Effect of Sb$_2$O$_3$ and/or CdO nanoparticle substitution on the properties of high-T$_C$ superconducting Bi$_{1.6-x}$Sb$_x$Pb$_{0.4}$Sr$_2$Ca$_{2-y}$Cd$_y$Cu$_3$O$_Z$ material

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Abstract
High temperature superconductors materials with composition Bi$_{1.6-x}$Sb$_x$Pb$_{0.4}$Sr$_2$Ca$_{2-y}$Cd$_y$Cu$_3$O$_Z$ (x = 0, 0.1, 0.2 and 0.3) and (y = 0.01 and 0.02), were prepared by using the chemical reaction in solid-state ways, and test influence of partial replacement of Bi and Ca with Sb and Cd respectively on the superconducting properties, all samples were sintered at the same temperature (850 °C) and for the same time (195 h). The structural analysis of the prepared samples was carried out using X-ray diffraction (XRD) measurements performed at room temperature, scanning electron microscope (SEM) and dc electrical resistivity was measured as a function of temperature. It was found that the sample prepared by partial substitution of Sb at ratio (x= 0.2) and Cd with ratio (y=0.01), has the maximum value of zero resistance temperature, where was the T$_C$(onset) and T$_C$(0) values for this sample are (140 K) and (123 K) respectively, and XRD results reveal that this sample has the highest percentage of Bi-2223 phase, as well as the analysis of XRD patterns of the prepared materials showed polycrystalline structure and signalize that all samples had an orthorhombic crystalline structure. It was concluded that Sb$_2$O$_3$ nanoparticles it may be acts as a flux agent in the matrix to lower the partial melting temperature of the samples and that leads to the promotion of the high-T$_C$ phase.

Key words
High-temperature superconductor, (BiPb)SCCO, Sb$_2$O$_3$ and CdO nanoparticles.

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Introduction

Since the discovery of high-$T_C$ superconductor materials, it was finding that the Bi system ((BiPb)SCCO) has great significance, and this related to their high critical current density, as well as have high transition temperature (high-$T_C$). But main problem is hard to obtain high-$T_C$ superconductor as single-phase (2223) in this Bi system, is the fact that the Bi-system with chemical formula $\text{Bi}_2\text{Sr}_2\text{Ca}_n\text{Cu}_{n+1}\text{O}_{2n+4+\delta}$, contain on three phases as follow: - The very low-$T_C$ phase when $n = 1$, low-$T_C$ phase when $n = 2$ and high-$T_C$ phase when $n = 3$ (where $n$ is the number of Cu-plains in crystalline structure). Bi-system are usually present as a mixture in the final products. So that, the producing of Bi-2223 as a single-phase, is considered the promised one between of these phases, because it is possible to produce the wires and rods for advanced current applications [1].

On the other hand, that the major worries of the Bi-system superconductor are the inter-grain low links and the presence of additional phases they were not superconductor material. Several researchers have revealed that the Bi-2201 phase is located mainly between superconducting grains, preventing the supercurrent flow [2, 3].

Since first discovery of Bi-system superconductivity, in late last century many efforts have been focused on the planning to prepared single phase of high temperature superconductor (2223). The configuration of Bi-phase has high zero resistance temperature, is safely enhanced through the partial exchange of Bi by Pb [4]. Moreover, the configuration of this phase is strongly depending on the process conditions such as sintering condition, annealing time and time of thermal processing. in this field several attempts have been made to promote the formation of the high $T_C$ phase: The accurate control of the sintering temperature and heat treatment for long periods (more than 200 hours [5-7], moreover synthesis atmosphere, sintering under low oxygen pressure, as well as the modification the components of a starting materials, such as, substitution for a part of Bi by combinations of Tl$_{0.3-x}$Sn$_x$ [8], part of Ca by Dy [9], part of Ca by holmium[10]. To date, an appropriate mechanism for the formation and strengthening of 2223 phase, for Bi-system as a single-phase has not yet been established.

It can be enhancing the properties of superconducting of chemical formula (BiPb)SrCaCuO by substitution or addition some materials have various bonding characters and various ionic radii. Therefore, several workers in this field [11, 12], they study influence of addition or substitutions of oxides of rare earth elements and other oxides in micro size particles on the properties and microstructures for (BiPb)SrCaCuO superconducting materials. On other hand, with nanotechnology developments, it was found that the nanoparticles powders have the individual chemical and physical properties, which are completely different from their counterparts in bulk size. at recent years, many researchers become more interested, in the field of the substitution or
addition nanoparticle powders to (BiPb)SrCaCuO system [13, 14].

