

# An effective research for diffusion annealing temperature and activation energy in Au surface-layered Bi-2212 ceramic composites

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**Abstract** This study tries to respond two fundamental questions such as (I) what is the role of Au inclusions in the Bi-2212 polycrystalline materials prepared at the diffusion annealing temperature of 600–850 °C on the stabilization of superconductivity in small homogeneous regions and (II) what are the diffusion coefficient and corresponding activation energy of Au inclusions for the Bi-2212 superconducting system. Solution of the former question is in association with the characteristic pairing and coherence transitions extracted from the detailed dc resistivity versus temperature measurements when the latter questions are answered by the change of sample resistivity after the removal of thin-layer from the surface with the aid of the Arrhenius relation for the first time. The experimental measurement results show that the optimum diffusion annealing temperature is observed to be 800 °C where the minor phase, local structural distortions, dislocations, lattice defects and disorders in the Bi-2212 compounds degrade considerably and reach the global minimum points. Similarly, the Josephson coupled energy resides in the maximum value, and thus the superconducting grains become more and more coupled due to the increment in the percolation of intergrains. Moreover, the diffusion coefficient of Au individuals is obtained to increase regularly from  $3.6186 \times 10^{-8}$  to  $2.79278 \times 10^{-7}$   $\text{cm}^2 \text{s}^{-1}$  with the enhancement in the diffusion annealing temperature. At the same time, the temperature-dependent diffusion coefficient and related activation energy values

of Au are computed to be about  $2.601 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$  and 1.523 eV, respectively. According to the results calculated, 1.523 eV is the required minimum activation energy for the penetration of Au (heavy metal) ions into the Bi-2212 crystal structure.

## 1 Introduction

For several years, superconducting cuprate materials with their own inherent greatly higher current and magnetic field carrying capacity and extremely smaller power consumption (energy losses) due to tremendously lower dissipations have attracted a lot of interest for the application-oriented material science, potential engineering, metallurgical, technological and industrial application fields. In fact the cuprates also have recently drawn a lot of attention in the fields of large scale applications as regards the medical diagnosis, nuclear magnetic resonance and magnetic resonance imaging for the medicine; motors, transformers, generators, magnetic energy storage, power transmission cable, levitated trains, network, magnetic separation, power grids, global electric power, nuclear fusion, particle accelerators, innovative energy infrastructure, future hydrogen society and spintronics for the adapted heavy-industrial technology [1–6] as a consequence of their newly/novel electronic, magnetic and optical performances. Nowadays the cuprates begin to take the first place in engineering applications due to the considerable decrement in size and weight of motor, generator and especially supporting equipment during the production processes [7]. Besides, the cuprate materials with lower material cost, simpler chemical composition and easier availability of chemical powders become the focus of attention for the potential applications.

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The superconducting cuprates regarding the Y-, Tl-, Ga-, Hg- and Bi-based high- $T_c$  compounds exhibit the perovskite structures with the  $\text{CuO}_2$  consecutively stacked layers. The Y-based superconductor with both the Cu–O chains and crystallization in orthorhombic phase is separated from the other high- $T_c$  superconductors obtaining the tetragonal structure [8]. Among the cuprates, especially the latter compounds including three different phases of Bi-2201, Bi-2212 and Bi-2223 phases present the useful characteristics such as easy and rapid phase formation, inexpensive and innocuous chemical contents. The first scientific study on the Bi–Sr–Ca–Cu–O system, originally based on  $\text{Bi}_2\text{Sr}_2\text{Cu}_2\text{O}_{7+y}$  material with the transition temperature of up to 22 K, was performed by Michel et al. in the year of 1987 [9]. After a short time (in 1988), with the introduction of calcium oxide and increment of the conducting  $\text{CuO}_2$  layers in the system the critical temperature have reached to around 110 K [10, 11]. The former material is named as Bi-2201, the latter ones are called as Bi-2212 and Bi-2223 according to their own number of the  $\text{CuO}_2$  planes. Further, the Bi-2201 system has no Ca plane in the crystal matrix; conversely, the Ca atoms dwell in a slab within the interior of  $\text{CuO}_2$  consecutively stacked layers in the Bi-2212 and Bi-2223 system. Accordingly, the main reason of enhancement in the critical transition temperature stems from the increment of  $\text{CuO}_2$  planes in the crystal texturing. Among three phases, the Bi-2212 one with the invariant of oxygen stoichiometry and especially lesser weak link problems and rather better thermodynamic stability exhibits the superior properties as compared to the other phases [12–14]. However, the narrowest operating temperature range and sensitivity over the magnetic field and current as well as the brittleness nature limit the superconducting cuprates in more application fields, the Bi-2212 compound as it does. Similarly, the other physical problems (structural layer, random distribution of grain coupling and even lower charge carrier density, larger penetration depth and shorter coherence length) are the most annoying problems belonging to the Bi-2212 superconducting compound for newly/novel applications [15, 16]. Hence, the researchers have endeavored to overcome the limitations by using some scientific techniques as regards the chemical substitution/doping/addition into the crystalline structure, transition metal evaporation onto the specimen surface and alteration of preparation conditions (composition, dopant type/quantity, operational procedure, calcination/annealing ambient atmosphere, heat-treatment including pressure, time and temperature) [17–20]. Of the scientific techniques, the transition metal evaporation method with easy reproducible process takes first place as the best technique to degrade the limitations [21]. At the same time, the evaporation method enables the researchers to determine the diffusion rate or diffusion coefficient (associated with how fast the foreign

