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Synthesis of High Temperature Superconductor $HgBa_{2-x}Y_xCa_{n-1}Cu_nO_{2n+2+\delta}$ Substituted with Yttrium for n=1 and n=2

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Abstract: ***The aim of this research is to study the effect of yttrium substitution on the pure phases superconductor $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ (n=1 & n=2). The solid state reaction method was used to prepare the samples in this work, followed firstly by the x-ray examination. The results of the XRD diffraction analysis showed that the structure for pure and doped phases was tetragonal. The change of yttrium concentration effects of all samples on the lattice parameters, electrical resistivity, the hardness, and the critical temperature Tc. The ratio of x=0.25 for n=2 gave the highest Tc. The results of the dielectric constant showed increasing with the Y addition increase for both phases (1 and 2) for high and low frequencies. While the hardness results showed a decrease in its values with Y addition increasing.***

Keywords: Superconductors, solid state reaction, dielectric constant, critical temperature

1. Introduction

Superconductivity is the property of completely electrical resistance lack in some materials when they cooled below a certain temperature called critical or transition temperature (Tc) which is vary from material to another. The explanation of this phenomenon was interpreted depending on the coupling of electrons (cooper pair) under some circumstances where they are follow together through the lattice [1,2]. The high temperature superconductivity has got the interest of researchers due to its extensive applications in electronics engineering and solid physics where some compounds have acquired the property of superconductivity at Tc=130 K or more, while the dream of the researcher is to rise the Tc to the room temperature [3,4, 5]. The superconducting properties in these materials depend widely on the structural properties like oxygen occupancy and charge carriers density [6]. Many researchers touched upon the historical development of the property of super electrical conductivity at high temperatures, also interested in studying the crystal structure and its impact on the degree of critical temperature Tc in some compounds such Hg-Ba-Ca-Cu-O system where the Tc reaches to 134K and under pressure, Tc in this compound could be raised to 160 K. [5,7]. Akram R. Jabur studied the effect of substitution Bi by Hg which gives a rise in Tc in the Bi_2-xHgxSr$_{2-y}$Ba$_y$Ca$_2$Cu$_2$O$_{10}$ compound [8]. The researcher Nihad Ali studied the substitution of Tl in Hg with different rates in the (Hg_{2-x} Tl_x $Sr_2Ca_2Cu_3O1_{0+\delta}$) compound type (2223) which leads to increase the critical temperature (142 K) and the grain size of superconducting phase [9]. Substituted Hg-based superconductor (($Hg_{1-x}M_x$)$Ba_2Ca_{n-1}Cu_nO_{2n+2+\delta}$) was studied by Dmitriy P. .This compound has record (Tc) value (166 K) at P = 23 GPa [10]. This research aims to prepare Hg based superconductor ($HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$) for phase one and two (n=1 and n=2) and study the effect of partial substitution of Y by Ba on the mechanical and electrical properties of these compounds and its transformation degree.

2. Experimental Part

Samples that submitted to the chemical formula: $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ for phase one and two and formula $HgBa_{2-x}Y_xCa_{n-1}Cu_nO_{2n+2+\delta}$ were prepared by using solid state reaction method to mix the calculated weights from pure oxides powders of HgO, BaO, CaO, Y_2O_3 and CuO by taking the molecular weight ratio to each element to the total molecular weight (for each formula) and specimen weight (2 gm) as shown in the table (1). The powders were mixed in agate mortar for 6 hours with addition of isopropanol ($C_2H_3O_5$) drops for more homogeneity. The mixture was dried using an oven at 100ºC then pressed into a disc shape pellets with diameter 5 mm and 2 mm thick, using hydraulic press under a pressure of 9 ton/cm^2 for one minute. The samples were sintered by using programmable controller furnace at 850 ºC for 12 hours with heating rate 5 ºC /min followed by cooling with same rate to the room temperature. Sample structure was obtained by using X-ray diffraction analysis ((XRD) type (Shimadzu), current (30 mA), voltage (40 KV) and (λ=1.540 A^0) and the crystal lattice parameters a, b and c were calculated using a program based on Full prof Suite toolbar. Vickers test method investigate the hardness for the samples and the standard four probe technique was used to calculate the electrical resistivity and then transition degree, the method of the work described in detail at reference [11]. The excess of oxygen content (δ) was determine by using a chemical method explained in detail at reference [12]. The dielectric constant was determined using (HF LCR meter) 6500p Series, Uk, frequency range from 20 Hz to 120 MHz.