When such nanoparticles are added to the (BiPb)SrCaCuO system, they will be settle easier and much more among the grains of these ceramic superconductors, as compared with the particles dopant have the micro size. Accordingly, when addition a suitable amount of nanoparticles powder with metallic character to the (BiPb)SrCaCuO, likely resultant that may be enhancement the connectivity between superconducting grains.

The objective of this work, two different nanoparticles was added at the same time to (BiPb)SrCaCuO system as substitution part of Bi$_2$O$_3$ micro particles by Sb$_2$O$_3$ as nanoparticles, and part of CaCO$_3$ micro particle by CdO nanoparticles, the samples were prepared with the general formula Bi$_{1.6}$-$_x$SbxPb$_{0.4}$Ca$_{2}$-$_y$Cd$_{y}$Cu$_3$O$_Z$ where (x = 0.0, 0.1, 0.2, and 0.3) and (y = 0.00, 0.01 and 0.02), as an attempt to get better the connectivity between superconducting grains, and increased a volume fraction for high –TC phase (2223) and then enhancing the superconducting properties.

**Material and methods**

**Materials used**

The materials with the chemical composition Bi$_{1.6}$-$_x$SbxPb$_{0.4}$Ca$_{2}$-$_y$Cd$_{y}$Cu$_3$O$_Z$ where (x = 0.0, 0.1, 0.2 and 0.3) and (y = 0.00, 0.01 and 0.02) were preparing by using the chemical reaction in solid-state ways. The raw materials as micro-particles Bi$_2$O$_3$, Sb$_2$O$_3$, PbO, SrCO$_3$, CaCO$_3$ and CuO supplied by Aldrich chemicals, and Sb$_2$O$_3$ and CdO nanoparticles supplied by Fluka chemicals were used. Atomic Force Microscope (AFM) is used to measuring the average particle size of the nano powders materials, which is shown that the average diameter for each of Sb$_2$O$_3$ and CdO nanoparticles (84.87 nm) and (85.53 nm) respectively. The results of particle size distribution for these Nano powders is show in the Fig. 1(a and b) respectively. The required quantities of the materials were weighed using 0.0001 g accuracy balance, Type (AB204-S) of METTLER Swiss companies, these powders were thoroughly mixed together by a wet ball mill in an agate mortar and pestle for an hour using an electrical grinder, after that, the samples were calcined in 800 °C for 24h with rate of heating 5°C/min by using temperature programmed muffle furnace. The calcined powders were grinded again to a fine powder then pressed into pellets under pressure of 0.6GPa. After that, all pellets were sintered in air of atmosphere, in 850°C for 195h, with heating and cooling rates of 5 °C/min and 2°C/min respectively. In this work six specimen were prepared according to chemical formula Bi$_{1.6}$-$_x$SbxPb$_{0.4}$Ca$_{2}$-$_y$Cd$_{y}$Cu$_3$O$_Z$, the first specimen was assorted as (a) with x =0.0 and y =0.0 and other specimens were classified as follow: - the second specimen was assorted as (b) with x = 0.1 and y = 0.00, (c) with x = 0.1 and y = 0.01, (d) with x = 0.1 and y = 0.02, (e) with x = 0.2 and y = 0.01 and (f) with x = 0.3 and y = 0.01 respectively.
**Sample characterization**

X-ray diffraction (XRD) technique was used to identify crystalline phases present in heat-treated samples. For this purpose, a Siemens D5000 (GERMANY) was used with the following features, step scan 0.02, and angular range 20–60, radiation source CuKα1 of wavelength 1.54 Å at 40 kV and 30 mA. DC electrical resistivity test, was done by using 4-points probe technique. Morphology of fractured surface was studied using Scanning Electron Microscopy (SEM) (Model VEGATS SI136XM).