atoms penetrate in the crystal) belonging to the metal ions throughout the crystal lattice. By use of the diffusion coefficient finding, the researchers decide that why the characteristic properties of superconducting cuprates changes with the doping mechanism [22]. In the present work, we conduct the gold inclusions inserted into the Bi-2212 crystal lattice by the evaporation method to improve the unfavorable characteristics provided above at different diffusion annealing temperature range from 600–850 °C. The changes in the structural, electrical, superconducting, flux pinning and mechanical properties have already been examined by means of the dc electrical resistivity, microhardness, powder X-ray diffraction, scanning electron microscopy, electron dispersive X-ray, bulk density and transport critical current density experimental measurements in detail elsewhere [7, 23, 24]. According to the experimental results observed, all the properties discussed were recorded to significantly improve increasing the diffusion annealing temperature up to 800 °C due to the introduction of Au impurities into the Bi-2212 crystal structure (confirmed by the dc electrical resistivity, XRD and EDX examinations). This was attributed to the fact that the optimum concentration of Au foreign impurities into the superconducting grains or over grain boundaries in the Bi-2223 crystal system. Accordingly, the over-doped nature of Bi-2223 structure transits to optimally doped state as a result of increment in both the density of electronic states at the Fermi energy level and the overlapping of the Cu-3d and O-2p wave functions. Similarly, the improvement of bulk density, mechanical durability and ideal flexural strength with the annealing temperature stemmed from the reduction of disorders, lattice strains, defects, dislocations, misorientations and local structural distortions in the Cu–O<sub>2</sub> planes. Besides, the variations of critical current density, phase volume fractions, average crystallite size, Lotgering index, lattice cell parameters *a* and *c* parameters are investigated with regard to the diffusion annealing temperature. That the Au<sup>+</sup> inclusions with higher electronegativity may preferentially be substituted by the Sr<sup>2+</sup> in the Bi-2212 crystal matrix was another important results deduced from the EDX results. Here, we focus not only on the changes in characteristic two-stage (pairing and coherence) transitions for the Au surface-layered Bi-2223 superconducting materials with the diffusion annealing temperature by use of the detailed dc resistivity ( $\rho$ -T), but also on the determination of the diffusion coefficient and related activation energy of Au nanoparticles throughout the Bi-2223 bulk superconducting system. To the best of our knowledge, until now the detailed survey has been published on neither the diffusion coefficient nor corresponding activation energy for the Au nanoparticles in Bi-2223 inorganic solid compound with respect to the diffusion annealing temperature, being one of the most striking point deduced from this work.

