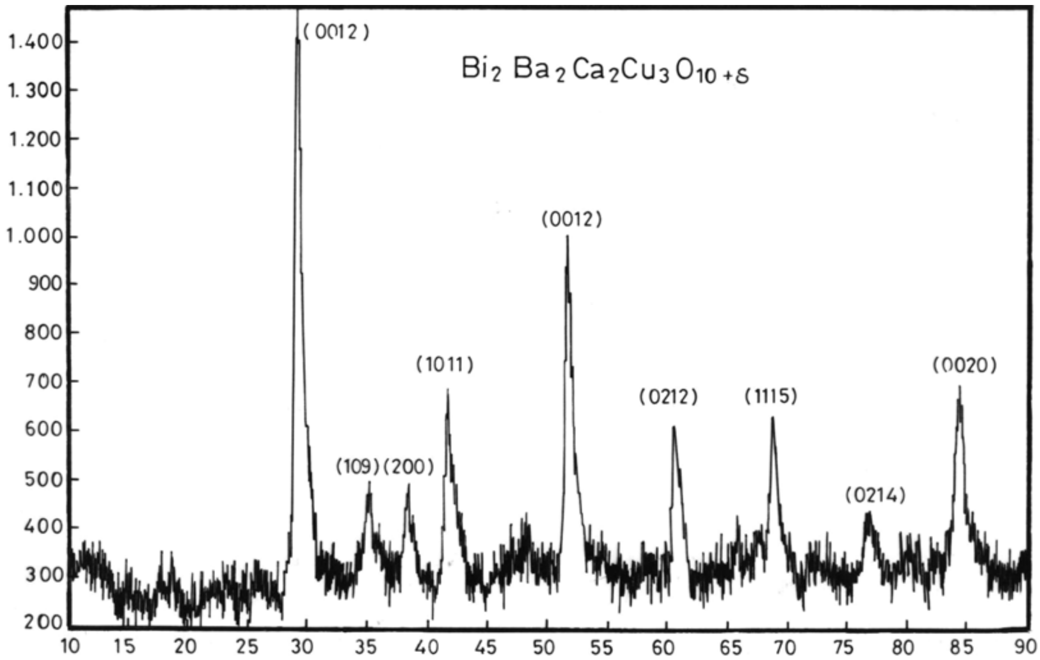

Fig (2): X – ray diffraction pattern of $Bi_2\ Ba_2\ Ca_2\ Cu_3\ O_{10+\delta}$ high temperature superconductor

Figures (3), (4), and (5) show x-ray diffraction patterns for the values: i) y=0 and x=0.05, 0.1, 0.2, ii) x=0, and y=0.1, 0.2, 0.3, iii) x=0.05 and y=0.1, 0.2, 0.3 respectively. From Fig.3, it was noticed that peak intensity decreases with increasing the value of the Tl content and small peaks like (0024) disappeared. In Table I we present the calculated values of lattice parameters. It is noticed that when no Sr is added and when Tl is partially substituted for Bi, with x =0.05, 0.1, the structure of the samples remained tetragonal with a slight increase in the c-parameter with decreasing x-values. The maximum increase in the c-parameter was up to 34.81Å for x=0.05. On the other hand, when x=0.2, the structure changed from tetragonal to orthorhombic phase with lattice parameters a=5.40 Å, b=5.21 Å, c=30.13 Å.

Fig.4 shows the x-ray pattern for the samples when only Sr is substituted for Ba with y=0.1, 0.2, 0.3 and x=0. The structure for samples with all y values remained tetragonal. It is noticed that there was a slight shift in the original peaks associated with the slight increase in the c-parameter. The maximum value calculated for the c-parameter was when y=0.1 and the lattice parameters were a=b≈5.40Å and c=34.72Å.

From Fig.5 and Table I, the structure of the samples changed in a different manner when Tl is substituted for Bi together with Sr substituted for Ba. For the samples with x=0.05 and y=0.1 the structure was tetragonal with a=b=5.46 Å, but for x=0.05 and y=0.2 or x= 0.05 and y=0.3 new peaks appeared which could be due to the formation of new phases of CuO and SrO. However, when x=0.1 and y=0.1 or x=0.2 and y=0.3, the structure of the samples was clearly orthorhombic.

