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## Effect of Nickel Substitution On Structural and Electrical Properties of $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_2\text{Cu}_{3-y}\text{Ni}_y\text{O}_{8+\delta}$ Superconductor Composite

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# Effect of Nickel Substitution On Structural and Electrical Properties of $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_2\text{Cu}_{3-y}\text{Ni}_y\text{O}_{8+\delta}$ Superconductor Composite

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**Abstract:** six specimens of the  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_2\text{Cu}_{3-y}\text{Ni}_y\text{O}_{8+\delta}$  ( $y=0.2,0.4,0.6,0.8,1.0$ ) superconducting compound were prepared by solid state reaction method, with sintering temperature equal to 1123K for 24 hours. The electrical resistivity was examined by the four probe technique, It was found that all the specimens have metallic behavior and increasing the critical temperature with increasing nickel concentration. The optimum critical temperature  $T_c$  was found equal to 137.5 K when the value of nickel concentration  $y=0.8$ . The structural of all the specimens studied by using XRD analysis, this examination showed the crystal structure of the specimens is polycrystalline with tetragonal structure. The volume fraction of high phase  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_2\text{Cu}_{3-y}\text{Ni}_y\text{O}_{8+\delta}$  superconducting (Hg-1223 phase) increase up to 81.737% by increasing Ni concentration for  $y=0.8$ , The increasing in nickel concentration causes due to increase the high-phase 1223 and increment of c parameter.

**Keywords:** Superconductors, transition temperature  $T_c$ , Hg-Ba-Ca-Cu-O oxide, XRD, AFM.

## 1. Introduction

High temperature superconductivity is a standout amongst the most vital research zones and Hg-Ba-Ca-Cu-O oxides are the most contemplated mixes around there. The motivation behind physicist is to acquire high temperature superconductors at room temperature. In the event that this is accomplished, numerous advances which will ease life in numerous fields, for example, transportation, vitality aura and append incredible significance to manner might be made. Mercury in HBCO superconductor mixes have consolidated  $\text{CuO}_2$  layers in these mixes and have expanded the basic temperature. Be that as it may, copper and oxygen has demonstrated superconductor properties when joined with Mercury.

High-temperature superconductors are materials that have a superconducting transition temperature  $T_c$  above liquid nitrogen point (77K). The high  $-T_c$  superconductor Hg-based,  $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$  has attracted much attention due to the highest transition temperature superconducting ( $T_c$ )  $\sim 135\text{K}$  at ambient conditions [1,2]and modest flux pinning strength [3], particularly at temperature above 77K.

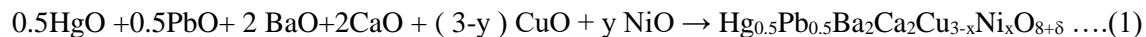


The structure of a Hg-Ba-Ca-Cu-O superconductors oxide are closely related to structure of perovskite, The series of compounds  $\text{HgBa}_2\text{Ca}_n\text{Cu}_{n+1}\text{O}_{2n+4}$  where  $n$  is an integer, are prototypes for the Hg family of superconductors. The first three members of the family, with  $n = 0, 1, 2$ , are often referred to as Hg-1201, Hg-1212, and Hg-1223, respectively. The lattice constants are  $a = 3.86$  for all of them, and  $c = 9.5, 12.6, \text{ and } 15.7$  for  $n = 0, 1, 2$ , respectively[4].

The extensive trouble experienced in the amalgamation of Hg bearing HTSC stages including Hg-1223 due to their outrageous affectability towards pollution from stickiness and carbon dioxide. The Hg-1223 samples are known to debase quickly after blend. In perspective of this, huge endeavors have as of late been made to enhance the security of the Hg bearing HTSC stages, especially the Hg-1223 stage. It is currently realized that the best method to enhance the solidness of the Hg-1223 stage is through reasonable cationic substitution for Hg. Ordinarily suited cations are those having oxidation states higher than that of  $\text{Hg}^{+2}$  more prominent than  $+2$ [5-10]. They get more oxygen in the oxygen insufficient HgO  $\delta$  layer prompting stage dependability. The higher oxidation state cations likewise lead to opening improvement in the gap insufficient as developed Hg-1223 stage, along these lines delivering ideal basic change temperature ( $T_c$ ). In this paper we ponder the successful of substitution Ni on electrical and basic properties for  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_2\text{Cu}_{3-y}\text{Ni}_y\text{O}_{8+\delta}$  superconductor.