## 2 Experimental details

In the previous studies, the role of Au impurity diffusion on the structural, electrical, superconducting, flux pinning and mechanical properties of polycrystalline Bi-2223 bulk superconducting materials produced by the conventional solid-state reaction route was previously reported with the aid of the dc electrical resistivity, powder X-ray diffraction, scanning electron microscopy, electron dispersive X-ray, bulk density, Vickers hardness and transport critical current density experimental measurements in [7, 23, 24] where one can also observe the experimental details related to the sample preparation procedures. In the current study, the same experimental procedure is followed as given in the papers mentioned above. In addition to this, the evaporation of Au inclusions onto one face of the sample obtained is exerted at the high vacuum of about  $10^{-5}$  Torr with the gold wire (high-purity, 99.999%) by means of AUTO 306 vacuum coater (EDWARDS) machine. Besides, the color of the sample surfaces exposed to diffusion process is observed as golden-blond owing to Au foreign inclusions. What's more, the control process is done by means of the visual inspection and mechanical damage method in order to check whether the Au nanoparticles have just adhered on the specimen surface or not. Additionally, the samples produced are exposed to the main heating process at different temperatures such as 600, 650, 700, 750, 800 and 850 °C. While the virgin sample is symbolised as pure, the other samples are denoted as Au-600, Au-650, Au-700, Au-750, Au-800 and Au-850 with respect to the annealing temperatures. Further, in this exhaustive study, the changes in characteristic two-stage (pairing and coherence) transitions for the Au surface-layered Bi-2212 superconducting materials with the diffusion annealing temperature is carried out by use of the detailed dc resistivity measurements performed in a He closed cycle system under 5 mA dc current through the sample surfaces in the temperature range of 55–105 K. Moreover, the determination of gold diffusion rate (coefficient) and related activation energy values are deduced from the differentiation of specimen resistivity after the successive removal of thin layers of about 20–130  $\mu\text{m}$  from the sample surface for the first time. The detailed computation method for the determination of Au diffusion fast rate and activation energy is provided in the following part of diffusion coefficient and corresponding activation energy of Au inclusions for Bi-2212 superconducting system. Likewise, the meanings of characteristic pairing and coherence transitions are thoroughly explained in detail in the following part of temperature derivative belonging to sample resistivities.

## 3 Results and discussion

### 3.1 Diffusion coefficient and corresponding activation energy of Au inclusions for Bi-2212 superconducting system

Here, we aim to answer three crucial questions: (I) what is the diffusivity of Au impurities for the Bi-2212 inorganic materials; (II) what is the relation between the Au diffusion coefficient and diffusion annealing temperature; (III) what is the related activation energy of Au nanoparticles? Prior to the answers of questions, it is to be described here that the diffusion coefficient (known as diffusivity) is attributed to the diffusion of an impurity along with the pore space of porous media [25, 26]. In this respect, the greater diffusion coefficient the impurity material exhibits, the faster the impurity diffuses into the target material. Thus, the researchers can easily determine the diffusion mechanism belonging to the impurity in the crystal lattices of the target material. For this aim, the most common four methods for the superconducting ceramics are arranged as the follows:

- (1) Variation of resistivity or conductivity after successive removal of thin layers on the sample surface
- (2) Change in the lattice constant parameters throughout the thin layer removal
- (3) Radio tracer, and
- (4) EDXRF technique.

The radio tracer and EDXRF methods include expensive procedures despite the similar results. The first two techniques (especially the first one) are reliable and useful enough to describe the diffusivity of impurity. In the current work, the diffusion coefficient belonging to the Au foreign impurities penetrated into the Bi-2212 system is found at different temperatures in the range of 600–850 °C. Experimentally, the resistivity distributions with respect to the specimen thickness after the successive removal of thin plate-parallel layers are systematically measured at the room temperature. The impurity diffusion (from a constant source into a semi-infinite solid) is defined by the following relations [27, 28]:

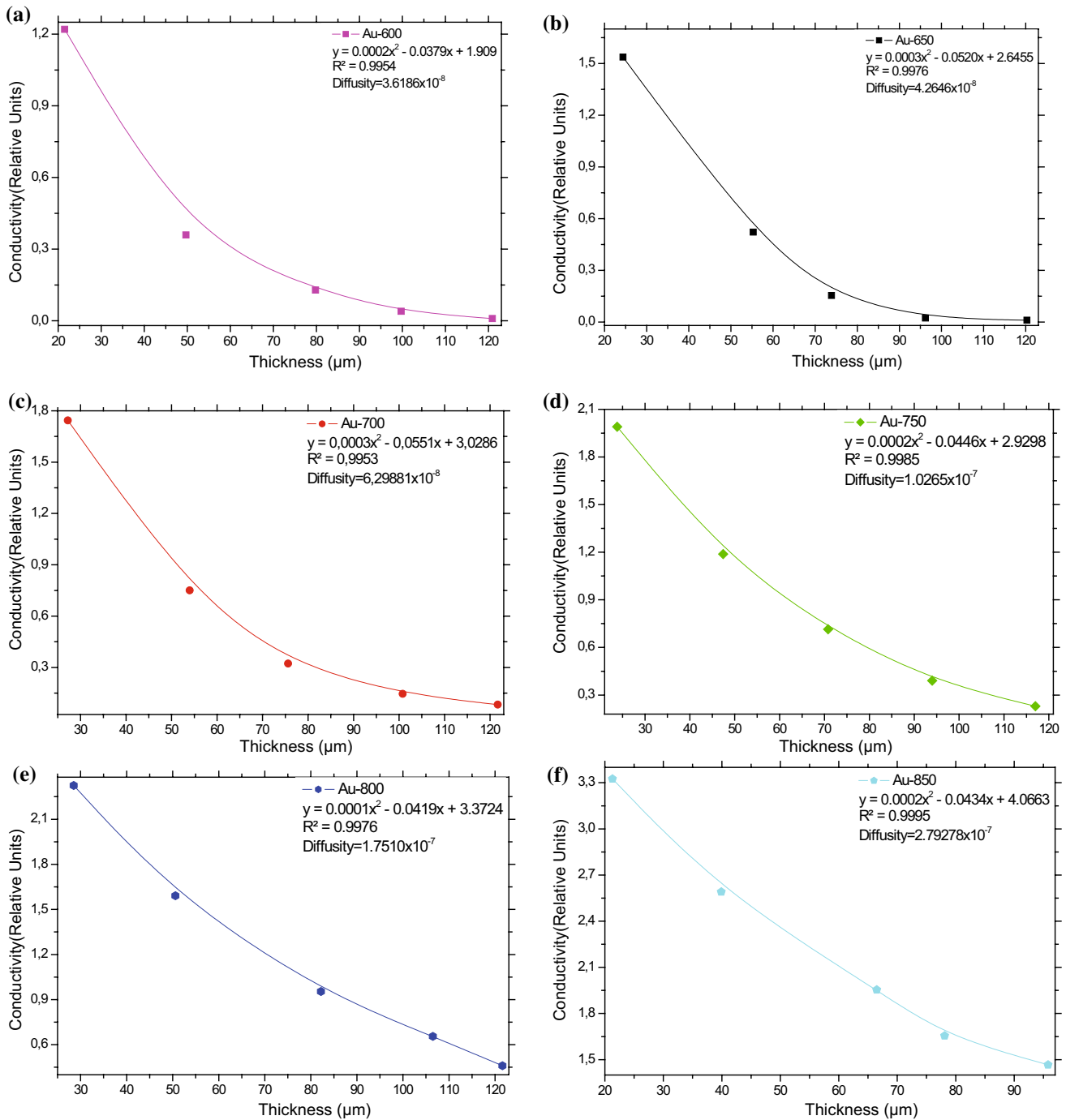
$$N(x_0, t) = N_0 \left[ 1 - \operatorname{erf} \left( \frac{x}{2\sqrt{Dt}} \right) \right] \quad (1)$$

here  $\operatorname{erf} \left[ \frac{x}{2\sqrt{Dt}} \right]$  denotes the error function with the argument of  $y = \left[ \frac{x}{2\sqrt{Dt}} \right]$

$$\operatorname{erf}(y) = \left( \frac{2}{\sqrt{\pi}} \right) \int_0^y \operatorname{erf}(-y^2) \quad (2)$$

where  $N_o = (0, t)$  is related to the constant concentration on the specimen surface when  $N(x, t)$  is in association with the impurity concentration at the distance  $x$  from the surface. Moreover, the abbreviated  $D$  displays the diffusion coefficient when  $t$  illustrates the diffusion-annealing time. According to the equations given, the change of resistivity or conductivity according to the sample thickness exhibits

the similar characteristics with the concentration distribution of the diffused impurity. One can see the variations of conductivity against the removal thickness from the sample surface for every material prepared in Fig. 1. It is to be stressed that the solid curve displays the calculated concentration profile pertaining to the Au foreign impurities embedded in the Bi-2212 superconducting system.



**Fig. 1** Variations of conductivity over removal thickness from sample surfaces belonging to **a** Au-600, **b** Au-650, **c** Au-700, **d** Au-750, **e** Au-800 and **f** Au-850 superconducting materials

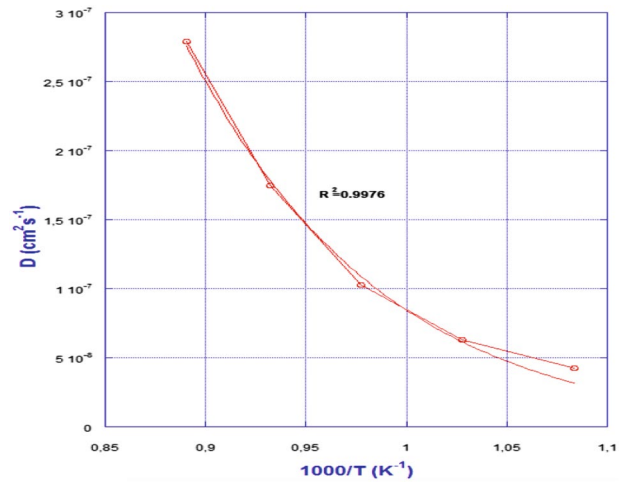
**Table 1** Calculated Au diffusion coefficients for every solid cuprate prepared