3. Results and Discussion

The X-ray diffraction patterns results of pure and substituted Hg-based superconductors are shown in figure (1 for n=1) and figure (2 for n=2) respectively. The main phases in these samples are high Tc phase H (1212) phase and low Tc phase L (1201) phase and amounts of ($BaCuO_2$, CuO) as impurities in addition to small quantities of other unknown

impurities. As for the parameters of the crystal lattice a, b, c and c/a, the results as shown in the table (1) and (2) respectively show that for all samples the crystal structure is tetragonal. The superconducting transition temperature $T_{c\ (offset)}$ for pure and doped samples for (1212) phase is higher than $T_{c(offset)}$ for 1201 phase due to increasing Ca-O layer as compared with those have no this layer. Also the range of superconducting transition (ΔT) at (2012) phase was limited between (25 to 30) K as compared with superconducting transition (ΔT) results in the phase (1201) which has showed a wide transition width as shown in figure (3) and (4) respectively. This may due to increase the ratios of impurities presence as observed in the XRD patterns or the presence of the non-superconducting region [11]. At the sample $HgBa_2CuO_{4+\delta}$, the critical temperature Tc (off) was increased from 95K to 102 K with increasing the Y content at x= 0.1 and x= 0.15 which leads to increase the percentage of oxygen as observed in table (1) while at x=0.2 and x= 0.25, it can be observed the absence of superconductor transition as shown in figure (3) and from energy gap results at the table 3 (Energy gap $\Delta(0)=1.76k_BT_c$, k_B *is Boltzmann constant*[13]). The sample $HgBa_2CaCu_2O_{6+\delta}$ has the maximum Tc (off) at x= 0.25 (125K) and the minimum transition width ΔT = 25. Also we can observe the decreasing in c/a ratio with increasing the Y content. This may due to the ionic radii mutuality of Y ion (0.89 Aº) by Ba ion (2.2 Aº) which takes a compensate places through the structure of the unit cell rather than the interstitial sites [14]. The results of hardness Vickers are observed in the figure [5]. The results show in general, decreasing in the hardness value with increasing the Y_2O_3 content may due to the presence of weak grain boundaries while at the phase 1201, it has been show an increase in the value of hardness at x= 0.15 may due to high percentage of monocrystalline phase state without interference in the grain boundaries[15].

