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# Effect of Composition and Dielectric Properties for (YBCO) Superconductor Compound in Different Preparation Methods

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## Abstract

The superconductor compound (YBa2Cu2.8Zn0.2O7+ $\delta$ ) is prepared by solid state reaction (SSR), Sol-gel (SG) and laser Pulse deposition (PLD) methods. We used the X-ray diffraction technique, which shows an orthorhombic crystalline system for all the samples, and increase in the high-phase (Y-123) and decrease in low-phase and vary in proportion according to the method of preparation with the emergence of some impurities. The behavior of the samples in terms of electrical resistance and critical temperature was investigated all samples showed superconducting behavior. The properties of the dielectric (real dielectric constant, imaginary dielectric constant, loss tangent, alternating electrical conductivity) were studied as a function of frequency and range (50 Hz-1MHz) at room temperature. The results are demonstrate the apparent change in the dielectric properties by using the method of preparation of laser pulse deposition PLD at the expense of other methods, Microscopic images of the samples were examined at very high magnitudes through the scanning electron microscopy (SEM). In addition to using the titration method to determine the proportion of oxygen.

**Keywords:** YBa2Cu3O7+ $\delta$ , superconductivity, critical temperature, XRD, dielectric constant.

## 1.Introduction

The functional use of high superconductors is basically bounded by the obtainable technique to prepare superconductor with properties appropriate for use in electrical and electronic applications [1]. After the discovery of ceramic-based materials (CuO) (Which has a critical temperature close to 100 K) in a short time, a unique vault event in the record of superconducting materials in 1987, when C.W.Chu and M.K.Wu co-operated the research group at the University of Houston with another group at the University of Alabama, published a report on their success in the preparation of the ceramic compound (Y-Ba-Cu-O), which has a temperature of its transformation 90 K [2]. where temperature exceeded the



transition temperature for the first time in history the boiling point of nitrogen gas, amounting to (77 K).which became the basis for many compositions that followed it when it alerted researchers to study this new generation of compositions in the hope of obtaining compounds that turn into superconductors at room temperature. The term (YBCO) is an abbreviation for the high-temperature superconducting system (HTSC) consisting of element oxides (Yttrium oxide, barium oxide and copper oxide) and its general chemical formula (YBa2CunxZnxO2n+1+ $\delta$ ) where (n =1,2.3), it has special attention because of its high frequency capability, the crystalline structure (Y123) is characterized by the arrangement of copperoxygen levels and chains [3]. it corresponds to the general HTS classification as a modification of Cu-1212 where Ca is completely replaced by Y. This alternative offers an additional negative charge in CuO2 layers due to high valence in Y (+3) compared with Ca (+2) [4]. it also has high critical temperature, good chemical resistance against moisture and does not contain rare earth elements, Therefore, great efforts have been made in studying the methods of preparation, treatments and properties of this system [5]. The sequence is stacking for layers (YBCO) along the c-axis (BaO-CuO2-Y-CuO2-BaO) for crystallization. The layers of the perovskite's structure (YBCO) are separated by CuO2 with the atoms of the atrium between the copper-oxygen levels. Superconductivity disappears when the oxygen content is less than 6.3 The superconducting properties and transition properties of high-temperature CuO compounds are very differentiated [6]. One of the common characteristics of all superconductors basic (CuO) is not to adopt the alternate conductivity on the frequency and as well as having a low dielectric constant at high frequencies (60 GHz) at room temperature, and that the polarization of the material may indicate or include the adoption of conductivity on the frequency and the qualitative difference of conductivity between the two directions (level (a, b) and trend (c)) suggests that there are two different connecting mechanisms that can work in both directions the first one is level(a,b) which show a qualitative resistance with metallic behaviour, while the other, which is in the direction of the axis (c) is still the subject of disagreement and controversy, the study showed that the true dielectric constant of the single crystal with oxygen deficiency of the system (YBCO) is (k = 10) towards the axis (c) at room temperature and outside this range the copper oxide layers did not show high polarization towards (c) and the study showed a decrease in the value of the true dielectric constant for the single crystallization while Of the crystallization system showed a very high dielectric constant (k = 600), the high dielectric constant of the (YBCO) system is due to the granular nature of the material, which can show the double coupling between the grains, the study showed that the alternating conductivity was not adopted for multi crystallization models and full oxygen at even frequency (200MHz) while the oxygen-deficient models were (100-600), which can be explained because they are capacity at the granular boundary or due to differing densities or porosity. The alternating conductivity of the completed oxygen and oxygen deficient models was measured in the direction of the axis (c) as well as the axes (a, b) to 1GHz, has been found do not support alternating conductivity on the frequency, and that the determination of the conductivity of jumping in systems with the identification of electron cases is support alternating conductivity on the frequency, that the jump of the electron between two sites can give a contribution to the alternating conductivity supported on the frequency and that the length of the alternating jump becomes similar or smaller than the continuous jump length for this, the alternating conductivity begins to bypass the continuous conductivity [7].