Diffusion-annealing temperature (°C)	Diffusion (cm <sup>2</sup> s <sup>-1</sup> )
600	$3.6186 \times 10^{-8}$
650	$4.2646 \times 10^{-8}$
700	$6.2988 \times 10^{-8}$
750	$1.0265 \times 10^{-7}$
800	$1.7510 \times 10^{-7}$
850	$2.7928 \times 10^{-7}$

According to the figures, the experimental evidences fit well with the theoretical curve. Hence, it is fair to conclude that the conditions of impurity diffusion are perfectly satisfied by Eq. (1). The diffusion coefficients computed are found to be about  $3.6186 \times 10^{-8}$ ,  $4.2646 \times 10^{-8}$ ,  $6.29881 \times 10^{-8}$ ,  $1.0265 \times 10^{-7}$ ,  $1.7510 \times 10^{-7}$  and  $2.79278 \times 10^{-7}$  cm<sup>2</sup> s<sup>-1</sup> for the Au-600, Au-650, Au-700, Au-750, Au-800 and Au-850 superconducting samples, respectively (Table 1). It is appear from the results that the diffusion coefficient increase regularly with the enhancement of the diffusion annealing temperature. This is associated with the fact that more and more Au individuals penetrate both into the superconducting grains and over the grain boundaries in the Bi-2212 crystal matrix. Based on the previous studies [7, 23, 24] and the following section, the optimum diffusion annealing temperature of 800 °C makes the incorporation of optimum Au inclusions into the crystal core suppress considerably the local structural distortions, dislocations, lattice defects and disorders in the Bi-2212 polycrystalline compounds. Consequently, the Au optimum nanoparticles penetration into the Bi-2212 system improves considerably the electrical, superconducting, physical and mechanical properties. Even, the other parts of the paper as well as the previous studies favor unequivocally the improvements.

In case of Au-850, onwards the Au nanoparticles begin to accumulate predominantly over the grain boundaries. Under the circumstances, the diffusions at higher temperatures are much more significant as compared to those at lower ones, confirming that the Au diffusion mechanism is seriously dependent upon the diffusion annealing temperature. Accordingly, we can pass the last question being about the activation energy value of Au impurities. For this aim, the mean values of diffusion coefficients over the diffusion annealing temperature for the Au nanoparticles inserted in the Bi-2212 superconducting matrix are graphically given in Fig. 2. What stands out clearly from the figure is that the Au impurity diffusion coefficient in the temperature range of 600–850 °C enhances with regard to the Arrhenius relation,

$$D = 2.601 \times 10^{-6} \exp \left[ \left( (-1.523 \pm 0.632)eV/k_B T \right) \right] \quad (3)$$



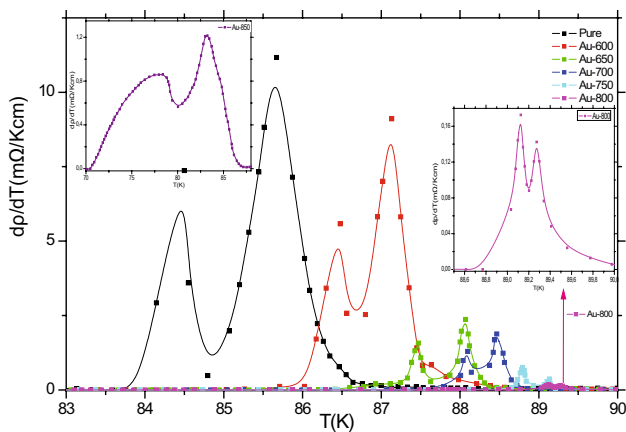
**Fig. 2** Temperature-dependent Au diffusion coefficients in the near surface of Bi-2212 polycrystalline superconducting ceramics

Equation 3 quarantines that the temperature-dependent diffusion coefficient and activation energy value pertaining to the Au impurities in the Bi-2212 system are about  $2.601 \times 10^{-6}$  cm<sup>2</sup> s<sup>-1</sup> and 1.523 eV, respectively. Based on the combination of relatively low value of diffusion rate and high value of activation energy (due to the penetration of heavy metal ions of gold into the crystal structure), it suffices to confirm that the minimum activation energy of 1.523 eV is required for the introduction of gold ions to the Bi-2212 crystal structure.

