Cryogenics 73 (2016) 1–7

Contents lists available at ScienceDirect

Cryogenics

journal homepage: www.elsevier.com/locate/cryogenics

Structural, electrical and mechanical properties of selenium doped thallium based high-temperature superconductors

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ARTICLE INFO

Article history:
Received 22 May 2015
Received in revised form 13 August 2015
Accepted 27 October 2015
Available online 2 November 2015

Keywords:
XRD
Se doping
Vickers microhardness
Resistivity

ABSTRACT

In this study, highly-refined chemical powders were synthesized by having them ready in appropriate stoichiometric proportions with conventional solid state reaction method so that they would produce the superconductor $TlPb_{0.3}Sr_2Ca_{1-x}Se_xCu_2O_y$ ($x = 0$; 0.4; 0.6; 1.0). This study aims to understand effect of the selenium doping on the superconducting, structural and mechanical properties of the aforementioned superconducting material. The effect of the doping rates on the structural and electrical properties of the sample has been identified. Electrical characteristics of the $TlPb_{0.3}Sr_2Ca_{1-x}Se_xCu_2O_y$ material were measured using standard four point probe method. Structural characteristics were examined with the powder X-ray diffractometer (XRD) and scanning electron microscope (SEM). Mechanical properties were analyzed with Vickers microhardness measurements on the sample surface. According to the results, it was observed that the reflection comes from the (*00l*) and parallel planes increased with Se doping. Particle size increases with increasing doping ratio. According to results of the mechanical measurements, all samples exhibit indentation size effect (ISE) behavior. Comparing the obtained results with theoretical studies, it was understood that Hays Kendall approach is the best method in determination of mechanical properties and analyzing microhardness of the materials.

1. Introduction

Ba–La–Cu–O based high temperature superconductors have been studied in some detail since its discovery by Bednorz and Müller [1]. Tl–Ba–Ca–Cu–O based superconductors have been mostly studied by Sheng and Hermann [2]. In the last 25 years many researchers studied on the effects of Cu–O chain on the $TlBa_2Ca_{n-1}Cu_nO_{2n+3}$ and $Tl_2Ba_2Ca_{n-1}Cu_nO_{2n+4}$ systems containing different Cu–O layers. Sheng et al. [3] reported the critical temperature value as 20 K at Tl–Sr–Ca–Cu–O system. Hayri and Greenblatt determined that the critical temperature decreases from 110 K for $x = 0$ to 20 K for $x = 2$ at $Tl_2Ba_{2-x}Sr_xCaCu_2O_8$ system with Ba–Sr partial substitution [4]. Valldor et al. investigated how the structural and electrical properties were changed by Tl–Hg partial substitution in 2212 Tl–Hg–Sr–Ca–Cu–O system. It is seen that the critical temperature values were in the range of 25–45 K for the samples they produced [5]. Subramania et al. reported that the critical temperature can increase to 115 K depending on Cu–O chain in Tl–Pb–Sr–Ca–Cu–O system [6]. Cigan et al., investigating the low La doped TlPb-1223 structure, found the critical temperature value in the range 113–116 K, for samples including Ba–Sr [7]. It is well known that, partial substitution with Tl–Ba and Pb–Sr makes the structure more stable and improves some properties like critical temperature [8]. On the other hand, substitution or doping in excess leads to negative behavior on the structure. For this reason both mechanical and electrical properties of the structure are affected. The reason of this affect is based on the valance and oxidation numbers [9]. The isovalent substitution of Ba for Sr increases the charge transfer from the charge reservoir layers to the $Cu–O_2$ planes and so hole concentration of the $Cu–O_2$ planes can be changed. $Cu–O_2$ can be seen as a key of superconductivity for high temperature superconductors. Oxygen deficiency or number of oxide layer in the structure is directly affects the superconductivity parameters. Low or high sintering temperatures during production, affects the oxygen content of the structure and affects the superconductivity properties creating distortions in the crystal structure.

Mechanical properties like, microhardness, elastic modulus (E), yield strength (Y), fracture toughness (K_{IC}) and brittleness are great importance for the investigation of superconducting applications. Many studies on superconductors with Vickers microhardness test

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http://dx.doi.org/10.1016/j.cryogenics.2015.10.019
0011-2275/

method has been reported in the literature [10–12]. Khalil investigated the mechanical properties of Ca–Y partial substitution on $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_{8-d}$ system and reported that hardness were increased at low Y concentration and the most hard structure were formed at $x = 0.35$ [13]. Celik et al. used the Vickers microhardness test method and applied different theoretical models for $(Sm123)_{1-x}(Nd123)_x$ superconducting system [14]. Dogruer et al. analyzed the effect of Cu diffusion on microhardness at MgB_2 bulk superconducting structure with Vickers microhardness test method and compared with the results obtained from theoretical methods as Meyer's, proportional sample resistance (PSR), modified PSR (MPSR), elastic–plastic deformation (EPD) models and Hays–Kendall (HK) approach [15]. Mohammed et al. analyzed the change of electrical and mechanical properties of nano-Fe_2O_3 doped $(Cu_{0.5}Tl_{0.5})$-1223 high temperature superconductor [16]. They reported that, critical temperature value decreases and microhardness increases with doping. Awad et al. found that, microhardness value of $(Cu_{0.25}Tl_{0.75})$-1234 superconductor system increases with nano-MgO doping and decreases with doping above 0.6 wt% [17]. In the literature there are very few studies investigating the mechanical properties of Tl–Pb–Sr–Ca–Cu–O structure. Especially, effect of Se doping on mechanical properties of this structure has not been analyzed before.

