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Effect of Partial substitution of Ag on the Structural and Electrical Properties of High Temperature $HgBa_2Ca_2Cu_3O_{8+\delta}$ Superconductor

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Abstract

The effect of the Ag partial substitution at Hg site in HgO_δ *layer on the structure ,*T_c *and oxygen content for Hg-1223 have been studied .High temperature superconductor composition* $Hg_{1-x}Ag_xBa_2Ca_2Cu_3O_{8+\delta}$*, with x=(0.0, 0.05, 0.1, 0.15, 0.2, 0.25 and 0.3) have been prepared using solid state reaction method. Annealing temperature 850* C^0 *for (24) hours with a rate of 5C°/min and under a pressure of 7 ton/*cm^2*. XRD analysis showed a tetragonal polycrystalline structure with high ratio of Hg-1223 phase and increase of c-axis lattice parameter with increase Ag rate. The surface morphology has been studied by using atomic force microscopes (AFM) in 3D. The results showed that all specimens have good crystalline and homogeneous surface. Also give a best Nano size value is 77.52 nm at x=0.3. Four probe technique is used to measured* T_c *. The highest* T_c *was found to be* T_c*=144 K for x=0.3, and oxygen contend was observed increase with increase Ag content.*

Keywords: *$HgBa_2Ca_2Cu_3O_{8+\delta}$, Superconductor, Solid state reaction metho , Structural Properties.*

1. Introduction

Superconductivity is a phenomenon, which was observed by Kamerlingh Onnes in 1911. When temperature decreases to below a critical value, electric resistance of a superconductor disappeared and the magnetic field is expelled [1]. The first member of $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ series, was $HgBa_2CuO_{4+\delta}$, fabricated by Putilin et. al. in 1993 [2]. In 1993 Schlling et. al. [3,4] concluded that the critical temperature was equal to 133 K for a compound $HgBa_2Ca_2Cu_3O_8$. This has achieved a significant jump in the critical temperature of superconducting compounds at high temperatures. The highest values of T_c which were calculated for $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ series were 97K [5] , 123K [6] 127K [7], and 135K [8], for Hg-1201, Hg-1234 phases, Hg-1212, and Hg-1223 respectively. The T_c value of Hg-1223 raise up to 164 K under high pressures of 30 GPa [9,10]. HBCCO superconductor phases most important series of all HTSC cuprates because of the high T_c and the extra oxygen existence appear by this series [11]. There are difficulties in preparation of Hg-based superconductors, because of the toxic mercury steam and the low decomposition temperatures of the compounds containing mercury and the relative instability of these materials. All cuprates are very sentient to carries doping and, it was found that the most efficient way to enhance the stabilization of the Hg-1223 phase is by partial substitution with cations having oxidation states higher than +2 i.e. higher than Hg^{+2} (e.g. Tl^{+3}, Re^{+4} etc) [12,13,14].

M. M. Abbas [15] reported that substitution of Cu ($0\leq x\leq 0.5$) at Hg site in HgO_δ layer in the Hg-1223 lead to enhanced T_c . The T_c $HgBa_2Ca_2Cu_3O_{8+\delta}$ was 118K .

While best value of T_c=153K and lattice parameters a=b= 3.841969, c= 15.79734 for sample with (x=0.3).

O. Babych et. al. [16] prepared 15% Pb, 5% Fe and 5% Cd doped mercury $HgBa_2Ca_2Cu_3O_{8+\delta}$ using Hg-free precursor $Ba_2Ca_2Cu_3O_{8+\delta}$ which obtained by sol-gel method. They noticed the superconducting plate grains size is (10–20) μm and there are non-superconducting phases of doped Hg-1223 ceramics surface. The Fe doped samples of $HgBa_2Ca_2Cu_3O_{8+\delta}$ exhibit decreasing T_c, with increase in the critical current in grain to 6800 A/cm^2.