### 3.2 Temperature derivative belonging to sample resistivities (dp/dT)

In our research for the current paper, we determine the changes (improvement or degradation) of granular nature pertaining to the virgin and Au diffused Bi-2212 bulk superconducting compounds with the annealing temperature in a range of 600–850 °C to answer the question is that whether the extra local microstructural distortions induce or not with the annealing temperature. Derivative of dc resistivity (dp/dT) versus related temperature is one of the best identification methods to explore the local microstructural distortions for the granular materials with their intrinsic structural inhomogeneity and grain boundary coupling problems.

Moreover, from the dp/dT curves the peak points on characteristic two-stage transitions are defined as the pairing transition ( $T_p$ ) or bulk genuine ( $T_c^{mid}$ ) observed at the higher temperature value when the coherence transition ( $T_{co}$ ) appeared at the lower temperature value. In more detail, the former temperature detected in the small homogeneous regions is connected with the stabilization of superconductivity whereas the latter parameter appearing in the long-range coherent state of the bulk superconducting sample



**Fig. 3** Temperature derivatives of resistivities against temperature graphics for the Au surface-layered superconducting materials. *Right and left insets* curves of Au-800 and Au-850 samples, respectively

corresponds to the location of resistance. Accordingly, the weak-interactions between the superconducting grains begin to move actively at the coherence transition temperature [28]. Here, the effect of diffusion annealing temperature on the induced disorders, distortions, porosity, defects, lattice strains, grain boundary weak-links and misorientations of the superconducting grains as well as the local structural distortions is analyzed by means of the  $dp/dT$  curves of all the superconducting materials as depicted in Fig. 3 clearly. We depict the figure in three parts due to the variation of peak amplitudes and temperature values: (I) the right hand side ascribes to the pairing ( $T_p$ ) and coherence ( $T_{co}$ ) transitions for the Au-800 sample, (II) the left hand side is in association with two-stage transitions for the Au-850 solid material, and (III) the middle graphic displays the transition temperature behavior of other superconducting materials. According to the figure, each material prepared exhibits the characteristic two-stage nature along with the transitions. Not only the  $T_p$  and  $T_{co}$  peak temperature values but also the broadening of  $dp/dT$  curves are noted to retrograde noticeably while the temperature values of peak points are found to shift towards such a higher temperature value with the annealing temperature value up to a critical value of 800 °C (right inset of Fig. 3) above which the changing trend of peak, broadening and peak-temperature values presents an opposite behavior, and the broadening gets larger and larger for the Au-850 sample (left inset of Fig. 3). This is association with the fact that the diffusion annealing temperature value of 850 °C causing the incorporation of excess Au nanoparticles in the Bi-2212 superconducting crystal matrix promotes/damages the Bi-2201/Bi-2212 phase and quality of interactions between the grains seriously, having already been confirmed by [7, 23, 24]. Further, the amplitude of  $T_{co}$  peak with more broadening in the  $dp/dT$  curve is found to be even larger than that of  $T_p$  (left inset of Fig. 3).

This means that the excess diffusion temperature shrinks the homogeneous superconducting regions in the Au-850 ceramic cuprate, and hence the formation of Bi-2212 phase reaches the unstable point as a consequence of the degraded overlapping (hybridization mechanism) of Cu-3d and O-2p wave functions. Regarding the values of  $T_{co}$  and  $T_p$  parameters for the Au-850 material, the former value is recorded to be 77.94 K while the latter one is found to decrease towards to the value of 83.22 (Table 2). The radical decrement of two-stage transition temperatures results from the rapid increment in the minor phase, permanent disorders, lattice strains, defects, local structural distortions and grain boundary coupling problems in the Cu–O<sub>2</sub> consecutively stacked planes [29, 30].