**Results and discussion**

XRD patterns was used to tested the crystalline structure, for samples with different chemical composition Bi$_{1.6-x}$Sb$_x$Pb$_{0.4}$Sr$_2$Ca$_2$Cd$_y$Cu$_3$O$_{z}$ where (x = 0.0, 0, 0.1, 0.2 and 0.3) and (y = 0.00, 0.01 and 0.02), all samples were sintered at the same temperature (850 °C) and for the same time (195 h). The XRD patterns for these samples were represented in Fig. 2, the analysis showed polycrystalline structure, as well as XRD patterns point out that all specimens had an orthorhombic structure, and most of the peaks belong
to the (BiPb)-2223 phase (high-\(T_C\) phase), with a few low-intensity peaks belonging to the (BiPb)-2212 (low-\(T_C\) phase), (from figure 2 where symbols H and L indicate to high-\(T_C\) phase [(BiPb)-2223] and low-\(T_C\) phase [(BiPb)-2212], respectively). From Fig.2 it can be noticed a different in peaks intensity, whereas peaks positions do not change with increasing the substitution content of Sb and/or Cd nanoparticles in the prepared samples, and this indicating to the change in the lattice parameters of the specimens. Addition of slight amount of Sb and/or Cd it was found is quite effective in decomposing the (BiPb)-2212 (low-\(T_C\) phase), and at the same time is escort the forming the (BiPb)-2223 phase (high-\(T_C\) phase); these results are in a good agreement with other workers results [12, 13].

The peaks intensities of (BiPb)-2223 phase for grafted samples are stronger than that sample free from Sb\(_2\)O\(_3\) and CdO nanoparticles. And whenever, the substitution ratio content of Sb by BI and Cd by Ca reach to 0.2 and 0.1 respectively, the peaks intensity of (BiPb)-2223 phase increased. This indicates that the doped by Sb\(_2\)O\(_3\) and CdO nanoparticles may enhanced the nucleation and the formation of high-\(T_C\) phase. To evaluation the volume fraction of the current phases corresponding to peaks intensities of (BiPb)-2212 and (BiPb)-2223 for prepared specimens, the following relation was used [15]

\[
\begin{align*}
\text{VH} \quad \frac{\Sigma (\text{BiPb}-2223)}{\Sigma (\text{BiPb}-2223) + \Sigma (\text{BiPb}-2212)} \times 100 &= (1) \\
\text{VL} \quad \frac{\Sigma (\text{BiPb}-2212)}{\Sigma (\text{BiPb}-2223) + \Sigma (\text{BiPb}-2212)} \times 100 &= (2)
\end{align*}
\]

where: \(\Sigma I\) (2223) and \(\Sigma I\) (2212) are the sum of the (hkl) XRD peaks intensities of (BiPb)-2223 and (BiPb)-2212 phase respectively.

The lattice parameters a, b, c, c/a volume fractions of the (BiPb)-phases for all prepared specimens were tabulated in Table 1, lattice parameters were evaluated by using hkl reflections of the spotted XRD pattern through a program of software based on Cohen’s least square method and d-values [16]. This table indicates that any change in the concentration of substitutional Sb and/or Cd, led to changing in the lattice parameters (a, b, c, c/a). The deformity in the c-axis regulated in the magnitude of charge transfer from Bi-O layer to Cu-O layer, and this lead to increase volume fraction higher for 2223 phase and this was improved the zero-resistance temperature \(T_C\). as well as, it can be observed from Table 1 that the maximum volume fraction of BiPb-2223 phase was gained for the specimen which has substitutions concentration content of Sb (x= 0.2) and Cd (y= 0.01). The substitution of Sb up to x= 0.2 and Cd up to y= 0.01 from nanoparticle, it was caused increasing the value of c parameter as shown from Table 1, whereas beyond it, unsystematic variation it was noticed, by increasing the amounts of Sb to more than 0.2, and Cd to more than 0.01, it was noticed, that the volume fraction of the Bi-2223 phase was decreased. These results could be attributed to several factors such as: The substitution of Sb\(_2\)O\(_3\) and CdO as nanoparticles and with crucial ratios of substitution content, according to this, such a crucial ratio is affecting on homogeneity, transient liquid viscosity which is formation at reaction temperature, and the formed rate of the BiPb-2223 phase [14]. Affecting on the viscosity of the transient liquid which is formed at the reaction temperature, its homogeneity, and the formation rate of the BiPb-2223 phase [14]. So, the nanoparticles in this ratio...
seem to act as phase stabilizer for Bi-2223; as well as, the increase in lattice parameter (c) revealed that cations of the, Ca$^{2+}$ in addition to Bi$^{3+}$ may partly be substituted by Cd$^{2+}$ and Sb$^{3+}$ ions respectively, similar behavior was obtained by other authors for different compounds of superconductor materials [17, 18]. While the shortened in lattice parameter (c) could be, attributed to the increment the amount of oxygen in BPbSCCO system, according to what mentioned in the literature [19].