M. Abdul-Nebi et. al. [17] studied $Hg_{0.5}Pb_{0.5-x}Sb_xBa_2Ca_2Cu_3O_{8+\delta}$ HTSC (x= 0, 0.10 and 0.15) prepared by solid state reaction method. They founded that the behavior of the composition which has no Sb is semiconductor. whereas the substitution of Sb content in the $Hg_{0.5}Pb_{0.5-x}Sb_xBa_2Ca_2Cu_3O_{8+\delta}$ convert from normal state (x=0.1) to superconducting state (x=0.15) with (T_c= 126K).

F. H. S. Eleuterio et. al. [18] analyzed granular

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composition and measure magnetic response of $Hg_{0.82}Re_{0.18}Ba_2Ca_2Cu_3O_{8.76}$ composition. The AC magnetic susceptibility of a powder sample have T_c=133 K with a particle size of 20 μm. The AC magnetic sample susceptibility shows two T_c at 133 and 98 K after crushed powder and sieved.

K. A. Jassim et.al. [19] prepared $Hg_{0.6}Tl_{0.4}Ba_2Ca_2(Cu_{1-x}Ag_x)_3O_{8+\delta}$ samples using solid state reaction for x=(0-0.7).XRD analysis exhibit a tetragonal structure for all specimens. Lattice constant and T_c are reduced with addition Ag.

In this paper we studied the effect of partial substitution of Ag on the oxygen content, structural properties and transition temperature of $HgBa_2Ca_2Cu_3O_{8+\delta}$ HTSC compounds.

2. Experimental methods:

The $Hg_{1-x}Ag_xBa_2Ca_2Cu_3O_{8+\delta}$ samples with different Ag (x=0.0, 0.05, 0.1, 0.15, 0.2, 0.25, 0.3) were prepared by using solid state reaction method using mixed oxides powder of HgO, AgO, BaO, CaO and CuO with a purity of 99.99%. The starting materials were mixed and ground in a gate mortar. Isopropanol is added during the grinding process to avoid the loss of parts of the powder during the grinding process. Then it is placed inside an electric oven at a temperature of 100 C^0 to remove the isopropanol alcohol. The powder was pressed into disc shaped pellets (1.5 cm) in diameter and (0.3) cm thickness, using hydraulic press under a pressure of (7 ton/cm^2) .The pellets were sintered in air at (850) C^0 for (24) hours with a rate of 5C°/min then cooled to room temperature by same rate of heating. Four probe method at temperature range (77-300)K was used to measure the resistivity(ρ) [20].Critical temperature (T_c) calculated using the relation:

$$\rho = (R * A)/L \ldots\ldots\ldots\ldots\ldots(1)$$

Where R is electric resistance, A is area of specimens and L is length of specimens. The structure properties of the 1223-phase was checked using X-ray diffraction technique using (Shimadzu XRD-6000) diffractometer with source Cu-Kα (1.5406 Å) radiation. The lattice parameters a, b and c were calculated by using d-values and (hkl) reflection of the observed XRD using standard card of Hg-1223 (ICDD-045-0615). The oxygen content determined using a chemical method called iodometric titration was described elsewhere [21]. The volume fraction for any phase determined by using the relation:

$$Vph = \frac{\sum I^{o}}{\sum I^{o} + \sum I1 + \sum I2 + \sum In} *100\% \ldots\ldots\ldots\ldots\ldots(2)$$