## 2. Experimental

The  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_2\text{Cu}_{3-y}\text{Ni}_y\text{O}_{8+\delta}$  with ( $y=0.2, 0.4, 0.6, 0.8, 1.0$ ) compounds were these specimens synthesized by procedure of (SSR), the required of amounts powders of pure oxides. the used high purity of oxides is almost pure (99.99% buy from England/GCC) of HgO, PbO, NiO, BaO, CaO and CuO., the molecular weights are calculated according to the following chemical formulas:



The powders were dried under 393K for 1.5h, to get rid of the water vapor, and then the weight of all the elements reacted by a sensitive digital balance to the extent of four decimal places. The powders were mixed and grinded by a 6-hour spiral electric mixer for optimal homogenization and for accurate powders. The resulting powders were dried in a drying oven under 393K for 1.5 h. The resulting powder mixtures were then compressed using a hydraulic piston under pressure of 7 ton /  $\text{cm}^2$  for two minutes, in the form of discs with a diameter of 1.5cm and a thickness of 0.2-0.3 cm. the specimens sintering at 1123K for time (24 hours) with a rate of (278K/min) to obtain a bonding material and to ensure optimum propagation between the atoms gradually. After that the specimens were cooled to room temperature at the same heating rate.

The structure of the prepared specimen was obtained by using x-ray diffractometer (XRD) type (Philips) have the following features, the source  $\text{Cu}_{k\alpha}$  current (20 mA), voltage (40 KV) and  $\lambda=1.5405 \text{ \AA}$ . Phase transformation for many composition was studied by using XRD to get the structure properties. The volume fraction of any phase ( $V_{\text{phase}}$ ) in the sample were determined by using the relation [11-13]:

$$V_{\text{phase}} = \frac{\sum I_a}{\sum I_1 + \sum I_2 + \dots + \sum I_n} * 100\% \dots \dots \dots (2)$$

Where  $I_a$  is the XRD peak intensity of the phase which were determined,  $I_1, I_2, \dots, I_n$  are the peaks intensity of all XRD. A computer program was established to calculate the lattice parameters a,b,c this program based on Cohen's least square method [12,15]

The density of the cell unit was measured, and then the percentages of the phases formed in the specimen. Vickers test method investigate the hardness for the specimens 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 [7]. The excess of oxygen content ( $\delta$ ) was determine by using a chemical method explained in detail at reference [12].