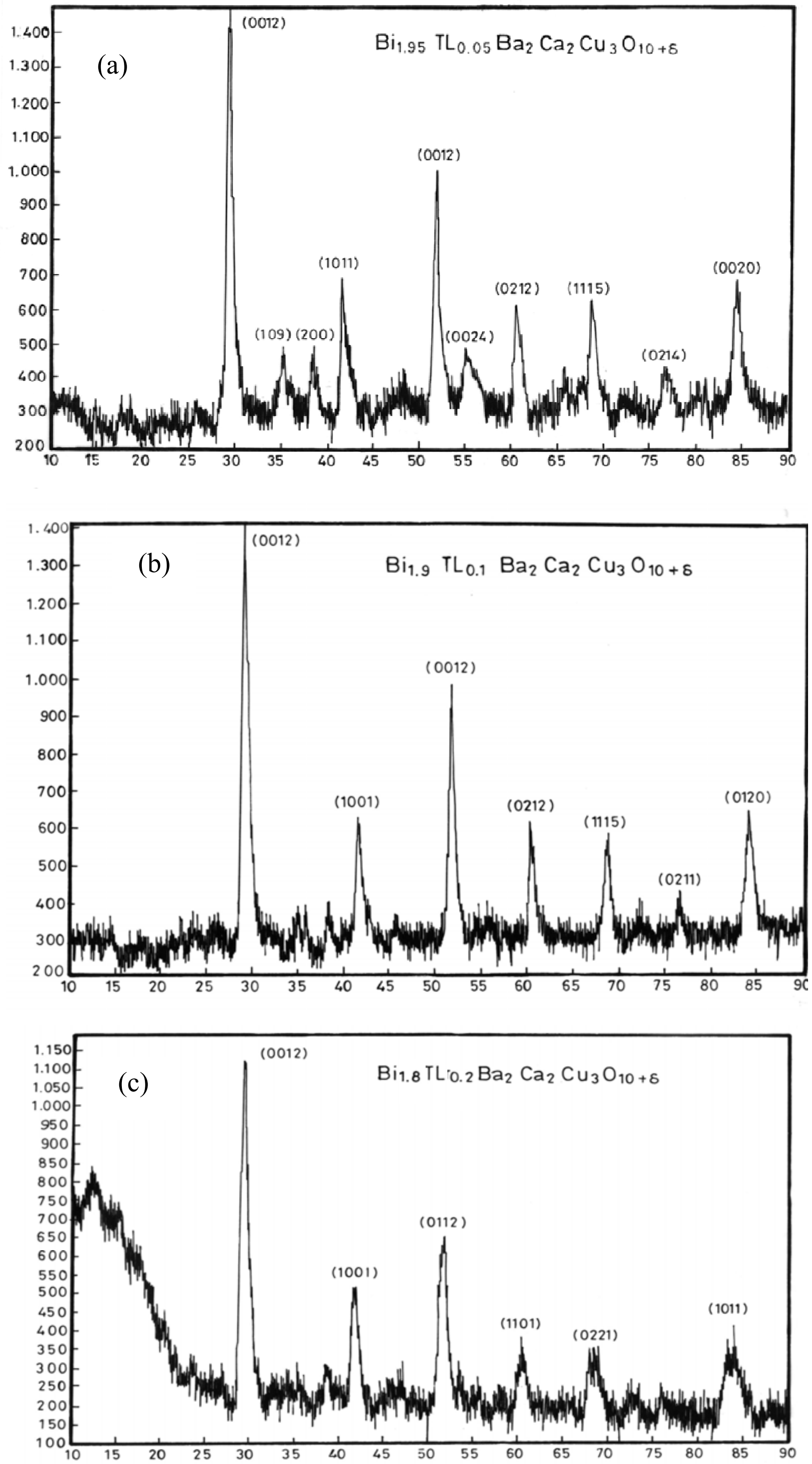

Fig (3): X – ray diffraction patterns for Bi_{2-x} Tl_x Ba_2 Ca_2 Cu_3 $O_{10+\delta}$ system for y=0 and: x = 0.05(a), x = 0.1(b), x=0.2(c).