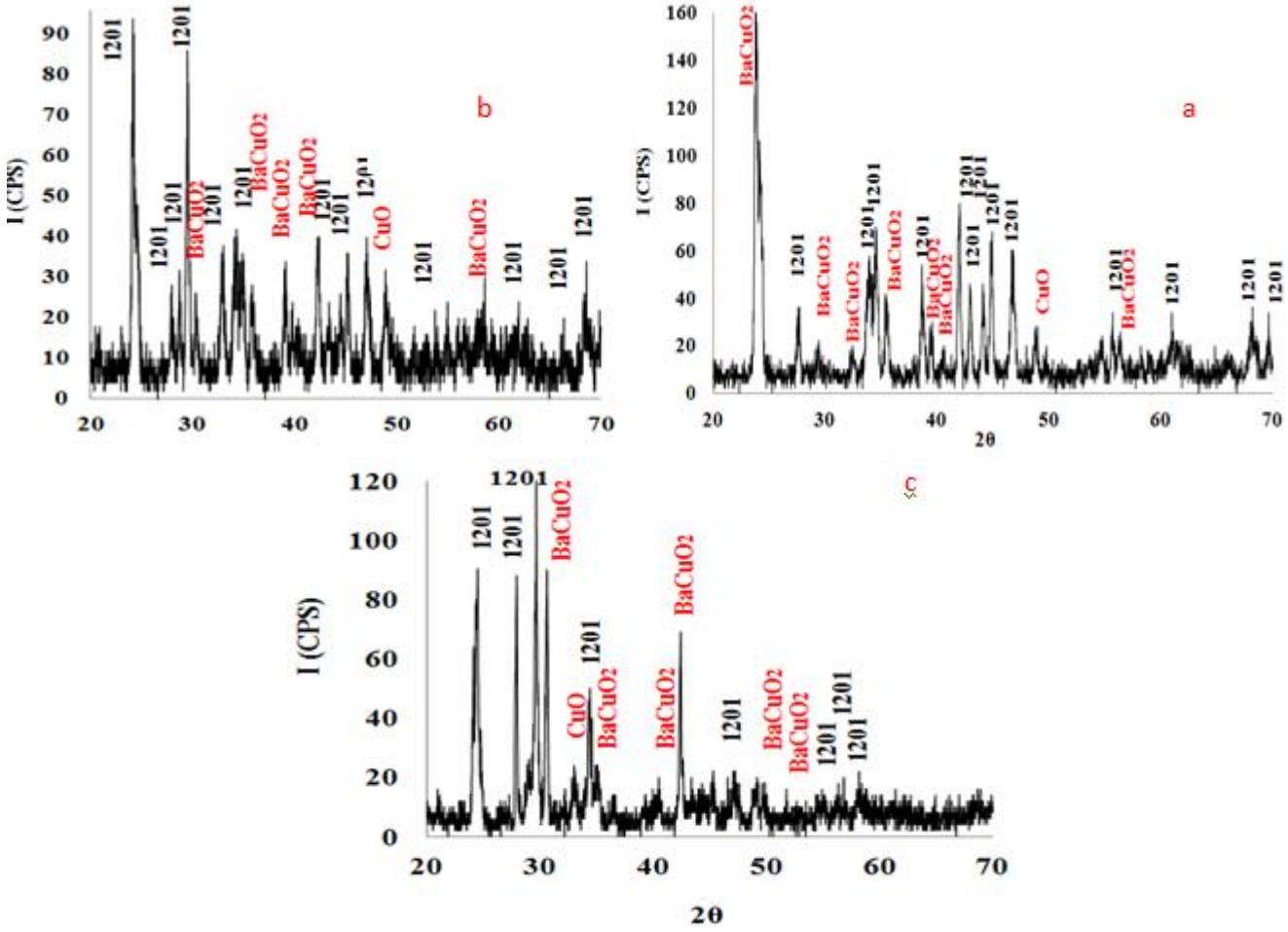

Figure 1: XRD patterns for the sample $HgBa_{2-x}Y_xCuO_{4+\delta}$, a when x=0, b when x=0.1, C when x=0.15. The main phase is 1201 and $BaCuO_2$ and CuO are the impurities.

Table 1: Values of lattice parameter, C/a, oxygen content (δ) and ρ_m for the sample $HgBa_{2-x}Y_xCuO_{4+\delta}$.

samples	x	a(Aº)	c (Aº)	c/a	$T_{C\ (mid)}$ (K)	ΔT_C (K)	ρ_m (gm/cm^3)	δ
$HgBa_2CuO_{4+\delta}$	0.0	7.510	12.728	1.694	123.5	57	0.8397	2.069
$HgBa_{2-x}Y_XCuO_{4+\delta}$	0.1	3.679	12.027	3.269	130	62	3.747	2.071
$HgBa_{2-x}Y_XCuO_{4+\delta}$	0.15	6.560	19.255	2.935	128	52	0.7405	2.098
$HgBa_{2-x}Y_XCuO_{4+\delta}$	0.2	------	------	------	insulator	-------	-------	-----
$HgBa_{2-x}Y_XCuO_{4+\delta}$	0.25	------	------	-------	semi	-------	-------	------