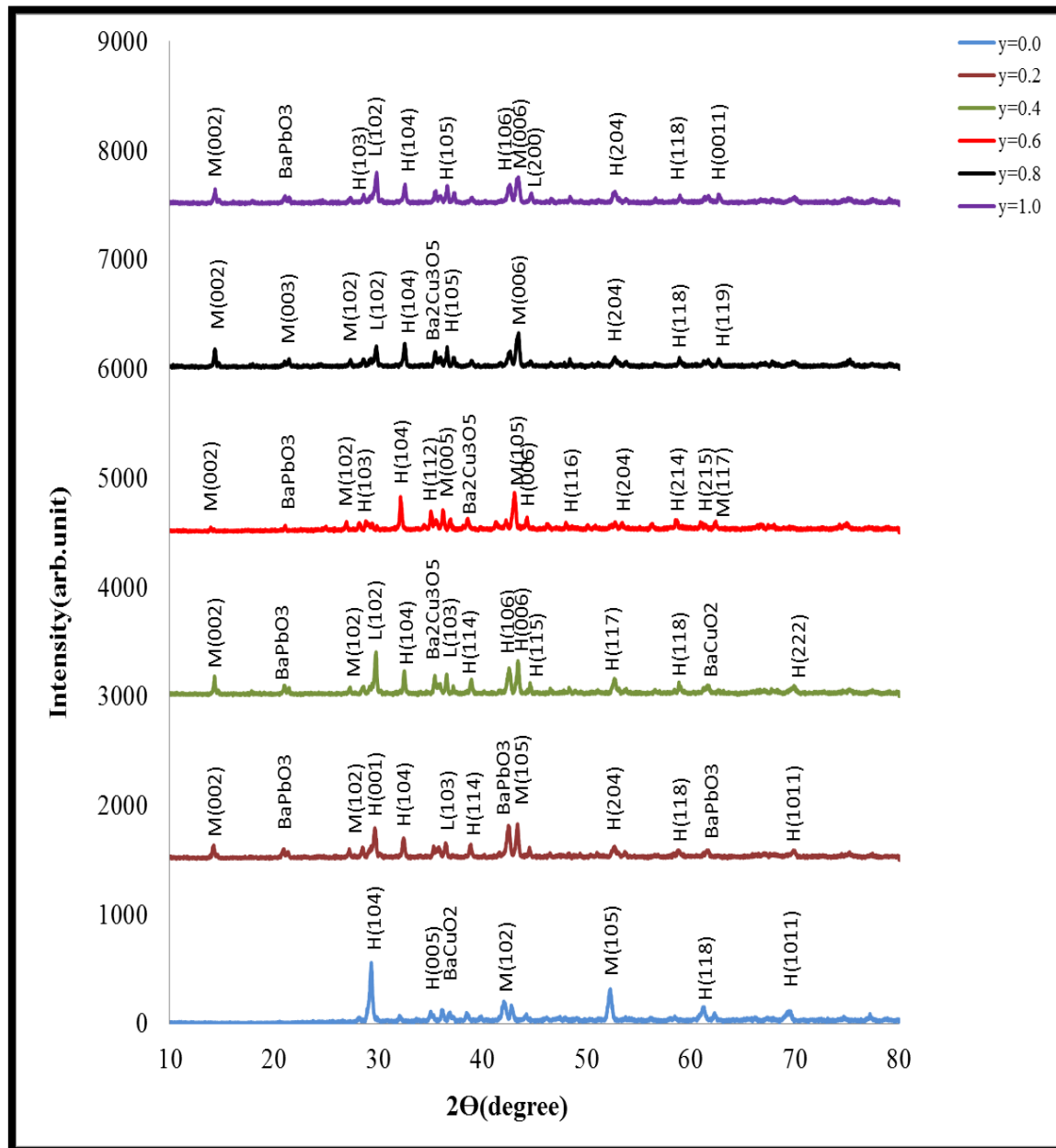
### 3. Results and Discussions

The X-ray diffraction patterns between  $10^\circ$  and  $80^\circ(2\theta)$  for the  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_2\text{Cu}_{3-y}\text{Ni}_y\text{O}_{8+\delta}$  bulk specimens with different Nickel concentration ( $y=0.2,0.4,0.6,0.8,1.0$ ) are depicted in Fig. 1. The corresponding hkl Miller indices belonging to  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_2\text{Cu}_{3-y}\text{Ni}_y\text{O}_{8+\delta}$ ,  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_1\text{Cu}_{2-y}\text{Ni}_y\text{O}_{6+\delta}$  and  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Cu}_{1-y}\text{Ni}_y\text{O}_{4+\delta}$  main lines are shown in the diagram. The results show that the all specimens in our work exhibit the polycrystalline superconducting phases. Specimens with increasing the values of Ni display the single-phase and a small amount of impurity as shown in figure 1. We observed from figure 1 increase in the intensity of High phase peaks and a decrease in low and medium peaks, in addition to the presence of displacements in the sites of these peaks. The major phases in these specimens are high transition temperature (Hg-1223 label H) and minority low transition temperature phases (L-1212), (L-1201) and amounts of the impurities. We observed by XRD analyzing increase the high phase ratio and decrease the different phases of the specimen with increasing the nickel concentration. The mass density of the unit cell was showed varying values, results are due to the temperature and time of sintering, these it gives more energy to the mass transport to get greater overlap between the molecules of materials. Thus reduce the surface area at the expense of the volume as well as long time of sintering is necessary to insert additional more layers of (CuO and CaO) in the structures of low phases [7-10].

