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Study The Effect of Substitution Ag on behavior of The High Temperature of $YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}$ Superconductor

Widad Mahmood Faisal

Hadramout university of science &Technology- Faculty of Engineering &Petroleum Department of Electronic &Communication –Yemen

ABSTRACT

Solid state reaction used to prepared the high temperature superconductor of $YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}$ compounds, Ag was isovalent substitution Cu for (x=0.0, 0.15, 0.25, 0.35, 0.45). Iodometric titration used to determine the oxygen content in the samples. The value of O_2 was varies from sample to another .The result of titration exhibited value of δ is increasing when increasing of concentration of Ag. The samples was analyzed by x-ray diffraction(XRD) patterns has been used to determine the purity of the used materials ,calculation of the lattice constants and determination of its phase .The study shows a high purity samples with an orthorhombic single phase ,resistivity measurement by four probe technique and morphological analysis by Scanning Electron Microscopy (SEM).These parameters are the forming pressure of the pellets , the sintering temperature was 950°C for sintering time (30hours) with the flow of oxygen gas of about (1.25)L/min. The maximum Tc value was 94.2K for $YBa_2Cu_{5.5}Ag_{0.45}O_{6.5+\delta}$, it is found Tc value increasing when increased x in the $YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}$ supercomputers.

1. INTRODUCTION

A fter the discovery of $YBa_2Cu_3O_{7-\delta}$ (YBCO)superconductors, extensive studies have been made on the effect of Ag additions in YBCO system with a view to understanding the mechanism of superconductivity and to improve the current carrying capacity of these materials (1-5)it has been reported that silver addition is effective in improving mechanical properties of the brittle superconducting oxides $YBa_2Cu_3O_{7-\delta}$(Y123) (6-8)For examples studies on textured Ag doped Y123 samples showed that the Ag addition improved fractured toughness and the resistance to thermal.(9).At mesoscopic level, silver fills the integranular spaces and increases plasticity and the resistance to crack formation(10).It has been found that Ag enhance the critical current density and magnetic –flux trapping (11,12).Also silver improves the grain growth and helps to obtain a better grain orientation lowering the electrical resistance in the normal state.(13,14).In general manner. Silver addition to Y(123)clearly enhances the process dominated by the weak links (15).Silver can introduced into the samples by various ways ,such as by mixing with metallic Ag, by mixing with Ag_2O or by electro chemical method (16,17).Due to its excellent chemical compatibility. The silver ion can by encountered into the Y-123grains (12,18).Yet as Copper and silver are from the same group of the periodic table .Ag atoms can substitute Cu(19).In the Y-123 grains and consequently many physical properties are affected by presence of microscopic granularity.(20). The samples were characterized by x-ray diffraction, electrical resistivity measurement and scanning electron microscopy.

2-EXPERIMENTAL

A series of samples were prepared by a conventional solid state reaction method using high-purity powder of Y_2O_3, $BaCO_3$, CuO, and Ag_2O with 99.99% purity .The mixture to make the specimens was prepared by homogeneously mixing and grinding prescribed amounts of all powders, into a gate mortar. Appropriate amounts of these powders were mixed with alumna mortar and pestle for (3 hours) in 2- propanole and dried. The calcinations process performed at (930^{o}C) for (20 hours) in order to remove the CO_2 amount from the mixture as gas and then crushed into fine powder. The calcinations and grinding procedure were repeated three times at least, that make insure the completely evolve of CO_2 –amount from the mixture. Measuring the weight of reactants with the required amounts using sensitive balance with (4-digitals), type (STATON) 462AL. The resulting powder then pressed into pellets.. The die has a stainless steel cylinder of 0.5 cm (13mm) diameter and (1.5-1.8) mm thick using manually hydraulic press RAKIN-EIMER, under a pressure of (0.6Gp).

The final step of sample preparation was high temperature treatment that causes particles of the materials to join together and gradually reduce the volume of pore space between them the powder is compacted into a pellet shape with a certain pressure then powder particles with then be in contact with one another at numerous sites, with significant amount of pore space between the particles.