Effect of the temperature on electrical resistivity for all prepared specimens according to formulas Bi$_{1.6-x}$Sb$_x$Pb$_{0.4}$Sr$_2$Ca$_2$Cd$_y$Cu$_3$O$_{Z}$ as a function of Sb and/or Cd nanoparticles substitution contents ($x = 0.0, 0.1, 0.2$ and $0.3$) and ($y = 0.00, 0.01$ and $0.02$) it was shown in Fig. 3. All these

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**Fig. 2:** X-Ray diffraction patterns of Bi$_{1.6-x}$Sb$_x$Pb$_{0.4}$Sr$_2$Ca$_2$Cd$_y$Cu$_3$O$_Z$ samples, as a function of Cd and Sb content. Where (a): Un-doped sample, (b): for sample have ($x = 0.1$ and $y = 0.00$), (c): for samples have ($x = 0.1$ and $y = 0.01$), (d): for sample have ($x = 0.1$ and $y = 0.02$), (e): for samples have ($x = 0.2$ and $y = 0.01$) and (f): for sample have ($x = 0.3$ and $y = 0.01$), where the symbol (H) indicates to the (BiPb)-2223 (high-$T_C$ phase) and the symbol (L) indicates to the (BiPb)-2212 (low-$T_C$ phase).
samples show superconducting transition. Moreover, all samples displayed a metallic character above onset temperature, where, all specimens when are at the ordinary state display that the electrical resistivity reduced with decrease in temperature. As well as it was observed that, the resistivity curves for some specimens have a gangway and this it may be believe due to the cohabitation of (BiPb)-2223 and (BiPb)-2212 phases. The critical onset temperature \( T_C \) (onset) and the temperature at zero-resistivity \( T_C (0) \) of all samples were listed in Table 1. From this table, it can be observed that the values of \( T_C \) (onset) and \( T_C (0) \) are in the range of (140 - 119) K and (123 - 92) K respectively. The substitution of slightly ratios of Sb for part of Bi and Cd for part of Ca apparently causes an increase in \( T_C \) (onset) values, such crucial ratios of substitutional nanoparticle, it may be related to formed a partial amount of liquid phase which can be facilitates in the formation of high-TC phase, based on what was previously reported in literature [14]. As well as, that the substitution of this nanoparticles at this ratio, it may be lead to the strong linkage and growing of the connection areas between grains of superconducting material, which have higher paving along the c-axis as concluded from both XRD patterns and SEM analyses. This outcome is in a perfect concord with previously reported literature [20]. Furthermore, the specimens with highest concentration of Sn \((x = 0.3)\) or Cd \((Cd= 0.02)\) nanoparticles in this study does not show the highest \( T_c \) value, therefore, excess substitution of Sn for Bi and/or Cd for Ca resulted a deterioration effect in superconducting properties. An excess addition in Cd-doped to higher than 0.01 lower the \( (T_C (0)) \) values to (109 K) but still larger than un-doped samples. Whereas, excess addition in Sb-doped to higher than 0.2 lower the \( (T_C (0)) \) values to (92 K), which is lower than un-doped samples.

\[ \text{Bi}_ {1.6-x} \text{Sb}_x \text{Pb}_{0.4} \text{Sr}_2 \text{Ca}_{(2-y)} \text{Cd}_y \text{Cu}_3 \text{O}_7 \]

**Fig.3:** The (electrical resistivity – temperature) curves of \( \text{Bi}_{1.6-x} \text{Sb}_x \text{Pb}_{0.4} \text{Sr}_2 \text{Ca}_{(2-y)} \text{Cd}_y \text{Cu}_3 \text{O}_7 \) samples as a function of partial substitution of Sn with \( x = (0.0, 0.1, 0.2 \text{ and } 0.3) \), and partial substitution of Cd with \( y = (0.00, 0.01 \text{ and } 0.02) \).
Table 1: Volume fraction of phases % for Bi$_{1.6-x}$Sb$_x$Pb$_{0.4}$Sr$_2$Ca$_{2-y}$Cd$_y$Cu$_3$O$_Z$ superconductors with different concentrations of substitution Sb and Cd.