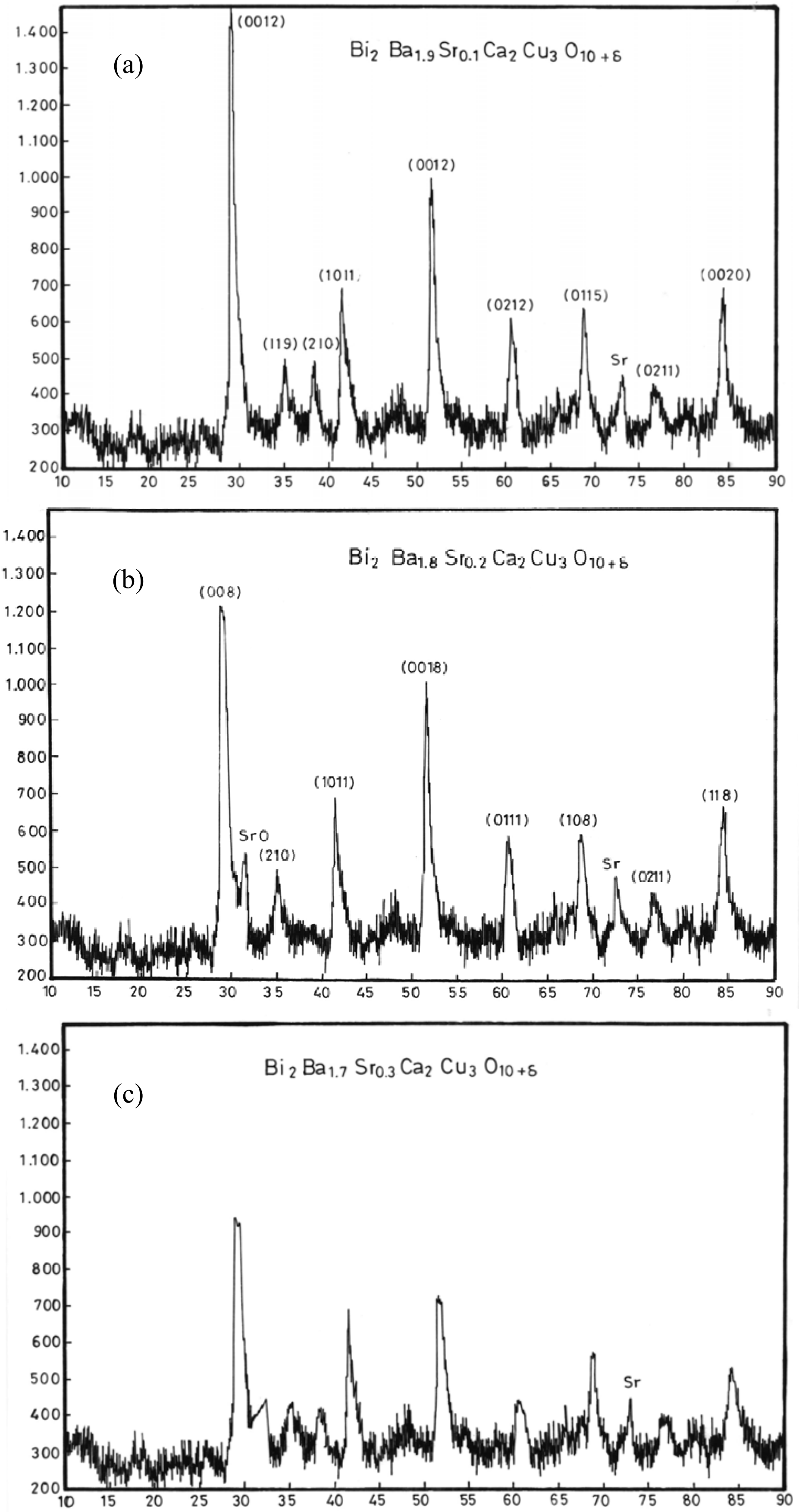

Fig (4): X – ray diffraction patterns for $Bi_2\ Ba_{2-y}\ Sr_yCa_2\ Cu_3\ O_{10+\delta}$ high temperature superconductor for x =0, and: y = 0.1(a), y = 0.2(b), y = 0.3(c).