In order to reduce the boundary energy, atoms diffuse to the boundaries, permitting the particles to be bound together and eventually causing the pores to shrink. If sintering is carried out for a long time the pores may be eliminated and the material becomes dense. The programming data for this process include, the rate of heating (60^{o}C/hour) up to (950^{o}C) for (30 hours) with the flow of oxygen gas of about (1.25)L/min and then with slow rate of cooling (30^{o}C/hours) reached 550^{o}C stopped for this temperature at 7 hours before it returned to room temperature. The significance of oxygen flow is to enhance the value of **δ** to reach the content of oxygen atoms in the samples to about (6.88). X-ray diffraction (XRD) data were obtained by powder diffraction utilizing Co(kα) radiation ,Scanning electron microscope, morphology of the crystalline grains in the samples prepared .The transition temperature of HTSC samples was determined using the four probe “resistive”.

3-RESULTS AND DISCUSSION

The superconductivity of Ag substitution in Cu for $YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}$ samples for (x=0, 0.15, 0.25, 0.35, 0.45) were studied using x-ray diffraction, temperature resistivity measurements and scanning electron microscopy.

3-1-X-ray diffraction studies

X-ray powder diffraction studies were carried out on Ag_2O added YBCO samples sintered at 950°C and cooled slowly to room temperature. Temperature at the rate of 2°C/min in flowing oxygen . Figure (1) represents the powder diffraction patterns of (YBCO) specimens with 0.0 to 4.5 wt % of Ag substitution type creating a $YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}$ .All samples showed an orthorhombic structure and no shift in the peak positions ,in the x-ray patterns of YBCO was observed because of Ag substitution. However in the doped samples , a few peaks were observed at 2θ =36.875° , 45.625 ° and63.750 ° apart from those of pure ($YBa_2Cu_3O_{6.5+\delta}$).The intensities of these additional peaks were found to increase with the increase in levels of Ag_2O substitution in Cu for YBCO samples .The presence of these extra peaks in the diffraction pattern indicates the formation of a second phase in the system .This new phase could not be readily identified from the data a available in the joint committee on powder diffraction standards (JCPDS) file .X-ray diffraction pattern as shown in figure (1) taken into account with a presence of computational program to determine the lattice constants and to show the effect of substitution Ag on the structure comparable with the pure system (YBCO-compound).Table (1) showing orthorhombic phase with the lattice constants, exhibited some deviation in their value compared with pure ,this is the first data investigated and comparison with Kupper man et al(21).who studied the effect of 15%Ag as additives to compound $YBa_2Cu_3O_{7-\delta}$ .Also with Diko et al(22).They condensed their study on using 5wt%Ag with respect to compound $YBa_2Cu_3O_{7-\delta}$. Also with Pavund et al(23).Using of 10wt%Ag with respect to $YBa_2Cu_3O_{7-\delta}$ .In other words, our studies confirmed by Taylor etal (24),who suggest that Ag play an important role due to the substitution as function of Cu site of $YBa_2Cu_3O_{7-\delta}$ .The Ag atoms change the dimension of the unit cell ,because the Ag ion size is larger than that of the Cu ion .It is also reported Ag atoms saturates at nominal concentrations higher than 2.5wt% Ag (25) .

3-2Temperature Resistivity Measurements

The temperature resistivity measurements on $YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}$ samples cooled slowly to room temperature were carried out by the four –probe method (26) using digital Nanovoltmeter type (KETHLY) model 180 with sensitivity of about(±0.1 nanovolt) used to measured the voltage drop , and D-C power supply type (Electronic Veneta DV30/V3) and a KETHLY model 1221 current source after removing a substantial part of the surface layer of specimens by polishing . Silver paste was used for point contacts and the temperature was measured by a copper-constant thermocouple with an accuracy of ± 0.2 K. Figure (2) shows the temperature versus resistivity curves for $YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}$ for (x=0,0 .15, 0.25, 0.35,0.45) when the samples were slow cooled at a rate of 2°C/min in oxygen atom sphere .The transition temperature (Tc =92.7 K) for x=0 and Tc= 92.8 K for x= 0.15 , Tc= 93.2 K for x= 0.25, Tc=93.6 K for x= 0.35,and Tc= 94.2 K for x= 0.45. These results indicated clearly that zero resistivity temperature is improved by excess substitution of Ag in Cu in the composition compared with pure compound.