The Lattice parameters a, b, c, c/a and volume fraction of the phases  $V_{\text{ph}}$  of  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_2\text{Cu}_{3-y}\text{Ni}_y\text{O}_{8+\delta}$  compounds as shown in table 1. It was shown in this table 1 that lattice parameters c, c/a and value of volume fraction ( $V_{\text{ph}}$ ) of Hg-1223 phase increasing with increase Ni, while the lattice parameter a, b, mass density, volume fraction of Hg-1201 and Hg-1201 phase decrease by increasing Ni concentration as comparable with the free specimen, the reason is due to the substitution of Cu by Ni, where the ionic radii of  $\text{Ni}^{+2}$  is longer than that of  $\text{Cu}^{+3}$  which increase c-parameter to be longer or get deformed

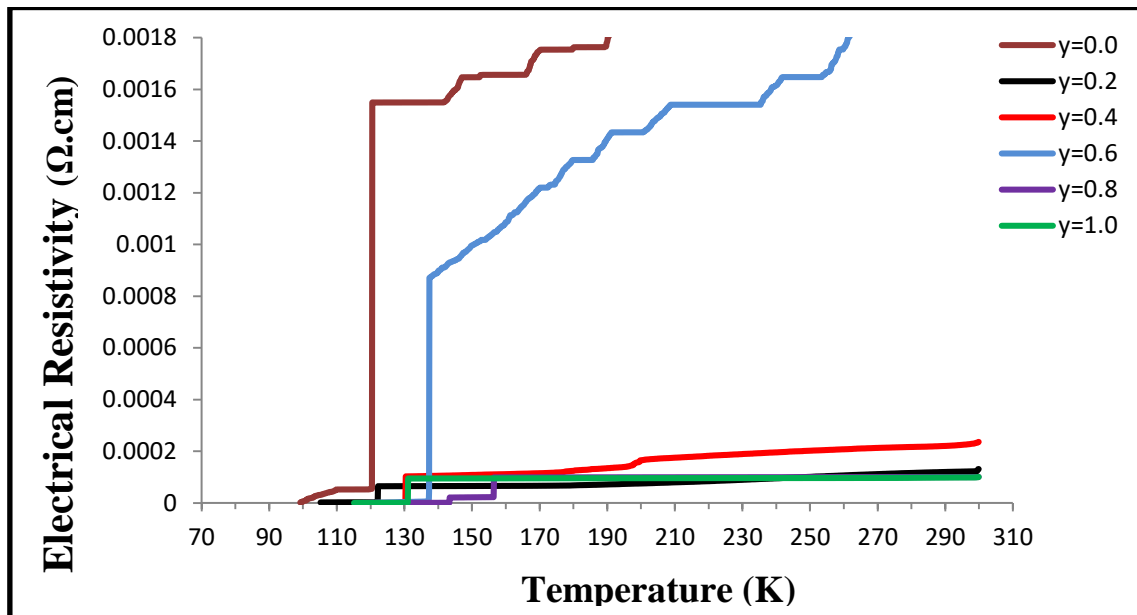
**Table 1.** Lattice parameters a, b, c, c/a, mass density and volume fraction of  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_2\text{Cu}_{3-y}\text{Ni}_y\text{O}_{8+\delta}$  (Hg-1223),  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_1\text{Cu}_{2-y}\text{Ni}_y\text{O}_{6+\delta}$  (Hg-1212),  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Cu}_{1-y}\text{Ni}_y\text{O}_{8+\delta}$  (Hg-1201) compounds and impurities

X	a=b.( $\text{A}^\circ$ )	c( $\text{A}^\circ$ )	c/a ratio	v( $\text{A}^\circ$ ) <sup>3</sup>	$M_m(\text{g}/\text{cm}^3)$	$V_{\text{ph}(1223)\%}$	$V_{\text{ph}(1212)\%}$	$V_{\text{ph}(1201)\%}$	impurities%
0	3.9019	15.7321	4.031908	239.5184	6.0824	74.102	8.826	7.953	9.117
0.2	3.8829	15.7651	4.060135	237.689	6.1224	71.2753	14.3902	5.7435	5.0503
0.4	3.8796	15.7891	4.069775	237.6464	6.1167	74.2033	10.0957	9.6064	6.0944
0.6	3.8081	15.8953	4.174076	230.5077	6.2922	76.395	13.003	5.865	4.734
0.8	3.8524	16.0635	4.169738	238.3982	6.0907	81.7370	12.7922	8.3812	5.1461
1	3.8053	15.8806	4.173285	229.956	6.3002	78.421	7.631	8.421	5.526