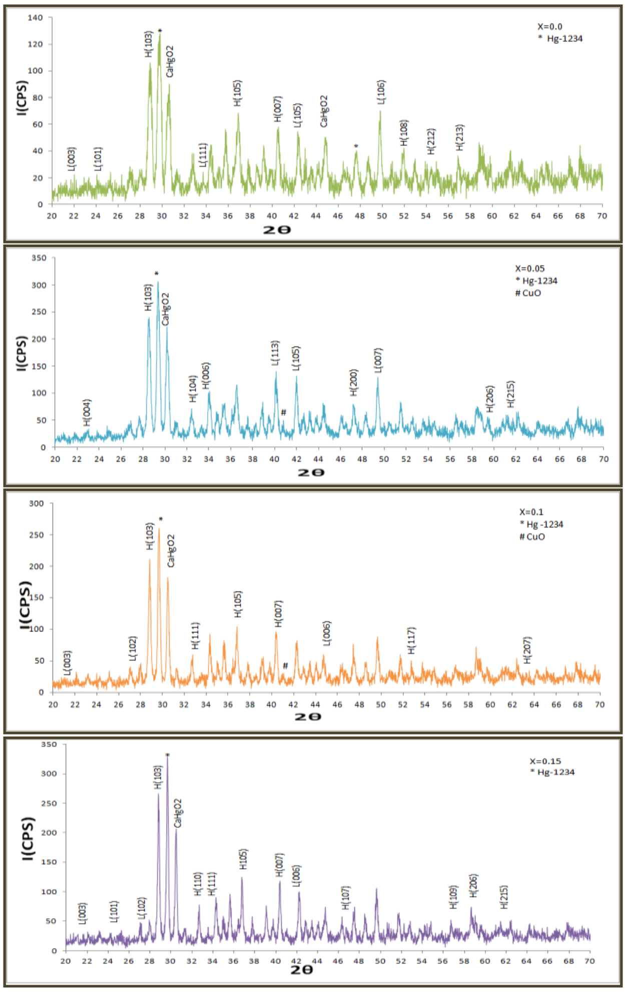
Where I° is the XRD peak intensity of the phase which were determined, I1,I2,…In are the peaks intensity of all XRD. The mass density (dm) calculated using the relation [22]:

$$d_m\left(\frac{gm}{cm^3}\right) = \frac{M_{wt}}{N_A \times V} \ldots\ldots\ldots\ldots\ldots(3)$$

Where N_A is Avogadro number (6.022*10^{23} mol^{-1}), M_{wt} is molecular weight, V is volume of unit cell which equal (a^2*c) for tetragonal system. Atomic Force Microscopy (AFM) micrographs were recorded by using scanning probe microscope type (SPM- AA3000), Contact mode, supplied by Angstrom Advanced Inc.

3. Results and discussion :

3.1. X-Ray Diffraction Results : The structures of $Hg_{1-x}Ag_xBa_2Ca_2Cu_3O_{8+\delta}$, with x=(0.0, 0.05, 0.1, 0.15, 0.2, 0.25, 0.3) were well established by X-ray diffraction analyses. Figure-I shows the X-ray diffraction patterns for samples which were prepared using solid state reaction. The positions and intensities of the diffraction peaks appear that samples mainly consist of a major 1223 phase and a small amount of a minor (1212 and 1234) phase and a small amount of impurity ($CaHgO_2$) which are agreement with reference [23]. The appearance of more than two phases could be related to the stacking faults along the c-axis. High-T_c phase (Hg-1223) appeared with increasing Ag content up to x=0.3.