Table (1): INSERT CAPTION????

Samples name	Tc(K)	Lattice constant(A °)	Volume(A^3)	δ

$YBa_2Cu_3O_{6.5+\delta}$	92.7	a=3.820 b=3.8855 c=11.4835	170.4451	0.380
$YBa_2Cu_{2.85}Ag_{0.15}O_{6.5+\delta}$	92.8	a=3.820 b=3.8855 c=11.4835	170.4451	0.383
$YBa_2Cu_{2.75}Ag_{0.25}O_{6.5+\delta}$	93.2	a=3.820 b=3.8855 c=11.4834	170.4436	0.388
$YBa_2Cu_{2.65}Ag_{0.35}O_{6.5+\delta}$	93.6	a=3.820 b=3.8855 c=11.4834	170.4436	0.41
$YBa_2Cu_{2.55}Ag_{0.45}O_{6.5+\delta}$	94.2	a=3.820 b=3.8855 c=11.4835	170.4451	0.421

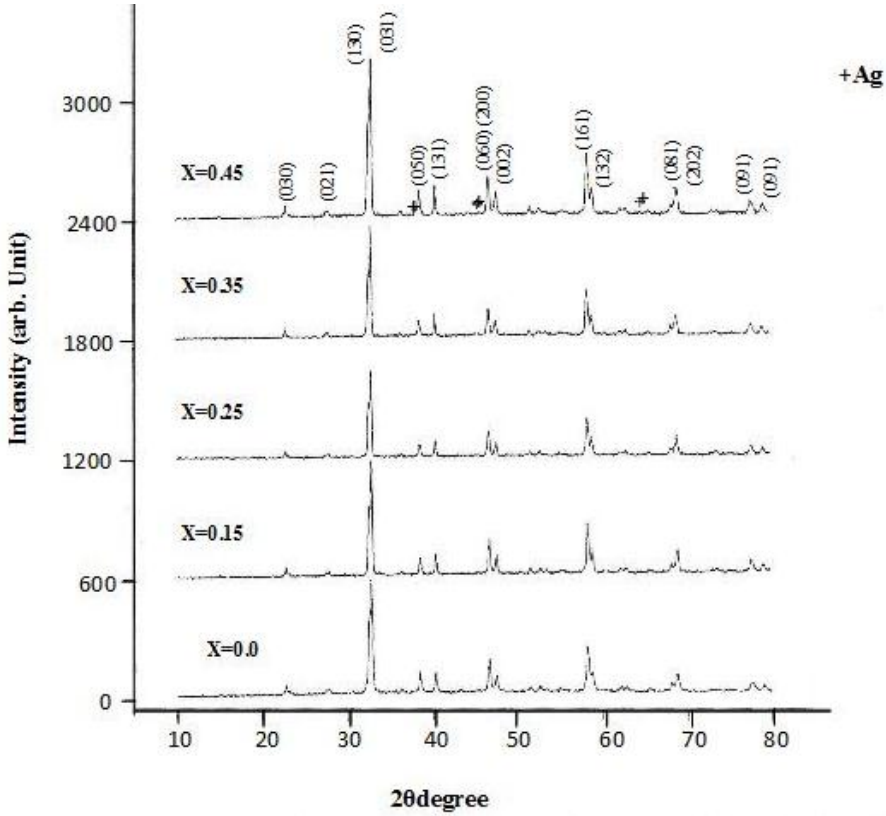

Figure (1) X-ray diffraction patterns for components $\mathbf{YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}}$