## 2. Experimental Methods

The samples were prepared in three different methods (solid state reaction(SSR), solgel(SG) and laser pulse deposition(PLD)) by using appropriate weights of high purity oxides and nitrate powders from according to the general chemical formula for SSR,PLD methods:  $Y_2 O_3+2BaO+3-xCuO+xZnO→YBa$  2 Cu 2.8 Zn 0.2 O (7+δ)

The chemical formula of the preparation method sol-gel(SG) they are as follows:

Y\_2 O\_3+2Ba [( [NO]] \_3)\_2+3-xCu( [NO]] \_3)\_2.2(H\_2 O)+xZn( [NO]] \_3 )\_2.6(H\_2 O)→10( [NO] 2)+11(H 2O)+YBa 2 Cu 2.8 Zn 0.2 O (7+ $\delta$ )

In the solid state reaction, the powders are dried at 125 ° C for two hours to dispose of water vapor, then all were weighed interaction materials by a sensitive digital balance of KERN-4 for four decimal digits. The powders were mixed and grinded with a spiral electric mixer for two hours to achieve optimum homogeneity and fine powders. Then dry the resulting powders in the drying oven under 250 ° C for one hour. The mixture is then formed in the form of cylindrical discs (1.5 cm) and thickness (0.15 cm - 0.25cm) using a manual hydraulic press and under pressure (7ton / cm2) for one minute. The plates are sintering at 860 ° C at 48 hours and at a heating rate (5 ° C / min) for coherent samples. Either in the mode of laser pulse deposition (PLD), The material resulting from the solid state reaction is used, including the sedimentation method and used, short and intense laser pulses to dissipate the target material. The particles are extracted from the target and condense on the surface of the slide. Sedimentation occurs in a vacuum chamber of air to reduce particle dispersion and is used in the Nd: YaG laser excision process [8]. Finally, in the SG method we can obtain high purity materials and homogeneous structures and can be produced under lower temperatures than other methods. The basic condition in this method is to choose a solvent that does not react with the ingredients. Where we weigh the required quantities of materials and each solution with acetic acid, nitric or distilled water using a magnetic mixer for 30 minutes. Add the rest of the solute elements to the basic compound and then mix them with a magnetic mixer at 60-70 ° C. After a while we notice the gel form of the mixture, then burn the mixture in the oven at a temperature of 600 ° C and for 12 hours to get rid of the nitrates. Grind the mixture, then press on the shape of the plates and then go to the sintering process at 800 ° C for 48 hours at 5  $^{\circ}$  C / min and then cool to room temperature at the same heating rate. After obtaining the samples, they were examined by XRD for structural properties, then the cell unit density was measured, the percentage of phases formed in the sample was then calculated on the basis of the following relationship [9]. (1)

$$Vph=(\Sigma I^{o})/(\Sigma I^{o}+\Sigma I1+\Sigma I2)*100\%$$

The mass density (dm) was calculated using the following equation [10].

$$d_m = Mwt/(N_AV)$$
<sup>(2)</sup>

dm-mass density in mg/cm3, NA- number of avocadro 6022\*1023 mol-1, Mwt-molecular weight, V-cell unit size equal to (a\*b\*c) molecular weight of orthorhombic system.

the measurement of electrical resistance as a function of the temperature using the technique of the four probes, where the samples were cooled using liquid nitrogen. To determine the inertial properties, which include the electrical dielectric constant (the real dielectric

constant( $\dot{\epsilon}$ ), the imaginary dielectric constant( $\ddot{\epsilon}$ ), the loss tangent(tan $\delta$ ), the alternating electrical conductivity( $\sigma$ a.c)), It was done through the capacity calculation(C) and conductivity(G) using the impedance analysis using a device(LCR) and frequency range of (50Hz-1MHz) and according to the following relationships [11].