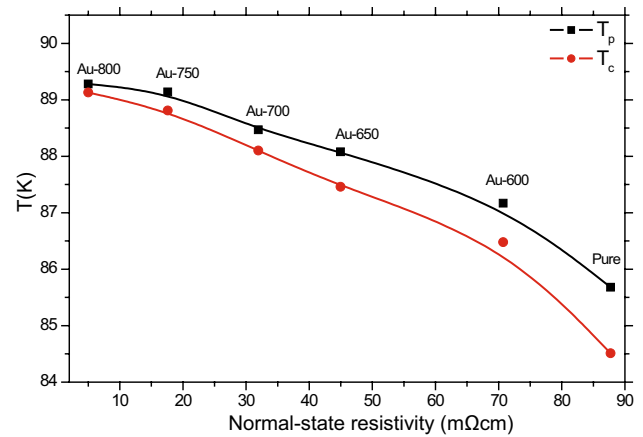
For the other materials, both the amplitudes of  $T_p$  and  $T_{co}$  peaks tend to degrade with the annealing temperature due to the increment in both the stabilization of superconductivity in relatively large homogeneous regions (formation of more cooper-pairs), and especially the decrement in the location of the resistance in long-range coherent state. As for the temperature values, each value increases significantly with enhancing the diffusion annealing temperature. In numerical, the  $T_p$  values are found to change from 85.68 K until 89.28 K whereas the  $T_{co}$  values are observed to shift from 84.51 to 89.13 K for the pure and Au-800 compound, respectively (Table 2). The increment in both the values points out that the reasonable Au nanoparticle diffusion contributes to the improvement in the stabilization of the superconductivity and the conductive locations in long-range coherent state. It is another probable result inferred from the current work that the amplitudes (in decrement trend) of  $T_p$  peaks belonging to the pure, Au-600, Au-650 and Au-700 materials are observed to be larger as compared to those of the  $T_{co}$  parameters, confirming that the Bi-2212 phase strengthens with the annealing temperature (middle of Fig. 3). On the other hand, for the Au-750 and especially Au-800 cuprate the  $T_p$  peak amplitudes are detected to be smaller than those of  $T_{co}$  values. Accordingly, the Au-800 sample exhibits the most stable Bi-2212 phase as a result of either the enlargement of homogeneous superconducting regions (formation of more cooper-pairs) or more stabilization of superconductivity in the small homogeneous regions.

**Table 2** Pairing transition ( $T_p$ ) and coherence transition ( $T_{co}$ ) temperature values for all the superconducting materials

Samples	$T_p$ (K)	$T_{co}$ (K)
Pure	85.68	84.51
Au-600	87.17	86.48
Au-650	88.08	87.46
Au-700	88.47	88.10
Au-750	89.14	88.81
Au-800	89.28	89.13
Au-850	83.22	77.94

As for the theoretical approaches, the  $T_p$  transition being responsible for the thermal fluctuations is received to reflect the transport and equilibrium behaviors in the normal state of a superconductor [31, 32]. Conversely, there is no detailed scientific information about the appearance of  $T_{co}$  transition observed at a lower temperature value. Thus, several models are ploughed to define and explain the coherence transition temperature findings. However, Percolation model, working in the isolated grains and grain boundary coupling between the superconducting grains, is known as a useful approach for the description of  $T_{co}$  transition [33, 34]. According to the Percolation model, once the Josephson coupled energy (abbreviated as  $E_j$ ) exceeds the thermal energy, the percolation of inter-grains occurs immediately and thus the superconducting grains become coupled. It is, of course, that the couplings are randomly produced in a certain probability depending on the temperature values. However, it should strongly be noted here that as soon as the coupling probability equals to the percolation threshold at a given temperature value (about the coherence transition region), the coupled superconducting grains begin to form the enormous cluster. Shortly, the discussion of  $T_{co}$  transition in the percolation model relies on the fact that more and more grains become coupled with the reduction of temperature due to the increase in the percentage of superconducting clusters in the path. Under the circumstances, zero resistivity is inevitable for a superconducting material at a temperature lower than the critical transition temperature value. In the present study, the shift of  $T_{co}$  transition to a higher temperature value with the annealing temperature (up to 800 °C) declares the significant increment in the percentage of superconducting clusters (made from the coupled superconducting grains) in the path.

We also endeavor to advance in-depth understanding of relationship between the metallic interaction and superconducting clusters in the path by means of Percolation model. Consequently, we gather two characteristic temperatures and normal-state resistivity findings (gathered from [7]) at room temperature for the cuprate materials except for the Au-850 compound due to its unstable characteristics and depicted graphically in Fig. 4. It is visible from the figure that the  $T_p$  and  $T_{co}$  transitions tend to diminish considerably as the normal-state resistivity increases or vice versa. This shows that once the temperature reaches to the  $T_{co}$  value, the first enormous cluster begins to form abruptly, and the resistivity measured in the path stems only from the grain boundary resistance [35]. Thus, one can be convinced quickly from the percolation model that the  $T_{co}$  transition value (beginning of the grain boundary resistance) may directly be in association with the phase transition from normal state to superconducting state. It is to be mentioned here that the increment of non-superconducting barrier regions through the grain boundaries makes new or intrinsic coupling



**Fig. 4** Differentiation of  $T_p$  and  $T_{co}$  transitions according to normal state resistivity

problems resurrect extensively. The material accordingly needs rather larger Josephson coupled energy to form the superconducting cluster or exhibit the superconductivity. In this respect, Percolation approach verifies the close relation between Josephson coupled energy and the phase transition from normal state to superconducting state.