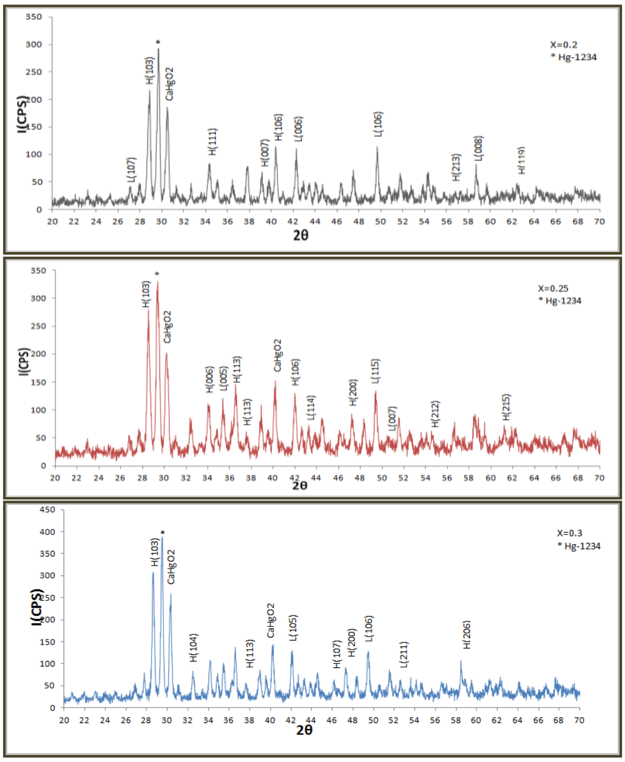


Figure I

XRD pattern of $Hg_{1-x}Ag_xBa_2Ca_2Cu_3O_{8+\delta}$, superconductor compounds with x=(0.0, 0.05, 0.1, 0.15, 0.2, 0.25, 0.3)

The lattice parameters a, b, c which presented in the Table-1 shows tetragonal symmetry structure for all our samples. The c-axis lattice constant increase with increase of Ag ratio. These results were almost identical to those reported in reference [24] and a-lattice parameter changes little. This result may be, because of the larger ionic radius of Ag^{+2} (1.26 A^0) than that of Hg^{+2} (1.02 A^0). This will be a driving force to the pairing generation of superconductor holes forming bosons which are the current carriers in our superconductor.

Table 1

Lattice parameters of $Hg_{1-x}Ag_xBa_2Ca_2Cu_3O_{8+\delta}$ compounds

x	a=b (A°)	c(A°)	$v(A^\circ)^3$	d_m (gm/cm^3)	V-ph(1223)
0	3.8383	15.7817	232.5652	6.2397	70.54
0.05	3.8385	15.7919	232.6185	6.2052	75.12
0.1	3.8384	15.7934	232.8632	6.1656	76.65
0.15	3.8381	15.7954	232.6528	6.1381	76.55
0.2	3.8388	15.7981	232.8202	6.1007	77.27
0.25	3.8379	15.7990	232.6580	6.0718	77.47
0.3	3.8380	15.8011	232.7099	6.0374	78.84

3.2. T_c and Oxygen Content Results : The change of T_c is related with δ. Oxygen content increase with increase Ag concentration as presented in the Table-2. Since the substitution of Ag in Hg site leads to produce chemical pressure in HgO_δ layer. generally increase the pressure mean increases the hole which lead to disorders in structure, and this disorder is found reflected the T_c (δ) behavior. At x=0.3, the oxygen content (δ) and T_c gets a maximum value. These results were almost identical to those reported in references [15].

Table 2

Oxygen content and critical temperature (T_c) for $Hg_{1-x}Ag_xBa_2Ca_2Cu_3O_{8+\delta}$ compounds

x	T_c(K)	δ
0	120	0.2931
0.05	126	0.3046
0.1	128	0.3078
0.15	133	0.3143
0.2	138	0.3189
0.25	140	0.4056
0.3	144	0.4423

Figure II shows the electrical resistivity as function of temperature for $Hg_{1-x}Ag_xBa_2Ca_2Cu_3O_{8+\delta}$, HTSC with (0≤x≤0.3). Samples shows a metallic behavior, followed by a superconductivity transition with T_c =120, 126, 128, 133, 138, 140, 144K respectively. This Due to firstly, the Ag partial substitution may lead to change in the carrier density of states indicating unlike magnetic moments, secondly, the positive contribution of the Ag element to the coupling process in the CuO layer which responsible for the superconductivity, leads to an increase in the c-lattice parameter, which leads to raise in T_c values.