$$\dot{\epsilon}$$
=Cd/A $\epsilon_{\rm o}$  (3)

$$\tilde{\varepsilon} = Gd/2\pi\rho\epsilon_{o} \tag{4}$$

$$\tan \delta = \tilde{\epsilon}/\dot{\epsilon}$$
 (5)

$$\sigma_{(a.c)} = 2\pi f \epsilon_0 \tilde{\epsilon}$$
(6)

Where: d- represents the thickness of the sample, A- represents the area of the electrodes,  $\varepsilon_o$ -represents the vacuum permission (8.85\*10-12F/m),  $\rho$ - is the electrical resistivity,f- represents the frequency. Also the microscopic images of the samples were examined through the SEM microscope of the compound (YBa2Cu2.8Zn0.2O7+ $\delta$ ).

## **3.Result and Discussions**

## **3-1. Structural Properties**

The crystalline structure of the samples that were prepared by (XRD) was also studied. It was found that the compound has a multi-phases orthorhombic structure.



Figure 1. X-ray diffraction pattern preparation by solid state reaction method.





Figure 2. X-ray diffraction pattern preparation by sol-gel method.

**Figure 3-a.** X-ray diffraction pattern preparation by pulse laser deposition (PLD) method, annealing at 400 degrees Celsius.

There was also a marked decrease in peaks intensity of all the samples prepared compared to the peaks of the sample prepared in the form of Sol-gel. **Figure 2.** Shows that this sample has the optimal structural properties, which are clearly shown by the height of the high phase and the lack of the slow phase, While the rest of the samples prepared by solid state reaction and laser pulse deposition showed a significant decrease in the ratio of the high phase and an increase in the percentage of the lower phases compared to the samples prepared by Sol-gel method, as shown in **Figures 1,3**.



Figure 3-b. X-ray diffraction pattern preparation by pulse laser deposition (PLD) method, annealing at 600 degrees Celsius.

**Figure 3-a.**For the sample prepared by the pulse laser deposition (PLD) after it has been sintering at a temperature of 400°C for two hours where the pattern of overlapping peaks was not clear and difficult to obtain from the crystalline structure or calculation of the lattice parameters therefore, the re-generation was also done at 600 °C for 2 hours, as in **Figure 3-b.** 

There was a marked improvement in the crystalline structure of the membrane after the second sintering compared with the first sintering. We observe a sharp decrease in the higher phase with a significant increase in the low phase. As well as an increase in lattice parameters (a,b) with a decrease in axis length (c) which is reflected in the decrease in the ratio (c/a) as well as in the density that can be explained on the basis of the increase in cell unit size, as shown in **Table 1.** Note that the behavior of the sample prepared by the deposition method by pulse laser, had a negative effect on structural properties this is evident in the change of constants and the ratio (c/a) as well as density, The change in the lattice constants of these samples may indicate the inclusion of oxygen, which may be due to the effect of the environment surrounding the preparation conditions, which have a significant effect in the superconducting compounds [12].

method	a(A°)	b(A°)	c(A°)	c/a	$V(A^{o})^{3}$	$d_m(\frac{g}{cm^3})$	$V_{ph(high)}$	$V_{ph(Low)}$
S.S.R.	3.521	3.543	12.60	3.5808	157.28	6.9506	%83.3	%16.7
S.G.	3.493	3.523	12.62	3.6137	155.33	7.0377	%89.5	%10.5
P.L.D.	3.522	3.562	12.48	3.5440	156.59	6.9813	%68.4	631.%

**Table 1.** Values of the parameters a,b,c with different substitution.

Note that the sample prepared by the sol-gel method is optimal in terms of structural properties compared with other preparation methods. It is possible that the preparation conditions will have a significant effect on the properties of the resulting samples in terms of (granular size of the powder, mixing method, grinding, sintering temperature, length of time sintering), where the effect of the temperature of the sintering and its length is not limited to the possibility of generating microscopic cracks in the sample, but also possible to reach a state closer to the solubility, which supports the growth of low phases and impurities at the expense of the high phase and its formation therefore, the temperature and duration of sintering are critical factors in the production of high-frequency samples. It has been shown that the best temperature of sintering is the temperature near the melting point [13].





**Figure 4.** The relationship between the resistance and the critical temperature of the prepared compound by: a-Solid State reaction (SSR), b-Sol-Gel (Sg), c-Pulse laser deposition (PLD).