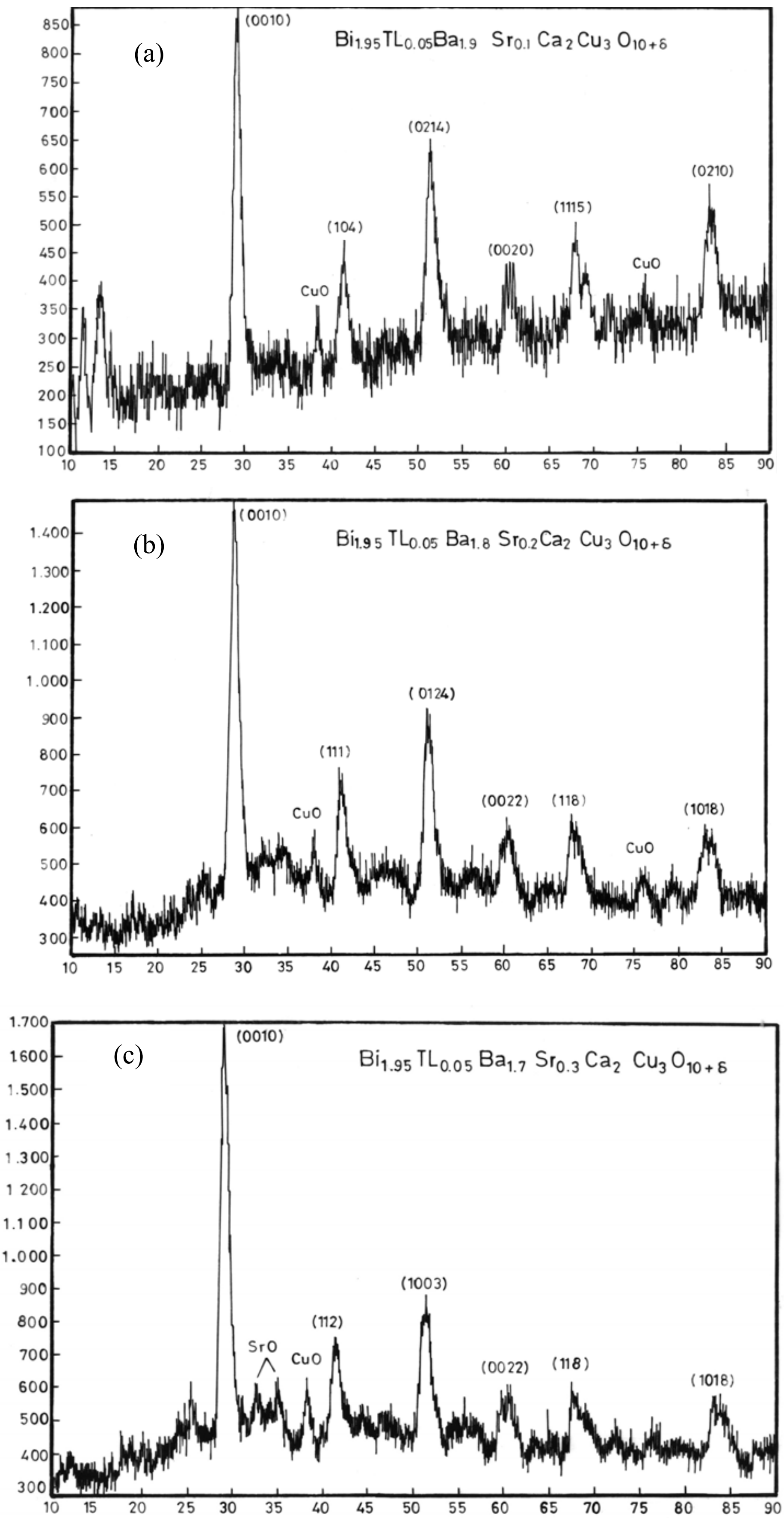

Fig (5): X – ray diffraction patterns for Bi_{2-x} Tl_x Ba_{2-y} Sr_yCa_2 $Cu_3O_{10+\delta}$ system for: a (x = 0.05, y = 0.1), b(x = 0.05, y = 0.2), and c (x = 0.05, y = 0.3).

