Influence of Ag/Cu Substitution on Structural Effect of New High Temperature Superconductor Y$_3$Ba$_5$Cu$_8$O$_{18}$

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Abstract

The class of superconductors ceramic materials has been made from copper oxides mixed with rare earth elements such as yttrium. The new Y-based high temperature superconductor Y$_3$Ba$_5$Cu$_8$O$_{18}$ (Y358), Gholipour et al. (2012), Tavana et al. (2010), Khosroabadi et al. (2014), with Ag/Cu substitution was synthesized through the sol-gel technique by making nano oxide Y, Ba, Cu and Ag. The influence of the doping of Ag atoms into the compound Y$_3$Ba$_5$Cu$_8$O$_{18}$ was studied by employing XRD pattern, SEM and the electrical resistivity measurements. The room temperature electrical measurements have shown that ceramic Y358 has metal behavior in the normal state. The compound of YBCO family was discovered containing the CuO$_2$ planes and the Cu-O chains. It seems that increasing the number of the CuO$_2$ planes and the position of the Cu-O chains have positive effects on the value of the transition temperature in the Y-based compounds. The analysis of the X-ray diffraction pattern was approved by MAUD software which the Ag ions substitute in symmetrical phase of the sample. The substitution of the Ag ions in the CuO$_2$ plane of symmetric phase has been compared with the Ag ions in the Y123 superconductor, which substitute in the Cu-O chain. The electrical resistivity of these superconductors was measured by 4prob method. The electrical resistivity graph approved the start of transition temperature above 100 K.

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1. Introduction

The effect of Ag doping concentration on the superconducting YBCO ceramics has been extensively studied in order to improve superconducting properties. The extremely research studies has been down on doping the YBCO family as YBa$_2$Cu$_3$O$_7$ (Y123) with transition temperature about 90 with one Cu-O chain and two CuO$_2$ planes, Wu et al. (1987). The superconductor ceramic Y$_6$Ba$_3$Cu$_6$O$_{18+\delta}$ (Y358) has transition temperature of 101 K, with difference of 10 K above the transition temperature of any YBCO family, Gholipour et al. (2012), Tavana et al. (2010), Khosroabadi et al. (2014). There are various structure presented for Y358. At first Structure with Pmmm2 that has five CuO$_2$ planes and three Cu-O chains and lattice parameter (a, b, c) = (3.845Å, 3.894Å, 31.093Å), Gholipour et al. (2012). Another crystal structure symmetry (pmmm) with 6 CuO$_2$ planes and 2 Cu-O chains and lattice parameter (a, b, c) = (3.838Å, 3.904Å, 31.043Å). This structure is formed from one block Y112 that is sandwiched between two Y123 unit cells. The advantages of silver doping may be related to its ability in the enhancement of the critical current density and increasing interconnections between the grains, Khosroabadi et al. (2014). The substitution or addition is very often employed as an effective means to control the electronic and the structural properties of solid state materials. The atomic arrangement in the Cu-O planes is the key factor to the electric transport in the superconducting YBCO ceramics. Therefore, it is worth to investigate the effects of a selected element which may replace Cu ions in the YBCO ceramics to modify the superconducting properties. The effect of Ag/Cu substitution on YBa$_2$Cu$_3$O$_7$ indicated the Ag doping improves the grain growth and their orientations, and modifies the inter-grain weak-links, Tepea et al. (2004). In the temperature processing of YBCO ceramics, liquid Ag fills into the pores of the structure and modifies the micro cracks, thus strengthening the role of pinning centers for the high current applications. The excess amount of silver plays a role of preventing factor for the growth of grains, Chuang et al. (1995). The effect of Ag in the Y123 superconductor, Cahen et al. (1987) researched substitution Ag in the Cu-O chain. In this study, we investigated the effect of Ag doping on superconductor Y$_3$Ba$_2$Cu$_{6-x}$Ag$_x$O$_{18-\delta}$ with doping value $x=0, 0.1, 0.2, 0.3, 0.4$ were synthesized through the sol-gel technique by making nano oxide Y, Ba, Cu and Ag. The characterization of the samples has been carried out by a susceptibility R–T, SEM measurements and the analysis of the X-ray diffraction pattern that was approved by MAUD software.

2. Experimental Detail

We synthesized samples of Y358 by the sol–gel technique. Appropriate stoichiometric compositions of Y: Ba: Cu = 3:5:8-$x$:$x$ (x = 0, 0.1, 0.2, 0.3, 0.4) of Cu(NO$_3$)$_2$.3H$_2$O (99.5%), Ba(NO$_3$)$_2$ (99 %), Y(NO$_3$)$_3$.5H$_2$O (99 %) and AgNO$_3$ (99%) powders were weighed at 10−5 g precision and solved in deionized water and we added these solutions to 0.5 M solution of citric acid (C$_6$H$_7$O$_8$). The pH of the final solution was adjusted to 6.88, Gholipour et al. (2012). Subsequently, this solution was evaporated on a stirrer hot plate maintaining the solution temperature at 83°C. The prepared gel was heated at 520 °C for 2 h in air atmosphere. Next, the resultant powder was calcined in air twice in order to remove the remaining nitrates from the specimen. The powder was pressed into pellets shaps. Then, these samples were calcined at 910–908 °C for 42 h in a controlled oxygen atmosphere with a specific thermal program. The ac electrical resistance measurements were made by using standard four-probe method and silver pasta and measured in the liquid nitrogen.
3. Characterization