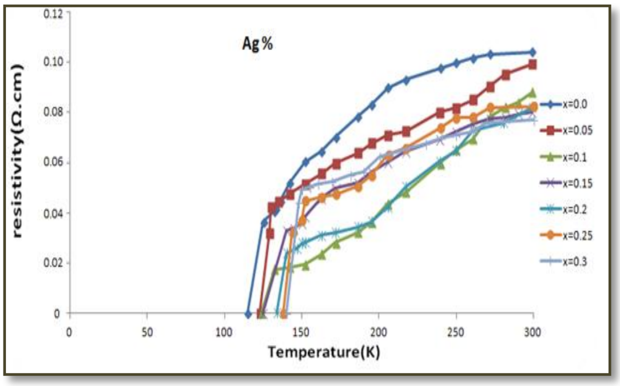


Figure II

The electrical resistivity vs. temperature behavior of $Hg_{1-x}Ag_xBa_2Ca_2Cu_3O_{8+\delta}$, for (0≤x≤0.3)

Figure III show T_c as a function of Ag content from 0.0 to 0.3. It is found that, with increasing of Ag contents, the T_c will increase.

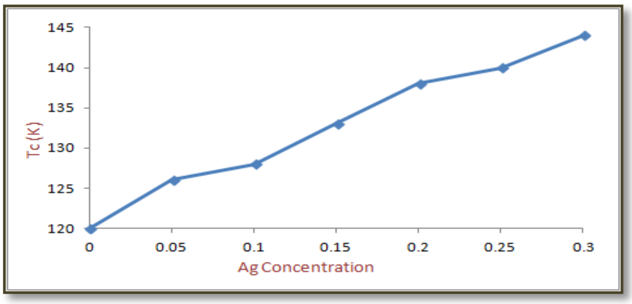
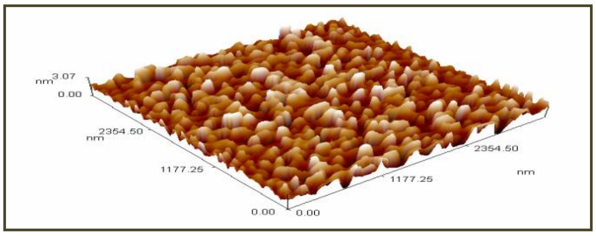


Figure III
The T_c vs. Ag concentration for ($0 \le x \le 0.3$).

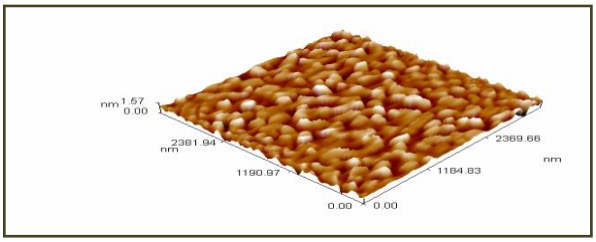
3.3. AFM Results : After the preparation of the samples by solid state interaction method. Using a nanometer agate mortar, the elements were converted to small dimensions and imaged by an atomic force microscope. Figure-IV represent 3-D AFM images of $Hg_{1-x}Ag_xBa_2Ca_2Cu_3O_{8+\delta}$, superconductor compounds for ($0 \le x \le 0.3$). It was noted that there are tortuosity, areas of high and low density with nano scale dimensions different from one site to another location within the sample .The Surface roughness and average diameter of $Hg_{1-x}Ag_xBa_2Ca_2Cu_3O_{8+\delta}$ compounds for ($0 \le x \le 0.3$) which presented in the Table-3 shows that all specimens have good crystalline and homogeneous surface give a best nano size value is 77.52 nm at x=0.3

Table **3**
Surface roughness and average diameter for $Hg_{1-x}Ag_xBa_2Ca_2Cu_3O_{8+\delta}$ *compounds*

X	**Surface roughness**	**average diameter**
0	0.522 nm	131.63 nm
0.05	0.202 nm	112.46 nm
0.1	0.418 nm	112.06 nm
0.15	0.84 nm	101.29 nm
0.2	0.266 nm	96.32 nm
0.25	0.62 nm	76.93 nm
0.3	0.668 nm	77.52 nm



(a)