Table I: Lattice parameters of the superconducting system $Bi_{2-x}Tl_xBa_{2-y}Sr_yCa_2Cu_3O_{10+\delta}$

Sample	Lattice Parameters		
	a (Å)	b (Å)	c (Å)
$Bi_2Ba_2Ca_2Cu_3O_{10+\delta}$	5.43	5.43	34.13
$Bi_{1.95}Tl_{0.05}Ba_2Ca_2Cu_3O_{10+\delta}$	5.43	5.43	34.81
$Bi_{1.9}Tl_{0.1}Ba_2Ca_2Cu_3O_{10+\delta}$	5.42	5.42	30.18
$Bi_{1.8}Tl_{0.2}Ba_2Ca_2Cu_3O_{10+\delta}$	5.40	5.21	30.13
$Bi_2Ba_{1.9}Sr_{0.1}Ca_2Cu_3O_{10+\delta}$	5.44	5.44	34.72
$Bi_2Ba_{1.8}Sr_{0.2}Ca_2Cu_3O_{10+\delta}$	5.41	5.41	30.18
$Bi_2Ba_{1.7}Sr_{0.3}Ca_2Cu_3O_{10+\delta}$	5.41	5.41	30.01
$Bi_{1.95}Tl_{0.05}Ba_{1.9}Sr_{0.1}Ca_2Cu_3O_{10+\delta}$	5.46	5.46	34.88
$Bi_{1.9}Tl_{0.1}Ba_{1.9}Sr_{0.1}Ca_2Cu_3O_{10+\delta}$	5.28	5.22	31.18
$Bi_{1.8}Tl_{0.2}Ba_{1.7}Sr_{0.3}Ca_2Cu_3O_{10+\delta}$	5.18	5.26	30.18

The measurements of resistivity versus temperature for samples with only Tl substituted for Bi; x=0.00, 0.05, 0.10, and 0.20, are shown in Fig.6. It was noticed that the critical temperature initially increased from 123K to 128K when x was increased from 0 to 0.05, and T_c started to decrease with increasing the content of the thallium, x=0.10 and 0.20. In Fig.7, we show the resistivity versus temperature for samples with only Sr partially substituted for Ba. Fig.8 shows resistivity versus temperature when both Tl and Sr are added for the smallest value of Tl, x=0.05, and with different amounts of Sr, y=0.1, 0.2, and 0.3. Figures (7) and (8) show a clear drop in the critical temperature with increasing the Sr. The values of the critical temperatures, T_c, determined from the Figures (6), (7), and (8) are summarized in table II.

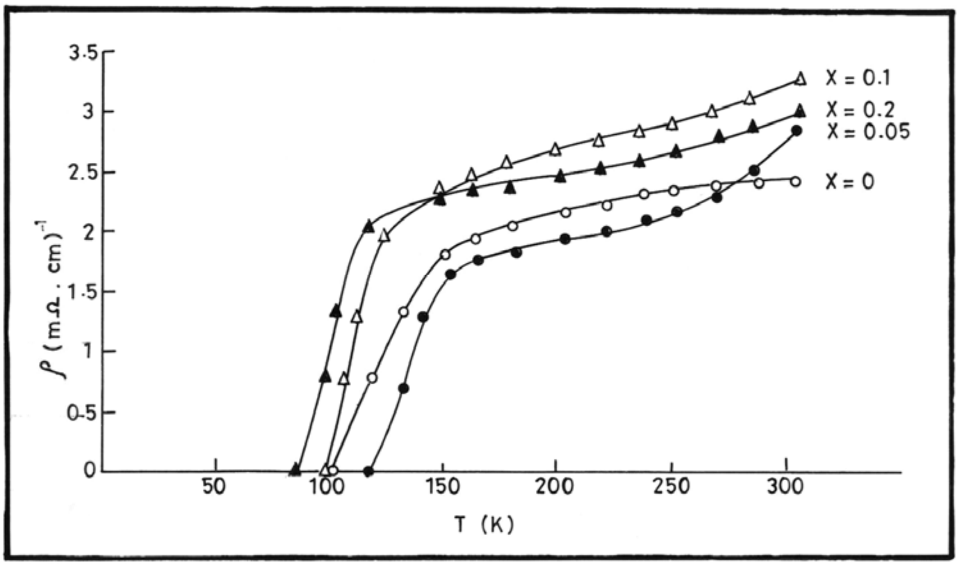

Fig (6): The resistivity versus temperature for the Bi_{2-x} Tl_x Ba_2 Ca_2 Cu_3 $O_{10+\delta}$ superconductors with x = 0, 0.05, 0.1, 0.2.