We employ XRD, supplemented with the MAUD software based on the Rietveld analysis, to identify phases of our superconducting samples. X-ray action analysis was performed with Cu-Kα radiation (\( A = 1.5406 \) Å). Fig. 1a shows the measured and calculated XRD patterns of Y358, and their difference. The designated Miller indices and volume fraction phase is 83% of structure pmmm.

Fig. 1b shows the XRD patterns of \( Y_3Ba_5Cu_{8-x}Ag_xO_{18-G} \) with \( x = 0, 0.1, 0.3 \) samples. Table 1 listed the result of the refined atomic positions and occupancy in the \( Y_3Ba_5Cu_{8-x}Ag_xO_{18-G} \) samples. The lattice positions of occupancy number Cu(1) become less when the amount of substitution was increase.

| atoms   | Ag content=0 | Ag content=0.1 | Ag content=0.3 |
|---------|--------------|----------------|----------------|
|         | x     | y     | z     | occupancy | x     | y     | z     | occupancy | X    | Y    | z     | occupancy |
| Ba(1)   | 0.5   | 0.5   | 0.12  | 0.99     | 0.5   | 0.5   | 0.117 | 0.99     | 0.5   | 0.5   | 0.12  | 0.99     |
| Ba(2)   | 0.5   | 0.5   | 0.26  | 0.99     | 0.5   | 0.5   | 0.26  | 0.99     | 0.5   | 0.5   | 0.26  | 0.99     |
| Ba(3)   | 0.5   | 0.5   | 0.5   | 0.99     | 0.5   | 0.5   | 0.5   | 0.99     | 0.5   | 0.5   | 0.5   | 0.99     |
| Y(1)    | 0.5   | 0.5   | 0    | 1.00     | 0.5   | 0.5   | 0    | 1.00     | 0.5   | 0.5   | 0    | 1.00     |
| Y(2)    | 0.5   | 0.5   | 0.38  | 1.00     | 0.5   | 0.5   | 0.377 | 0.99     | 0.5   | 0.5   | 0.38  | 1.00     |
| Cu(1)   | 0     | 0     | 0.06  | 1.00     | 0     | 0     | 0.065 | 0.93     | 0     | 0     | 0.064 | 0.83     |
| Cu(2)   | 0     | 0     | 0.19  | 1.00     | 0     | 0     | 0.194 | 1.00     | 0     | 0     | 0.19  | 1.00     |
| Cu(3)   | 0     | 0     | 0.33  | 1.00     | 0     | 0     | 0.302 | 1.00     | 0     | 0     | 0.33  | 1.00     |
| Cu(4)   | 0     | 0     | 0.43  | 1.00     | 0     | 0     | 0.43  | 1.00     | 0     | 0     | 0.43  | 1.00     |
| O(1)    | 0.5   | 0     | 0.05  | 0.99     | 0.5   | 0     | 0.15  | 0.99     | 0.5   | 0     | 0.05  | 0.99     |
| O(2)    | 0     | 0.5   | 0.05  | 1.01     | 0     | 0.5   | 0.15  | 0.99     | 0     | 0.5   | 0.05  | 0.99     |
| O(3)    | 0     | 0     | 0.12  | 1.00     | 0     | 0     | 0.13  | 0.99     | 0     | 0     | 0.12  | 0.99     |
| O(4)    | 0     | 0.5   | 0.19  | 1.00     | 0     | 0.5   | 0.08  | 0.99     | 0     | 0.5   | 0.19  | 0.99     |
| O(5)    | 0     | 0.26  | 1.00  | 0     | 0     | 0.26  | 0.99  | 0     | 0     | 0.26  | 0.99     |
| O(6)    | 0.5   | 0     | 0.33  | 1.00     | 0.5   | 0     | 0.4   | 0.99     | 0.5   | 0     | 0.33  | 0.99     |
| O(7)    | 0     | 0.5   | 0.33  | 1.00     | 0.5   | 0     | 0.4   | 0.99     | 0     | 0.5   | 0.33  | 0.99     |
| O(8)    | 0.5   | 0     | 0.43  | 0.99     | 0.5   | 0     | 0.41  | 0.99     | 0.5   | 0     | 0.43  | 0.99     |
| O(9)    | 0     | 0.5   | 0.43  | 0.99     | 0     | 0.5   | 0.41  | 0.99     | 0     | 0.5   | 0.43  | 0.99     |
| O(10)   | 0     | 0     | 0.5   | 1.00     | 0     | 0     | 0.5   | 0.99     | 0     | 0     | 0.5   | 0.99     |
| Ag      | 0     | 0     | 0     | 0.065    | 0.08  | 0     | 0.07  | 0.3      |
The peaks for Ag phase weren’t observed in the specimen with x = 0.1, and 80% Ag occupied the lattice position of Cu(1) that clearly is shown in fig 2. The other ion Ag is out of the grain during sintering of the bulk. The twin peaks at 46.68° and 47.38° indicate the orthorhombic structure for x= 0, 0.1, 0.3 as we expect.