**Figure 4.** Illustrates the behavior of electrical resistivity as a function of the temperature, where we note that all the samples had a metallic behavior in the region before (Tc-offset), which then turns the material into the superconducting state. The reason for the sudden drop in resistivity is the possibility that other impurities and secondary phases can add other energy levels in the (CuO) layers, which correlate with the (CuO) level, causing a bond between the two levels to create the Exciton. This compound also showed a difference in the critical temperature measured for the three different methods, it was the highest critical temperature is for the prepared compound in a sol-gel where it reached (98.5) K followed by critical

temperature of the recorded compound in a solid state reaction reaching (96) K and then which was measured for the pulsed laser membrane that reached K (91), As in **Table 2.** 

method	T <sub>C(Off)</sub> (K)	<b>T</b> <sub>C(ON)</sub> ( <b>K</b> )	$\Delta \mathbf{T}_{\mathbf{C}}(\mathbf{K})$	T <sub>C(mid)</sub> (K)	E <sub>g</sub> (eV)	δ
S.S.R.	96	103	7	99,5	0.0292	0,017
S.G.	98,5	105	6,5	101,75	0.0299	0,193
P.L.D.	91	96	5	93,5	0.0277	0,054

Table 2. Relative temperature, energy gap and oxygen content in three ways.

Table 2. Shows that there is a marked increase in the critical transition temperature Tc(off) of the samples prepared by the sol-gel method. The reason for this characteristic behavior is due to the high phase formation rate, which is reflected in obtaining a higher critical transfer temperature than The critical temperature of the samples prepared by the methods of solid state reaction and pulse laser deposition. This is due to the decrease in the different methods and conditions of preparation and this condition can cause distortion in the structure and this deformity can lead to the emergence of point defects in the lattice, which can be grouped together in the form of clustered problem of defects in the stack, which can facilitate the process of impurities and growth of low phases therefore, there is a decrease in the value of the critical transition temperature [14]. We also observe an increase in the transition width ( $\Delta Tc$ ), which can be attributed to the mixed state of the high phase and low phases with high phase dominance (Y-123). In addition, the increase in the critical transfer temperature of samples prepared by the sol-gel method can be attributed to a state of strong bonding and increased contact between the granules during the sintering process resulting in a decrease in porosity. All of this discussion agrees with obtained results of oxygen content values which listed in Table 1. [15]. As a result of the increase in the critical transition temperature, Energy for this sample. The electronic survey was carried out by the (SEM) electron microscope (YBa2Cu2.8Zn0.2O7+ $\delta$ ) in different preparation methods. As we observed regularity in homogeneity of the material as we observed from the appearance of dark areas and luminous regions and at a different magnification degree 20.5 (µm). We observed that the proportion of light areas is more than the dark and was homogenous. As observed in different shapes and images by different methods of preparation of the compound homogeneity of the material and the emergence of granules large size and these sizes of granules consistent with the results of (XRD) in terms of the emergence of sharp peaks as the tops of the sharp indicates the increase in the size of granules as the decrease of the width of the peaks means increasing the size of granules According to Schiller's law). Increasing the size of granules means decreasing the area of the granular boundary, which in turn impedes the movement of superconducting currents. As shown in Figure 5,6,7. the granules of this sample are composed of granules with a plate type and a small number of granules. We also note that the distribution of granule size is fairly homogenous. We also observed the formation of black plate particles and small grains. The characteristic of black platelets in morphology is believed to be the result of the high phase as indicated by Lopera et al [16].



Figure 5. Shows the SEM of the SSR at degree of magnification (20,5)µm.



Figure 6. Shows the SEM of the SG at degree of magnification (20,5)µm.



Figure 7. Shows the SEM of the PLD at degree of magnification  $(20,5)\mu m$ .

## **3-2. Dielectric Properties**

To measure the properties of the dielectric for samples, LCR-Meter and (50Hz-1MHz) were used at room temperature. The measurements included (real and imaginary dielectric constant, loss tangent, alternating electrical conductivity).



Figure 8. Change the true dielectric constant as a function of the frequency.