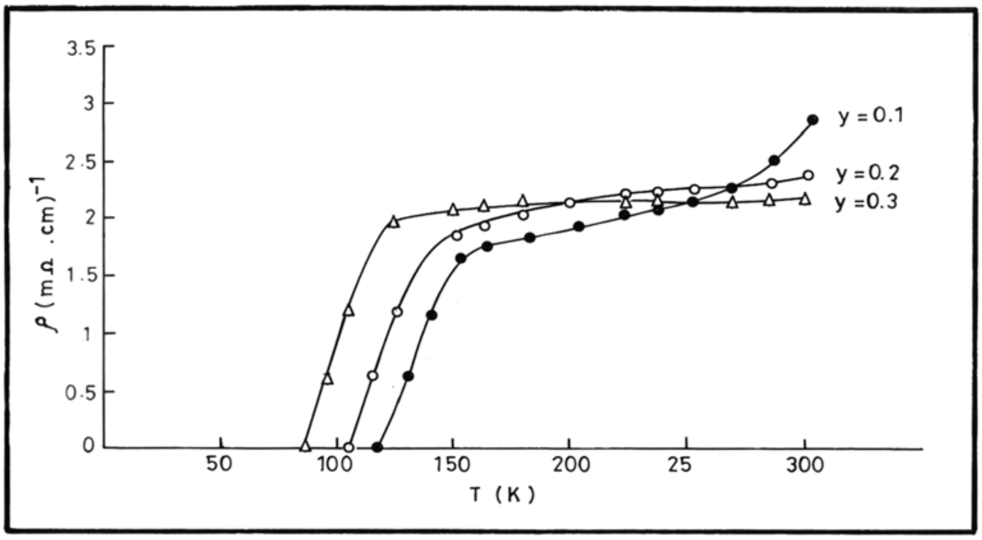

Fig (7): The resistivity versus temperature for $Bi_2\ Ba_{2-y}\ Sr_y\ Ca_2\ Cu_3\ O_{10+\delta}$ high temperature superconductor with y = 0.1, 0.2, 0.3.

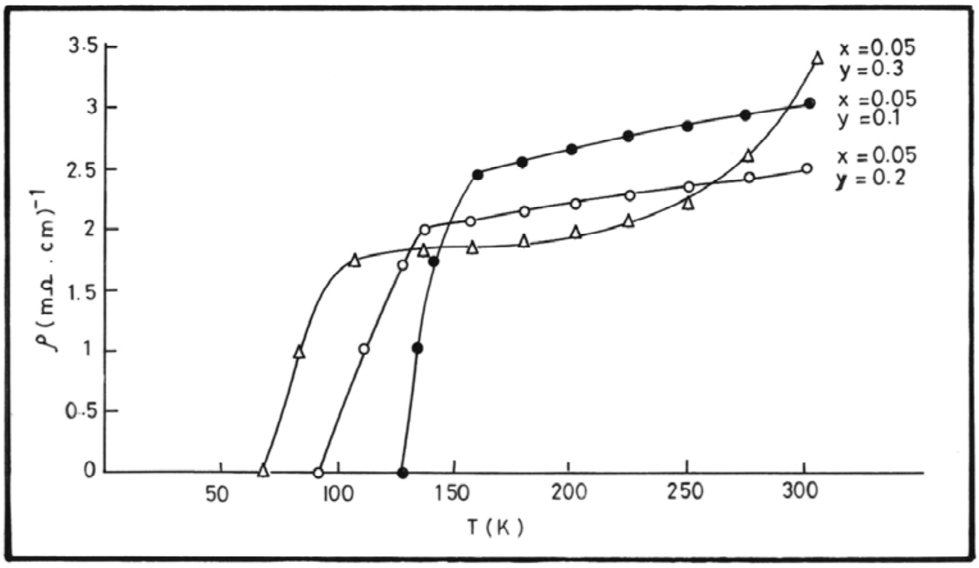

Fig (8): The resistivity versus temperature for $Bi_{2-x}\ Tl_x\ Ba_{2-y}\ Sr_y\ Ca_2\ Cu_3\ O_{10+\delta}$ for x = 0.05 and y = 0.1, 0.2, 0.3.