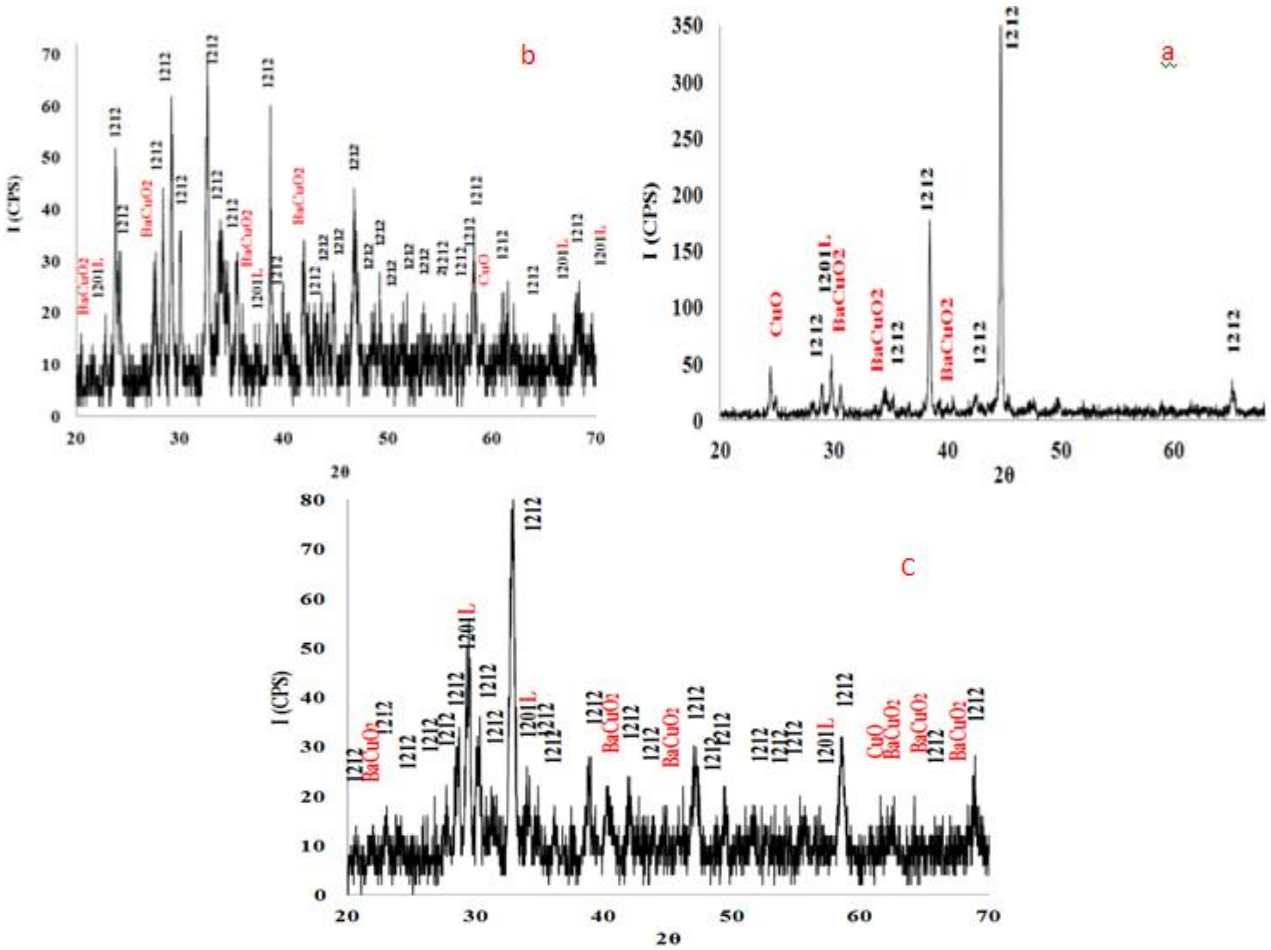

Figure 2: XRD patterns for the sample $HgBa_{2-}xYxCaCu_2O_{6+\delta}$, a when x=0, b when x=0.1, C when x=0.25. The main phases are H 1212, 1201L and $BaCuO_2$ and CuO are the impurities.