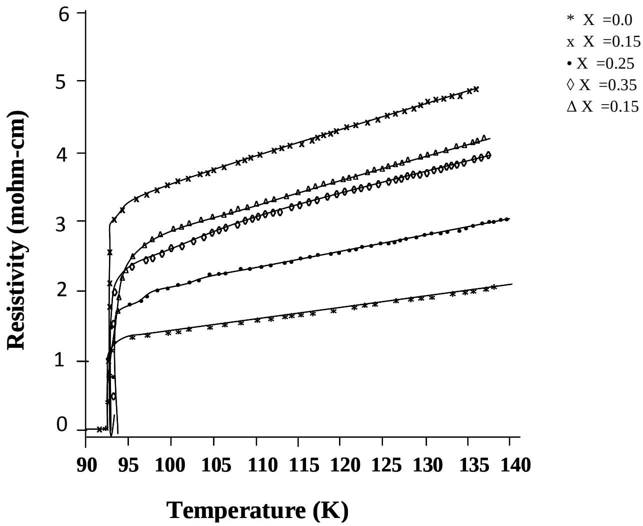

Figure (2): The resistivity veruse temperature for HTSC for $\mathbf{YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}}$

3-3- Surface Morphology

To compare the microstructure of pure and Ag substitution on cu in the $YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}$samples .SEM micrographs were taken by a JOEL JSM 4600 scanning electron microscope is used for studying the microstructure of the samples coating with metal and operated at 15KV.Figure (3) shows the microstructure of compound $YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}$ for (x=0, 0.15,0.25,0.35,0.45) cooled slowly after sintering at 950^{o}C .The micrographs clearly show that the size of the plates shapes become is between (19.23-11.4) μ m .It was found that the grain growth of plate .like increased rapidly with increasing Ag substitution and reaches to maximum size at the interior region of the fracture surface as shown in figure (3)E. The large homogeneity of the chemical species .This agree with stability of

3-4-Enhanced Oxygen absorption in $\mathbf{YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}}$ samples

The “Iodometrics titration “ method used to find oxygen content of oxide superconductor $YBa_2Cu_3O_{6.5+\delta}$,the superconducting prosperities of YBCO are intimately connected with the tetragonal – orthorhombic phase transition (27,28) which in turn depends on the oxygen stoichiometry (29, 30) . Therefore processing atmosphere and annealing temperature are of great importance in obtaining high temperature superconductivity In the case of YBCO materials it has been extensively reported that slow cooling of the sintered $YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}$ samples in flowing oxygen or annealing between (500 -550)^{o}C for several hours is an essential requirement for obtaining orthorhombic phase (31). As the tetragonal tp orthorhombic transformation takes place below 700^{o}C by absorbing oxygen, samples quenched in liquid nitrogen from temperature above 700^{o}C were always semiconducting and they did not show superconductivity (32). In our on the effect of Ag substitution in YBCO, we observed that the widely accepted procedure of slow cooling at 2 ^{o}C/min or annealing at 550 ^{o}C for several hours after high temperature sintering is not necessary to obtain superconducting in YBCO samples. Table (1) showing the values (δ) of oxygen content for all samples.