Table II: Critical temperatures measured and Oxygen contents for the superconducting system $Bi_{2-x}Tl_xBa_{2-y}Sr_yCa_2Cu_3O_{10+\delta}$

Sample	**Critical temperature T_c(^{o}K)**	**Oxygen content 10+δ**
$Bi_2Ba_2Ca_2\ Cu_3O_{10+\delta}$	123	10.22
$Bi_{1.95}Tl_{0.05}Ba_2Ca_2\ Cu_3O_{10+\delta}$	128	10.28
$Bi_{1.9}Tl_{0.1}Ba_2Ca_2\ Cu_3O_{10+\delta}$	108	10.21
$Bi_{1.8}Tl_{0.2}Ba_2Ca_2\ Cu_3O_{10+\delta}$	100	10.18
$Bi_2Ba_{1.9}Sr_{0.1}Ca_2\ Cu_3O_{10+\delta}$	125	10.26
$Bi_2Ba_{1.8}Sr_{0.2}Ca_2\ Cu_3O_{10+\delta}$	120	10.18
$Bi_2Ba_{1.7}Sr_{0.3}Ca_2\ Cu_3O_{10+\delta}$	110	10.14
$Bi_{1.95}Tl_{0.05}Ba_{1.9}Sr_{0.1}Ca_2\ Cu_3O_{10+\delta}$	140	10.38
$Bi_{1.9}Tl_{0.1}Ba_{1.9}Sr_{0.1}Ca_2\ Cu_3O_{10+\delta}$	112	10.26
$Bi_{1.8}Tl_{0.2}Ba_{1.7}Sr_{0.3}Ca_2\ Cu_3O_{10+\delta}$	95	10.21

It was observed that the critical temperature was enhanced drastically for small amounts of Tl and Sr and decreased significantly with increasing Sr content. The decrease in T_c values could be attributed to the change in structure from tetragonal to orthorhombic phase. The increase in T_c could mean that there was an increase in the mobility of electrons in the b-direction of Cu-O plane. Finally it is noticed that increasing the volume of the lattice cell, which could be caused by growth of a larger number of Cu-O layers in the unit cell, can lead to an increase in T_c.

The oxygen content in each sample was measured by using iodometric titration method and results are shown in Table II. It is noticed, from Table II, that higher critical temperatures are associated with higher values of oxygen content and the increase in the oxygen content might affect the electronic behavior by shifting the Fermi level.

Conclusion

The partial substitution effects on the structure & electrical properties of Bi_{2-x} Tl_x Ba_{2-y} Sr_y Ca_2 Cu_3 $O_{10+\delta}$ superconducting compounds have been investigated. X-ray diffraction studies showed that the structure is tetragonal with a=b=5.43Å, c=34.13Å for x=0, y=0. The structure almost remained tetragonal for all x values when no Sr was added. The c-parameter increased only for x=0.05 and then a decrease in the c-parameter with increasing x value was observed. On the other hand, for x=0 and y= 0.1, 0.2, 0.3 the structure was tetragonal with a slight decrease in the lattice parameters with increasing y. There was an observed change of structure to orthorhombic with increasing the content of both Tl and Sr resulting in a drop in T_c.

The critical temperature was very sensitive to the Tl content in the absence of Sr since it increased to 128K but deceased noticeably with increasing x value. A similar behavior was observed for Sr content. However a large enhancement in the critical temperature was observed in the presence of lower concentrations of both Tl and Sr. Finally it is noticed that the increase in the volume of the lattice, which could be caused by growth of a larger number of Cu-O layers in the unit cell, and the increase of the oxygen content are associated with higher critical temperatures.

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