Table 2: Values of lattice parameter, C/a, oxygen content (δ) and ρ_m for the sample $HgBa_{2-x}Y_x CaCu_2O_{6+\delta}$.

samples	x	a(Aº)	c (Aº)	c/a	$T_{C\,(mid)}$ (K)	ΔT_C (K)	ρ_m (gm/cm^3)	δ
HgBa2CaCu2O6+δ	0.0	5.995	18.353	3.0615	135	30	1.1194	2.089
HgBa2-xYXCaCu2O6+δ	0.1	7.557	19.315	2.5558	137	30	0.676	2.045
HgBa2-xYXCaCu2O6+δ	0.15	------	------	------	insulator	-----	-------	-----
HgBa2-xYXCaCu2O6+δ	0.2	------	------	------	semi	-----	-------	-----
HgBa2-xYXCaCu2O6+δ	0.25	8.253	10.140	1.2285	137.5	25	1.095	2.059

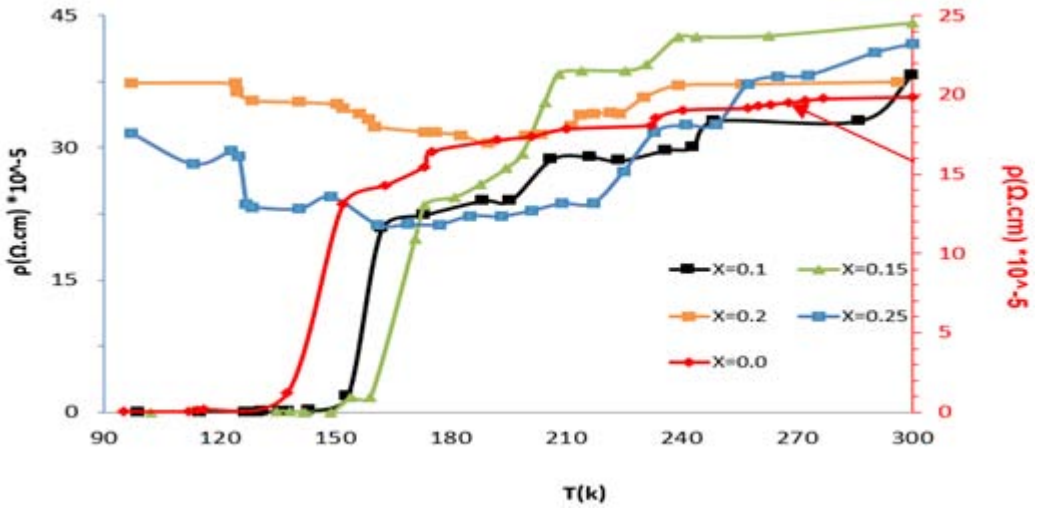

Figure 3: The resistivity dependence on Temperature for the sample $HgBa_{2-x}Y_xCuO_{4+\delta}$ at x= 0.0, 0.1, 0.15, 0.2, 0.25