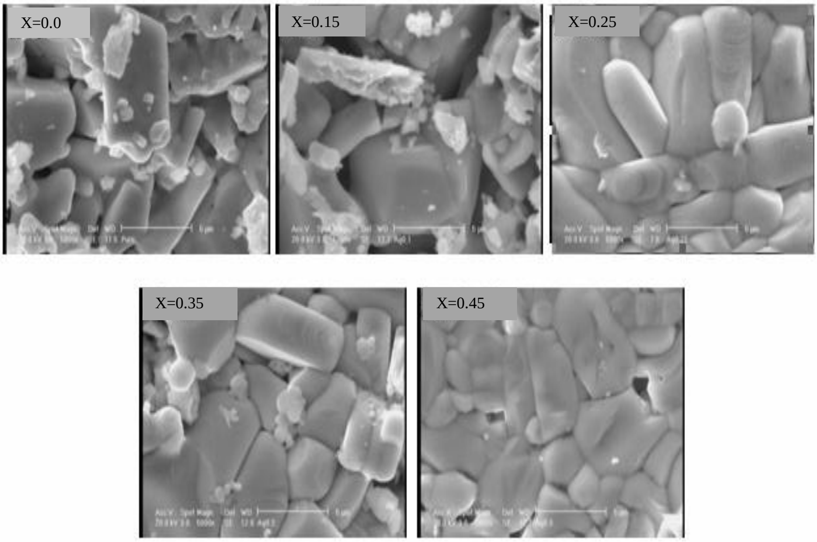

Figure (3): Morphology of fracture surface of compound $\mathbf{YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}}$

4-Discuusion

The x-ray diffraction studies of all samples ($YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}$)showed that there is no structure change in superconducting YBCO compound due to Ag substitution .A few additional peaks at 2θ =36.87 ,45.625 and 63.75 other than those of pure YBCO ,indicate the presence of a second phase in the composite .It is found that the extensively used procedure of flow cooling or annealing at 550^{o}C for several hours for oxygenation is not essential to obtained superconductivity in Ag substitution in Cu, superconducting transition was obtained by quenching Ag substitution in Cu directly from a sintering temperature 950 ^{o}C .In the present study one may suspect that tetragonal to orthorhombic phase transformation temperature has been raised to a higher temperature (950 ^{o}C),due to Ag_2O additional resulting in an orthorhombic structure on quenching the sample to room temperature . If that is true then sample quenching in liquid nitrogen should also have shown orthorhombic structure .All the electrical measurement were taken on samples after polishing and removing the outer surface layers. X-ray diffraction patterns were also taken on powdered samples which again confirms that the observed effect in quenched samples in a bulk property .Indicating that the rate of oxygen absorption has been tremendously increased by the addition of small amount of Ag_2O in YBCO and the materials become superconducting by air quenching .However it is not clear that the enhanced oxygen absorption is due to the second phase formed ,it is likely that second phase ($YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}$)may be acting as for the oxidation a catalyst for the oxidation of YBCO materials . Appearance of texture in SEM images for all samples under study such as plate –like accompanied the formation of superconducting phase.

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دراسة تأثيرالتعويض الجزئ للفضة على النحاس في المركب الفائق التوصيل ($YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}$)

وداد محمود فيصل

جامعة حضرموت للعلوم والتكنولوجيا- كلية الهندسة والبترول – قسم الهندسة الالكترونية والاتصالات - اليمن

ملخص

تم تحضير نماذج من ($YBa_2Cu_{3-x}Ag_xO_{6.5+\delta}$) الفائق التوصيل عند درجات الحرارة العالية بطريقة تفاعل الحالة الصلبة . اظهرت الدراسة تراكيب النماذج باستخدم تقنية حيود الاشعة السينية والمحضرة عند درجة حرارة تلبيد 950 درجة مئوية ، انها في طور معيني القائم (Orthorhombic). كما تم دراسة التعويض الجزئي ل(x) على التركيب والخواص الكهربائية ل $YBa_2Cu_{3-x}Ag_xO_{6.5+}$ واستخدمت عدة قيم (x=0.0,0.15,0.25,0.35,0.45).ولابعاد وحدة الخلية اعتمدت طريقة قياس المقاومةالكهربائية لايجاد قيم درجات الحرارة الحرجة (Tc) لهذه النماذج وذلك باستخدام منظومة تبريد تعمل بالنتيتروجين السائل وكانت اعلى قيمة عند (x=0.45) حيث كانت قيمة درجة الحرارة الحرجة تزداد بازدياد قيم (x). تم فحص النماذج باستخدام المجهر الالكتروني الماسح (SEM) .