**Figure 1.** XRD pattern of  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_2\text{Cu}_{3-y}\text{Ni}_y\text{O}_{8+\delta}$

The resistivity as a function of temperature was testing through using the four-point probe technique with temperature range (77-300K). Figure. 2 shown the behavior of the electrical resistivity as a function of the temperature, it was found from this figure that all the specimens have metallic behavior and increasing the critical temperature with increasing nickel concentration. The optimum transition temperature founded from  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_2\text{Cu}_{2.2}\text{Ni}_{0.2}\text{O}_{8+\delta}$  compound ( $T_c=137.5\text{K}$ ), then it began to decrease when the concentration of Ni became Ni=1.0. We noted that the transition width transition  $\Delta T_c$  (different between  $T_{c(\text{onset})}$  and  $T_{c(\text{offset})}$ ) has a small value and this indicates the homogeneity of the specimen[12] as shown in table 2. We notice from table 2 that the increase of oxygen content ( $\delta$ ) lead to improves  $T_{c(\text{onset})}$  and  $T_{c(\text{offset})}$  because the changing the concentration and oxygen content effected on structure properties and superconducting properties and generation or elimination of holes concentration in the structure of the specimen[12,13].



**Figure 2.** The resistivity as function of temperature for  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_2\text{Cu}_{3-y}\text{Ni}_y\text{O}_{8+\delta}$  specimens with  $y=0.0, 0.2, 0.4, 0.6, 0.8$  and  $1.0$

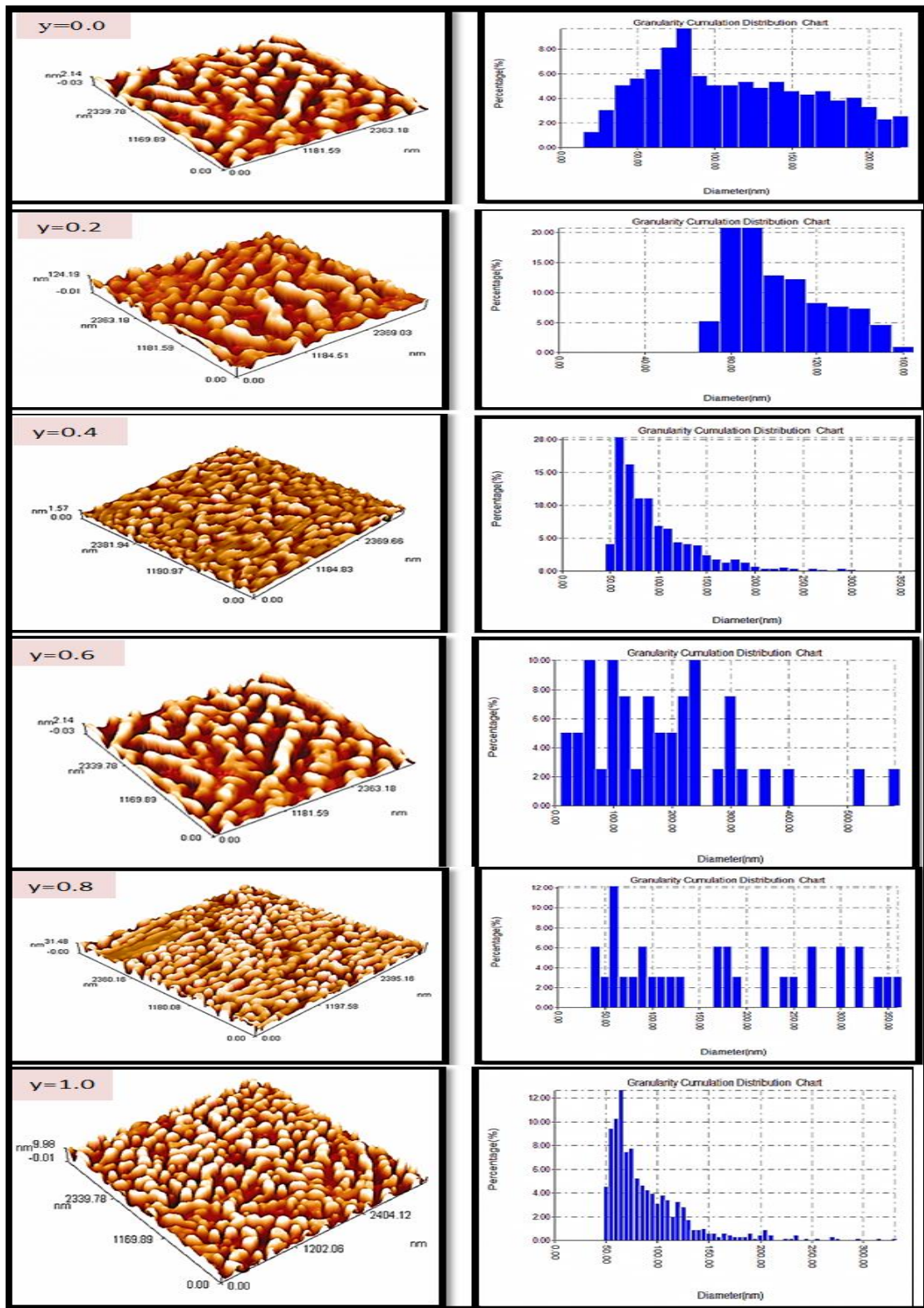
The Energy gap, Vickers micro hardness (HV), Young's modulus (E) and yield strength (Y) were calculated, the values are summarized in Table 2. It was shown that the variation of Energy gap, Vickers micro hardness Number increase with increasing Ni concentration.