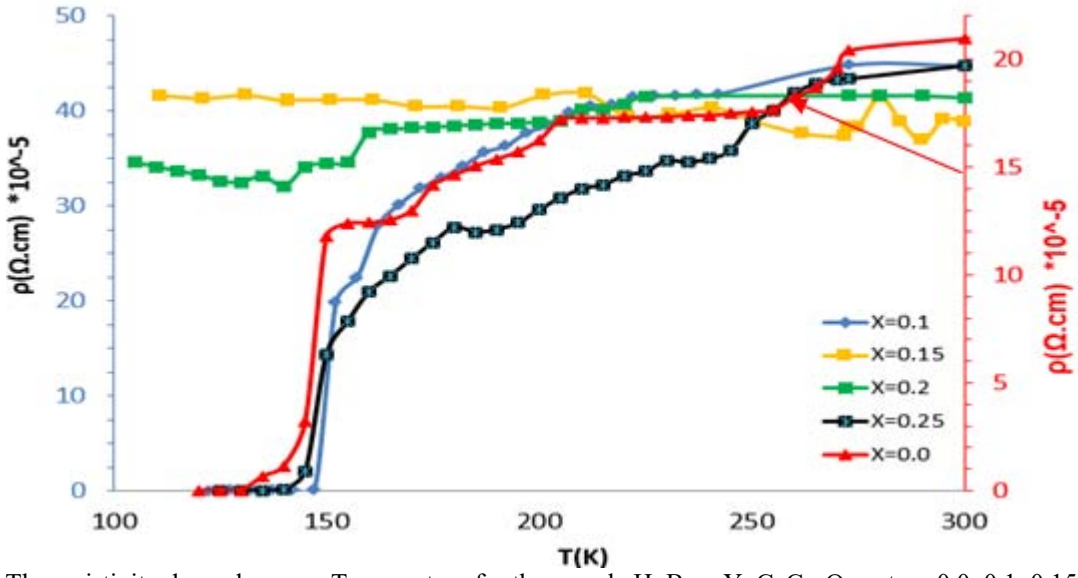

Figure 4: The resistivity dependence on Temperature for the sample $HgBa_2xYxCaCu_2O_{6+\delta}$ at x= 0.0, 0.1, 0.15, 0.2, 0.25.

Table 3: The values of energy gabs for the samples at n=1 and n=2

Samples (1201)	Energy gap	Samples (1212)	Energy gap
$HgBa_2CuO_{4+\delta}$	0.028924	$HgBa_2CaCu_2O_{6+\delta}$	0.036536
$HgBa_{1.9}Y_{0.1}CuO_{4+\delta}$	0.030142	$HgBa_{1.9}Y_{0.1}CaCu_2O_{6+\delta}$	0.037144
$HgBa_{1.85}Y_{0.15}CuO_{4+\delta}$	0.030446	$HgBa_{1.85}Y_{0.15}CaCu_2O_{6+\delta}$	3.309413
$HgBa_{1.8}Y_{0.2}CuO_{4+\delta}$	5.5131	$HgBa_{1.8}Y_{0.2}CaCu_2O_{6+\delta}$	0.646616
$HgBa_{1.75}Y_{0.25}CuO_{4+\delta}$	0.143951	$HgBa_{1.75}Y_{0.25}CaCu_2O_{6+\delta}$	0.038058