| X, y | a Å | b Å | c Å | V Å$^3$ | c/a | T$_c$(onset) | T$_c$(0) | Volume fraction of phases established % |
|------|-----|-----|-----|--------|-----|-------------|--------|---------------------------------------|
|      |     |     |     |        |     |             |        | (BiPb)-2223 (BiPb)-2212               |
| x = 0.0 y = 0.00 | 5.33 | 5.419 | 37.12 | 1072.15 | 6.964 | 123 | 96 | 63.472 | 36.528 |
| x = 0.1 y = 0.0 | 5.326 | 5.417 | 37.123 | 1071 | 6.97 | 123 | 111 | 72.241 | 22.759 |
| x = 0.1 y = 0.01 | 5.324 | 5.415 | 37.129 | 1070.4 | 6.974 | 130 | 115 | 81.194 | 14.472 |
| x = 0.1 y = 0.02 | 5.329 | 5.409 | 37.126 | 1070.1 | 6.967 | 127 | 109 | 69.472 | 24.574 |
| x = 0.2 y = 0.01 | 5.320 | 5.416 | 37.138 | 1070 | 6.981 | 140 | 123 | 84.647 | 11.386 |
| x = 0.3 y = 0.01 | 5.334 | 5.421 | 37.119 | 1073.3 | 6.959 | 119 | 92 | 67.539 | 32.46 |

The SEM images of the fracture surface morphology for all prepared samples Bi$_{1.6-x}$Sb$_x$Pb$_{0.4}$Sr$_2$Ca$_{2-y}$Cd$_y$Cu$_3$O$_Z$ as a function of contents for Sb-substitution and/or Cd-substitution nanoparticles (x = 0.0, 0.1, 0.2 and 0.3) and (Cd= 0.00, 0.01 and 0.02) is shown in Fig. 4 (a–f); where the photographs of fracture surface morphology for un-doped sample was shown in Fig. a, and fracture surface morphology of the other samples labeled as b with x = 0.1 and y = 0.01, c with x = 0.1 and y = 0.01, d with x = 0.1 and y = 0.02, e with x = 0.2 and y = 0.01 and f with x = 0.3 and y = 0.01 respectively.

The morphology of all samples shows small size plates and stacks of thin flake like grains. A noticeable change in distribution of grains, shape of the grains and size of grains was observed, and this indicating to the influences of Sb and Cd substitution on the morphology of the samples, based on the foregoing literature, the occurrence of grains with plate-like structure is a signature of the Bi-2223 phase formation from the Bi-2212 matrix mostly due to the change in the chemical composition of the samples and prolonged sintering process [5].

Fig. 4 (a and b) shows the surface morphology of un-doped specimen and doped specimen with Sb-substitution at 0.1 % concentration of Sb, each morphology includes many stacks composed of many layers, apparently each grain grows in random directions. So, the partial substitution of Bi elements by Sb nanoparticles, led to the linkage and growing of the connection areas between grains of superconducting material and this reducing mean free volume between the grains, and that may be the reason for the increase the values of zero resistance temperature (T$_C$). Moreover, from photographs of SEM Fig. 4(c and e), it was found that the partial substitution of Bi elements by Sb with ratios (x= 0.1 and x = 0.2) at the same time with partial substitution of Ca elements by Cd (y= 0.01) as nanoparticles, that led to increase in the grains size and it is makes the morphology more homogeneous. So, it can be observed there are a considerable effect in development a large number of grains, where grows in uniform directions and in a homogenous distribution. Over and above, it can be seen that there are both needle shapes and grain structure in the
morphology of these samples (Figs. 4 c and e) and this related to clearer of collocation of their crystals with the c axis, with the c axis, and that led to increase zero resistance temperature ($T_C$) values of these samples as compared with other samples prepared in this work. Clearly, it can be observed, `that the fracture surface morphology for specimen (Fig.4 e) with chemical composition $\text{Bi}_{1.4}\text{Sb}_{0.2}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_{1.99}\text{Cd}_{0.01}\text{Cu}_3\text{O}_Z$ is more identical and have clearer of collocation of their crystals with the c axis, moreover, the plate-like grains of superconducting material were aligned, and this made the specimen more dense and conductive as compared with other samples. This morphology in concord with other reported earlier in literature [21].