In this work, effects of Se doping on structural, electrical and mechanical properties of $TlPb_{0.3}Sr_2Ca_{1-x}Se_xCu_2O_y$ ($x = 0$; 0.4; 0.6; 1.0) bulk superconducting structure were systematically investigated.

2. Experimental details

A well-known solid state reaction method were used to prepare the Se doped Tl-1212 superconductor. Commercial Tl_2O_3, PbO, SrO, CaO, CuO (Alfa Aesar) and Se (Sigma–Aldrich) powders were used to prepare the samples. 2.5 g powders were prepared for each sample. Se powder, substituted with Ca at the ratios of 0.4, 0.6 and 1.0, were added into the mixture. For simplicity, we will use the abbreviations TPSO-00, TPSO-04, TPSO-06 and TPSO-1 for the samples with $x = 0$, 0.4, 0.6 and 1.0, respectively. All chemicals were mixed in the proper ratios in an agate mortar for 1 h to form the 2.5 g samples. Obtained mixtures were pressed with 980 MPa pressure to form discs of 10 mm in diameter. Tablets formed were clung to gold plate and sintered at 860 °C under an oxygen flow for 4 h in a programmable tube furnace. The samples were allowed to cool down to the room temperature in the furnace. The cold tablets were crushed and powdered again. The resultant powder again pressed to form new tablets after mixed in the agate mortar for 0.5 h. the new formed tablets were sintered again at the same conditions at 860 °C for 2 h. This process was repeated for every ratio of Se doping.

The crystalline phases were characterized by Bruker D8 Advance X-ray powder diffractometer using Cu Kα radiation in the range of $3° \leqslant 2\theta \leqslant 80°$ at a scan speed of 2°/min. TESCAN brand VEGA3 SB model scanning electron microscope (SEM) were used to examine the nature surfaces of the samples.

Resistivity measurements of the samples, cut into a rectangular bar shape, were performed using four-point contact technique made with silver paste in order to minimize the contact resistance. Temperature dependence of resistivity of the samples was measured in a cryostat by running 10 mA direct current in the temperature range 40–150 K. The critical temperature of the samples, viz, the transition temperature (T_c^{onset}) is the temperature value where the resistivity decreased sharply and superconductivity started.

In order to investigate the effect of Se doping on the mechanical properties, Vickers microhardness measurements were performed on the sample surfaces at the room temperature using Shimadzu brand HMV-2 model digital microhardness tester. The applied load was changed in the range 0.245–2.940 N for 10 s. Microhardness values were determined with an average of 5 readings at different parts of the sample surfaces making sure that the indentations do not overlap.

3. Result and discussion

3.1. Structural characterization

3.1.1. XRD analyses

The XRD patterns of the Se doped Tl-1212 superconductor samples were shown in Fig. 1. Crystal structures were calculated with Bruker-EVA 14.0.0.0 analyzing program including ICCD PDF2-2002 library. Tl-1212 (01-086-0458-ICCD) tetragonal structure as main phase, Tl-1223 (00-044-0251) high T_c phase, Tl_2O_3 (00-001-0849-ICCD) and $CaSrCu_3O$ (00-048-0761-ICCD) as impurity phases were determined for undoped (TPSO-00) and Se doped (TPSO-04, TPSO-06 and TPSO-1) samples. From Fig. 1, it is seen that the peak intensity of the samples belonging to (*00l*) increases with doping. In particular, the peak intensities from (001) to (006) increased very much (Fig. 2). No serious change was observed for peaks belonging to other planes. The increase of the peak intensity is related with the increase of the grain size. The peak intensity, reflected from the surface, increase with the increase of the grain size. Furthermore, the SEM results show us the increase of the grain size and the decrease of the porosity. Eventually, we can say that, the peak intensity increase with increasing grain size.

Lattice parameters, calculated from XRD peaks were given in Table 1. Significant change in lattice parameter *a* was not observed. But, a slight reduction has been seen in parameter *c*. Hence, lattice volume changed accordingly.

3.1.2. SEM analyses

Fig. 3 shows the SEM images of TPSO-00, TPSO-04, TPSO-06 and TPSO-1 samples. Surface structure, orientations, grain sizes and accumulation in the grain boundaries were investigated from these micrographs. It can be seen from Fig. 3 that, surface morphology of the samples changed with doping. Samples show non-homogenous small or large granular structure. The grain sizes increase and porosity decrease with Se doping.

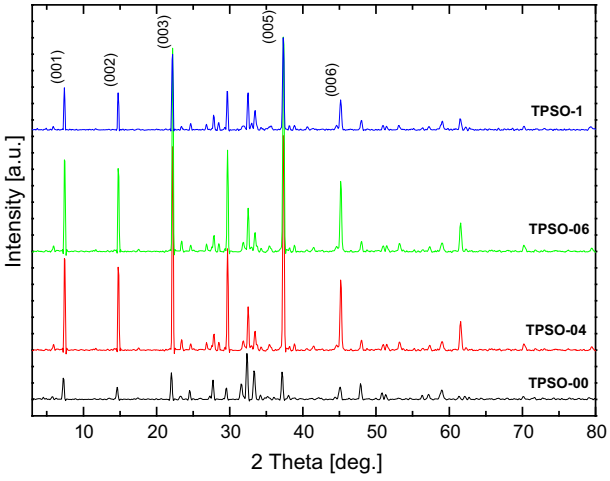

Fig. 1. XRD patterns of *TPSO-00*, *TPSO-04*, *TPSO-06* and *TPSO-1*.