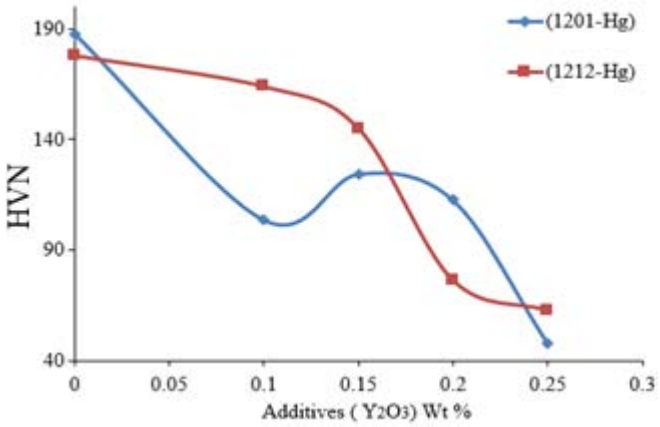

Figure 5: Vickers Hardness versus Y_2O_3 addition for the samples at n=1 and n=2

As a function of the changes in the applied frequencies on the samples in the range from 50 Hz to 1 MHz, we can observe the changes in the dielectric constant values (έ) as shown in the figures (6 , a and b) respectively, we will discuss the behavior of superconductor samples only. The figure (6,a) shows a rise in (έ) values at low frequencies from 6.139 at the sample $HgBa_2Cu_2O_{4+\delta}$ (x= 0.0) to 103.342 at the sample $HgBa_{1\cdot85}Y_{0\cdot15}CuO_{4+\delta}$ (x=0.15) also increasing in this values at 1MHz with increasing Y_2O_3 addition from 4.871 (at x=0) to 58.477 (at x=0.15). Also the values of dielectric constant for the samples at n=2 (fig. 6, b) shows an increase in their value from 17.569 at 50 Hz for the sample ($HgBa_2CaCu_2O_{6+\delta}$, 1212 x= 0.0) to 80.123 (for the sample $HgBa_{1.75}Y_{0.25}CaCu_2O_{6+\delta}$, 1212, x= 0.25) while the results at 1 MHz were increased from 8.072 for the sample $HgBa_2CaCu_2O_{6+\delta}$ (x=0.0) to 34.626 for the sample $HgBa_{1.75}Y_{0.25}CaCu_2O_{6+\delta}$ (x= 0.25). So in general, the dielectric constant decreased with frequency increasing because of the charge carriers at high frequencies cannot orient themselves by pursuing the frequency of the applied electric field where they need longer period of time [16]. The maximum results of the (έ) for both phases 1201 and 1212 was at the maximum content of (Y) ions. This is may be due to the agglomeration of yttrium particles in the crystal phase at higher concentration of Y ions which occupies partial sites in the crystal lattice more than replacement sites with Ba [17].

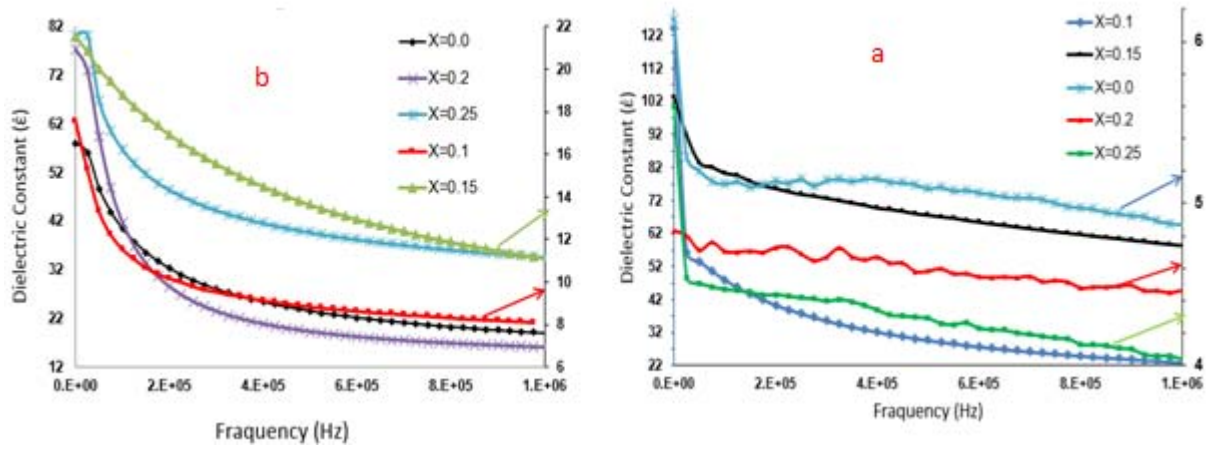

Figure 6 (a) Variation of dielectric constant (έ) of $HgBa_{2-x}Y_xCuO_{4+\delta}$ versus frequency for x = (0.0, 0.1, 0.15, 0.2, 0.25), **(b)** Variation of dielectric constant of $HgBa_{2-x}Y_xCaCuO_{6+\delta}$ versus frequency for x = (0.0, 0.1.0.15, 0.2, 0.25).

4. Conclusions

Yttrium substituted – Hg based superconductor ($HgBa_{2-x}Y_xCa_{n-1}Cu_nO_{2n+2+\delta}$ for n=1 and n=2) were synthesized successfully by solid-state reaction method and characterized by different experimental techniques, structural, electrical, superconducting and mechanical properties. The crystal structure was Tetragonal for pure and doped samples for both 1201 and 1212 phase. The maximum value of the critical temperature was at the phase1212 when yttrium content was 0.25. The hardness of the samples was decreased with increasing the (Y) addition for both phases, while the best value was at the pure phase 1201. Dielectric constant has been decreased with the increasing the frequency and increasing with increase the (Y) addition, the highest value at high frequency was at the phase 1201 when (Y) content was 0.15 wt%.

5. Acknowledgment

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