Table 2. lattice parameter of Y_{3}Ba_{5}Cu_{8-x}Ag_{x}O_{18-6} with x=0, 0.1, 0.3

| Samples | a   | b   | c   |
|---------|-----|-----|-----|
| x=0    | 3.838 | 3.904 | 31.043 |
| x=0.1  | 3.849 | 3.9006 | 30.928 |
| x=0.3  | 3.867 | 3.896 | 30.777 |

Fig. 2. Crystal structure of Y_{3}Ba_{5}Cu_{8-x}Ag_{x}O_{18-6}

Table 2 shows the lattice parameter of Y_{3}Ba_{5}Cu_{8-x}Ag_{x}O_{18-6} with x=0, 0.1, 0.3. The lattice parameters for samples did not deviate from an orthorhombic cell, thus indicating Ag doesn’t affects the orthorhombic structure of the Y_{3}Ba_{5}Cu_{8-x}Ag_{x}O_{18-6}.

The goodness of fit (S) is described by S = R_{wp}/R_{exp}, where R_{wp} is the weighted residual error and R_{exp} is the expected error. Table 3 shows the goodness of fit (S) of Y_{3}Ba_{5}Cu_{8-x}Ag_{x}O_{18-6} (with x= 0, 0.1, 0.4). Refinement has been continued until a convergence was reached for a value of S close to 1, which confirms the goodness of refinement. Fig. 3 shows the resistive transition R–T of the samples. It is clearly shown the influence of Ag doping in Y358; the transition temperatures were changed by the Ag content. As indicated in Fig. 3, the transition temperatures of the samples decrease from 100.5 to 98 K with the increase Ag, except x = 0.1 (Tc = 101k). The zero-resistivity critical temperature for x= 0.1 (101K) is slightly higher than that for the pure sample (100.5 K). The Tc of Ag-doped sample decreased with average thermal rate of 17 degrees Kelvin on content.

The resistivity of Y358 at normal state was decreased linearly with temperature that shows some Ag ions fill the space between the Y358 grains and decrease the porosity of the superconducting sample; this leads to a short-circuiting of the grains in the normal state. The less change in ∆Tc shows Ag ions has not created any impurity phase.
Table 3. The parameters for calculation fitness

| Samples | Rwp | Rexp | S   |
|---------|-----|------|-----|
| x = 0   | 0.044 | 0.03 | 1.46 |
| x = 0.1 | 0.048 | 0.03 | 1.601 |
| x = 0.3 | 0.047 | 0.03 | 1.58 |

Fig. 3. The electrical resistivity of the Y$_3$Ba$_5$Cu$_{8-x}$Ag$_x$O$_{18-δ}$ sample versus temperature

Table 4. The value of the resistivity Y$_3$Ba$_5$Cu$_{8-x}$Ag$_x$O$_{18-δ}$

| Sample | T$_c$(on) | T$_c$(off) | T$_c$(mid) | ΔT$_c$ |
|--------|-----------|-----------|-----------|--------|
| x=0    | 105.5     | 98        | 100.5     | 5      |
| x=0.1  | 105       | 97.5      | 101       | 4.5    |
| x=0.2  | 105       | 95        | 98.5      | 5.5    |
| x=0.3  | 101.5     | 95        | 97.5      | 4      |
| x=0.4  | 105       | 93        | 98        | 6      |

Figure 4 shows the surface morphologies of Y$_3$Ba$_5$Cu$_{8-x}$Ag$_x$O$_{18-δ}$ by scanning electron microscopy (SEM). If Cu ion is not replaced by Ag it, prevents the growth of grains. It is clearly seen from the micrographs that the grain sizes increase when the Ag placed on structure.

Fig. 4. The FE-SEM images of the Y$_3$Ba$_5$Cu$_{8-x}$Ag$_x$O$_{18-δ}$ samples. (a) x = 0.1, (b) x = 0.4
4. Conclusions

The high temperature superconductor $\text{Y}_3\text{Ba}_5\text{Cu}_{3-x}\text{Ag}_x\text{O}_{18.5}$ with doping value $x=0, 0.1, 0.2, 0.3, 0.4$ were synthesized through the sol-gel technique. In this study, we investigated the effect of Ag doping on the superconducting and transport properties of Y358 ceramics. The analysis of the X-ray diffraction patterns of our samples don’t show the peaks for Ag phase. The substitution of the Ag/Cu is in the CuO$_2$ plane of symmetric phase while the Ag/Cu in the Y123 superconductor was occurred in the Cu-O chain, Cahen et al. (1987). The transition temperatures of the samples decrease from 101 to 98 K and the increasing Ag. The normal state resistances of the samples have metallic behaviour.

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