## 4 Conclusion

In the current study, we not only examine the change in the stabilization of superconductivity belonging to the Au surface-layered Bi-2212 compound with the diffusion annealing temperature range of 600–850 °C by means of characteristic two-stage (pairing and coherence) transitions derived from the detailed dc resistivity measurements, but also find the diffusion coefficient and related activation energy of Au nanoparticles through the bulk Bi-2212 superconducting system using the changes of specimen resistivity over the thin-layer removal from the surface for the first time. We also shed some lights on the crucial changes in the stabilization of superconductivity in the small homogeneous regions, overlapping of Cu-3d and O-2p wave functions, amplitude of pair wave function and formation of cooper-pairs with the enhancement of diffusion annealing temperature by means of the theoretical approach of Percolation model. Likewise, the model enables us to explain the mechanisms between the metallic interaction and superconducting clusters in the path. To sum up, this comprehensive work shows the following major results:

- Both the  $T_p$  and  $T_{co}$  are directly related with the formation of cooper pair.
- With the increment up to the critical diffusion annealing temperature value of 800 °C, either the supercon-

ductivity in small homogeneous regions in the Bi-2212 inorganics compound sample more and more stabilizes; or the homogeneous regions related to the formation of more cooper-pairs tend to enlarge considerably. Thus, the superconducting behavior of Bi-2212 phase improves.

- Moreover, the optimum annealing temperature value of 800 °C leads to augment harshly the Josephson coupled energy so that the superconducting grains become coupled due to the appearance of the percolation of intergrains. In other words, not only the percentage of superconducting clusters in the path about the  $T_{co}$  transition region, but the coupling probability also increases systematically with the diffusion annealing temperature.
- In case of 850 °C annealing temperature, the structural problems regarding the permanent disorders, defects, dislocations, local structural distortions and grain boundary coupling problems as well as the minor phases (encountered in Ref. [7]) appear immediately and thus the weak-connections between the superconducting grains begin to play more active role in the crystalline structure so that the resistivity region in the path expands along with the Au-850 material.
- Similarly, both the significant decrement of the  $T_c^{mid}$  transition value and increment in the amplitude point of  $T_c^{mid}$  peak may be attributed to the degradation in the overlapping (hybridization mechanism) of Cu-3d and O-2p wave functions (known as the carrier localization or hole filling). In other words, the excess annealing temperature damages the amplitude of pair wave function ( $\Psi_0$ : parameter connected with the number of superelectrons;  $\Psi = \Psi_0 e^{-i\varphi}$ ) [36].
- Percolation model is useful and reliable approach for the definition of the relationship between Josephson coupled energy and the transition from normal state to superconducting state.
- The diffusion coefficient parameter of Au inclusions tends to augment monotonously from  $3.6186 \times 10^{-8}$  until  $2.79278 \times 10^{-7} \text{ cm}^2 \text{ s}^{-1}$  with increasing the diffusion annealing temperature. The major reason with the scenario of increased diffusion coefficient with the diffusion annealing temperature is in attribution to the fact that more and more Au individuals penetrate both into the superconducting grains and over the grain boundaries along with the Bi-2212 crystal lattice with the diffusion annealing temperature. In this respect, the optimum diffusion annealing temperature (leading to the introduction of optimum Au inclusions to the crystal texturing) is found to be about 800 °C for the improvement of the electrical, superconducting, physical and mechanical properties of Bi-2212 system. In other words, the optimum Au concentration primarily proceeds through the local structural distortions, dislocations, lattice defects and disorders in the Bi-2212 polycrystalline compounds.
- Furthermore, the temperature-dependent diffusion coefficient and related activation energy values of Au foreign impurities in the Bi-2212 system are calculated to be about  $2.601 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$  and 1.523 eV, respectively. According to the results obtained, rather lower diffusion rate and higher activation energy values result from the penetration of heavy metal ions of gold when the gold ions require the minimum activation energy value such as 1.523 eV to penetrate into the Bi-2212 crystal structure.

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