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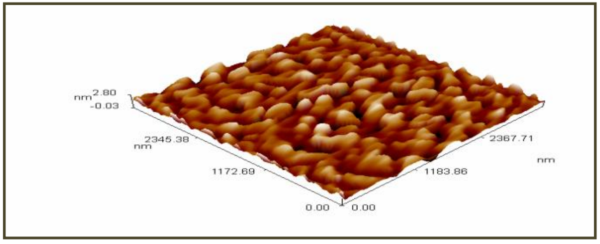
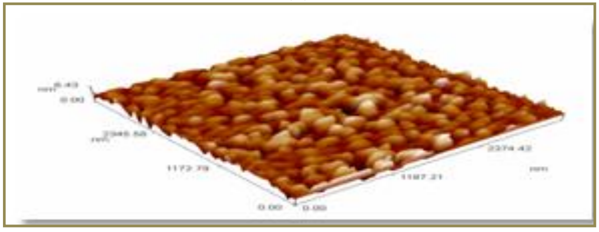
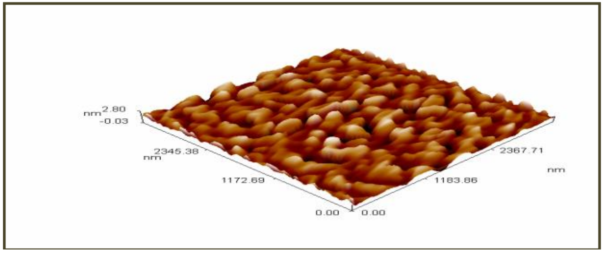


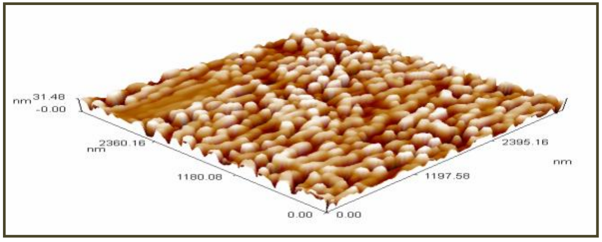
(c)



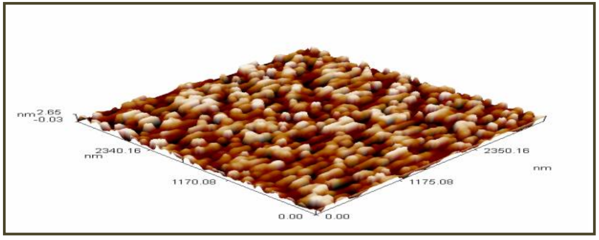
(d)



(e)



(f)



(g)

Figure IV
3D AFM images of $Hg_{1-x}Ag_xBa_2Ca_2Cu_3O_{8+\delta}$, superconductor compounds for (a) x=0, (b) x=0.05, (c) x=0.1,(d) x=0.15, (e) x=0.2, (f) x=0.25, (g) x=0.3.

Conclusions

In the present paper, we have investigated $Hg_{1-x}Ag_xBa_2Ca_2Cu_3O_{8+\delta}$, superconductor compounds for $(0 \leq x \leq 0.3)$ which prepared by solid state reaction method. XRD pattern analyses have showed tetragonal structure with high ratio of Hg-1223 superconductor phase, and, increase of the c-axis lattice constant for the samples doped with Ag as compared with this has no Ag content. The best value for x is that the best substitution ratio for Ag in the compound $Hg_{1-x}Ag_xBa_2Ca_2Cu_3O_{8+\delta}$, is at x = 0.3 where a high percentage of phase Hg-1223 appears. The T_c of un-doped Hg-1223 was (120K). The substitution of Ag in Hg for the compounds $Hg_{1-x}Ag_xBa_2Ca_2Cu_3O_{8+\delta}$, has exhibited a maximum value of T_c (144 K) at x=0.3 in addition, oxygen content δ have been found increases with increasing Ag concentration since the substitution produced of local pressure, hole carrier concentration, variation electronic state and its distribution. AFM results showed that the values of the surface roughness and average diameter that samples have good crystalline and homogeneous surface and give a best Nano size value is 77.52 nm at x=0.3

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