Figure 8. Shows an increase in the values of the real dielectric constant of the YBCO system samples, especially in the pulsed laser deposition method and significantly while the decrease in the value of the sample prepared by the method of sol-gel and of solid state reaction, the reason is due here to the method of preparation, which may generate secondary spores and impurities in addition to the impact of the presence of pores, which increase the impact of the mechanism of polarization, The low value of the dielectric constant can be attributed to differences in the electronegative of the components of each sample [17]. which could result in an increase in concentration of charges and gaps in samples, which is observed at 50 Hz by an increase in the values of the real dielectric constant or because of the change in the nature of the material within the conductive levels and transformational in the charge carriers which works on reduction of some defects, secondary phases and impurities, this is consistent with XRD tests, as well as pores that can reduce the effect of the polarization mechanism, thus reducing the value of the real dielectric constant of the material than it is in the samples for different compounds. In frequency (1 MHz) we notice a decrease in the value of the dielectric constant due to the cancellation of the work of some polarization mechanisms.



Figure 9. Change the imaginary dielectric constant as a function of the frequency.

The imaginary portion of the electric dielectric constant in **Figure 9.** Refers to the absorption and dispersion of energy at the joints (granular boundary, localized defects, localized stacking, as well as crystalline defects) under the influence of alternating electric field, where we note the value of the imaginary dielectric constant decreases with increasing frequency due to polarization mechanisms and distribution within the sample. It is noted that the frequency constant increases the values of the imaginary dielectric constant of the sample prepared by the pulse laser deposition method compared to the samples prepared by other methods, this can be attributed to the increase in the number of dipoles that lead to increased energy lost due to the rotation and friction of the dipoles. The decrease in the amount of dielectric constant is attributed to the fact that the movement of carriers of the charge and the number of the number during the article may slow down when the convergence of any phase in the material and because of the existence of multiple and these phases, it can lead to the depletion of absorbed or dispersed energy, Therefore, there is a decrease in the value of the imaginary dielectric constant [7].



**Figure 10.** Change  $(\tan \delta)$  as a function of the frequency.

In **Figure 10.** Change the dielectric of loss tangent  $(\tan \delta)$  as a function of the frequency of prepared compound samples, where we note the loss of loss tangent by increasing the frequency within the sample and for all the samples. where we note decreased shadow lossby increasing frequency within the sample and for all samples,Losses are produced by friction and thermal irritation, which works to resist the routing of dipoles and impede its rotation with the influential field, the energy needed to keep this rotation causing loss of capacity, As the frequency increases, the surface and surface leakage current increases (The result of the presence of moisture and impurities), and the difference in the values of  $(\tan \delta)$  and other dielectric factors may be due to the heterogeneity of the distribution of the phases within the sample because of the possibility of different distribution of heat on the sample inside the furnace during the process of sintering of the sample as well as between samples [7].



**Figure 11.** Change ( $\sigma$ ac ( $\Omega$ .cm)) as a function of the frequency in different methods.

Figure 11.Shows the change in alternating conductivity with the samples frequency of the prepared compound, where it is noted that the conductivity increases with increasing frequency, according to equation (6). This increase can also be attributed to the presence of spot defects that are dominant in ionic and covalent materials As well as due to the increase of surface leakage generated on the surface of the sample due to moisture and impurities. This leads to increased alternating conductivity until it reaches the state where the maximum energy is absorbed from the field in which the amplified field frequency is equal to the normal frequency of the material in which the maximum value of the alternating electrical conductivity. The difference in the behavior of conductivity through the model of frequency probation can be explained on the basis of different methods of preparation of the sample and its multiple phases, which causes asymmetry in the behavior of the alternating conductivity relative to the directions of the cell axes where the conductivity towards the axis (c) may be different from its behavior towards Level (a,b). In general, the low values of alternating conductivity ( $\sigma a.c$ ) can be due to the effect of the temperature and the time of the sintering, were lead to a decrease in porosity and increase in density, which indicates the reduction of crystalline defects and reduce the dissipative energy, which means a decrease in the value of alternating electrical conductivity [18].

#### 4. Conclusions

Of the compound (YBa2Cu2.8Zn0.2O7+ $\delta$ ) and the prepared by (solid state reaction method(SSR), sol-gel method (SG) and pulse laser deposition method) showed (Xray) tests that all the samples have a orthorhombic crystalline structure and the higher phase ratio (Y123) was increased for samples prepared by SG compared to samples prepared by other methods, while the electrical resistivity tests showed that all samples conduct superconducting behavior with high critical temperature of samples prepared by SG compared to other methods. There was also an increase in the inertial properties of the samples prepared in PLD compared to those prepared by other methods, and they depended largely and directly on the frequency of the alternating electrostatic field. From this we conclude that the preparation of samples in the superconducting system (YBCO) depends on the method and conditions of preparation for its effect on the structural, electrical and inertial properties of the system, leading to a change in superconductivity properties.

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