**Table 2.** Transition temperature  $T_{c(\text{onset})}$ , ( $T_{c(\text{offset})}$ ), transition width ( $\Delta T_c$ ), oxygen content ( $\delta$ ) Vicker's Hardness  $H_v$ , Young modulus (E) and Yield strength (Y) for  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_2\text{Cu}_{3-y}\text{Ni}_y\text{O}_{8+\delta}$

X	$T_{c(\text{off})}$ (K)	$T_{c(\text{on})}$ (K)	$\Delta T_c$ (K)	$T_{c(\text{avg.})}$ (K)	$E_g(\text{eV})$	$\delta$	Hv	E	Y
0	105	120	15	112.5	0.0342	0.0620	130	10654.8	43.33
0.2	108	122	14	115	0.0349	0.0764	147	12048.12	49
0.4	122	130	8	126	0.0383	0.1796	155	12703.8	51.66
0.6	121	143	22	132	0.0392	0.1324	162	13277.52	54
0.8	132	143	11	137.5	0.0418	0.1752	170	13933.2	56.66
1	120	131	11	125.5	0.0381	0.1374	178	14588.88	59.33

Figure (3) shows 3D AFM image and Granularity accumulation distribution chart of  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_2\text{Cu}_{3-y}\text{Ni}_y\text{O}_{8+\delta}$  structure synthesis by solid stat reaction method distributed uniformly on the surface. It was obvious from figure(4) that the structure has small ordered particles with semispherical shape.





**Figure 3.** Reveals the (3-D) AFM images and the chart distribution of  $Hg_{0.5}Pb_{0.5}Ba_2Ca_2Cu_{3-y}O_{8+\delta}$

#### 4. Conclusions

In the current research, we synthesized  $\text{Hg}_{1-x}\text{Pb}_x\text{Ba}_2\text{Ca}_2\text{Cu}_{3-y}\text{Ni}_y\text{O}_{8+\delta}$  compounds high- $T_c$  superconducting ( $y= 0, 0.2, 0.4, 0.6, 0.8, 1.0$ ) by solid state reaction method and we tests the structural, electrical, mechanical proprieties. It was found from the XRD analysis that all the specimens are polycrystalline with a tetragonal structure and the majority phase correspond to Hg-1223 phase. The critical temperature of superconductor was calculated, it was found that all the specimens have metallic behavior and increasing the critical temperature with increasing nickel concentration. The optimum transition temperature founded from  $\text{Hg}_{0.5}\text{Pb}_{0.5}\text{Ba}_2\text{Ca}_2\text{Cu}_{2.2}\text{Ni}_{0.2}\text{O}_{8+\delta}$  compound ( $T_c=137.5\text{K}$ ). We noted that the different between  $T_{c(\text{onset})}$  and  $T_{c(\text{offset})}$  (transition width transition  $\Delta T_c$ ) has a small value and this indicates the homogeneity of the specimen. It is noted that the energy gap and mechanical properties increase when increasing Ni concentration.

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