On the other hand, adversely effected is concerning to the directions and distribution of grains growth, that occurs when increase the concentration of Sb substitution and Cd substitution to larger than ($x=0.2$) and ($y=0.01$) respectively, as shown from Figs. 4 (d and f), this samples have a structure morphology, consist from plate-like grains in a randomly distribution. Furthermore, it can be observed from this figure to an existence of luminous areas with little quantities of grains, this region may have attributed to quasi melted region, it seems that the increase of Sb substitution and Cd substitution to larger than ($x=0.2$) and ($y=0.01$) respectively, leads to forming another phase like-liquid during the sintering process of prepared material, and this may be acts as an insulating material exist in the granular boundary of superconductor material. And resultant in hindrance the formation of superconducting material and then increases the electric resistance at grain boundaries and finally leads to a lower the zero-resistance temperature ($T_C$) of prepared materials. These results are agreement with previously reported by Baqiah et al. [15].

![SEM image of fractured surface morphology where: (a): un-doped sample and (b): for sample have with $x = 0.1$ and $y=0.00$. (At magnification $x10000$).](image-url)
SEM image of fractured surface morphology where: (c): for samples with $x = 0.1$ and $y = 0.01$, (d): for sample with $x = 0.1$ and $y = 0.02$. (At magnification x10000).

SEM image of fractured surface morphology where: (e): for samples with $x = 0.2$ and $y = 0.01$, (f): for sample with $x = 0.3$ and $y = 0.01$. (At magnification x10000).

Fig. 4: SEM image of fractured surface morphology for all prepared samples $\text{Bi}_{1.6-x}\text{Sb}_x\text{Pb}_0.4\text{Sr}_2\text{Ca}_2\text{Cd}_y\text{Cu}_3\text{O}_Z$ as a function of Sb and/or Cd nanoparticles substitution contents ($x = 0.0$, 0.1, 0.2 and 0.3) and ($Cd= 0.00$, 0.01 and 0.02). Where (a): undoped sample, (b): for sample have $x = 0.1$ and $y= 0.00$, (c): for samples have $x = 0.1$ and $y = 0.01$, (d): for sample have $x = 0.1$ and $y= 0.02$ (e): for samples have $x = 0.2$ and $y = 0.01$ and (f): for sample have $x = 0.3$ and $y = 0.01$. (At magnification x10000).

Conclusions

In the present research work it is concluded that:-

Joint systems with Low and high zero resistance temperature, were found to be embed in all the synthesis samples. All studies in this work showed that partial replacement of $\text{Bi}_2\text{O}_3$ by $\text{Sb}_2\text{O}_3$ nanoparticles with a concentration of no more than $x=0.2$, and partial replacement of $\text{CaCO}_3$ by $\text{CdO}$ nanoparticles with a concentration of no more than $y = 0.01$, foster the created of $(\text{BiPb})$-2223 phase to fulfill higher zero resistance temperature values. XRD analysis detect that the specimen with partial replacement of Sb with $x=0.2$ and with partial replacement of Cd=0.01 has the highest percentage of $(\text{BiPb})$- 2223 phase, and electrical resistivity results reveal that this sample have the lower value of electrical resistivity at room temperature $\rho_{(RT)} = 4.184$ mohm-cm,
and the elevated value of zero electrical resistivity temperature \( T_C (0) = 126 \) K. So, based on the foregoing, it is concluded that Sb\(_2\)O\(_3\) nanoparticles it may be acts as a flux agent in the matrix to lower the melting point of BiPbSrCaCuO system and finally to increase the zero-resistance temperature \( T_C (0) \) values.

SEM morphology confirm that the partial substitution of Bi elements by Sb nanoparticles, led to the linkage and growing of the connection areas between grains of superconducting material and this reducing mean free volume between the grains, and that may be the reason for the increase the values of zero resistance temperature. Finally; with appropriate substitution ratio of nanoparticles Sb\(_2\)O\(_3\) and/or nanoparticles CdO in the prepared material, it was correlating with the growing in figuration of (BiPb)-2223 phase and the enhance the links between grains.

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