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The effect of different dwell times in the ball milling process on the superconducting properties of $Bi_{1.8}Sr_2Ca_{1.1}Cu_{2.1}O_y$ (Bi-2212) ceramics

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Abstract

In this paper, the effect of the applied different dwell times (1, 4, and 8 h) during ball-milling on the basic properties of Bi-2212 ceramics has been investigated. $Bi_{1.8}Sr_2Ca_{1.1}Cu_{2.1}O_y$ was selected as the initial composition to obtain the high amount of Bi-2212 phase. All samples have been prepared by the conventional solid state reaction method. It has been observed that increasing dwell time negatively affected the superconducting properties of Bi-2212 samples because of the formation of structures with weak grain links. The magnetic properties of samples were determined from the magnetic hysteresis loops (*M-H*) between $\mp$ 2 T at 10 K, showing that the width of *M-H* loops has been significantly decreased with increasing dwell time.

In addition, the magnetic critical current densities (J_c) of the samples were calculated from their magnetic hysteresis loops using the Bean's model. The highest value of J_c has been observed in the sample exposed to dwell time of 1h.

Keywords: $Bi_{1.8}Sr_2Ca_{1.1}Cu_{2.1}O_y$; XRD; SEM; Magnetic-hysteresis loop; Critical current density

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1 Introduction

The high emissions of dangerous gases such as carbon dioxide and carbon monoxide, which are released when fossil fuels are burned to obtain energy, are among the most important causes of the destruction of global ecosystem. Moreover, the consumption of the energy as the consequences of both developing technology and global population growth is increasing significantly. Thus, the efficient use of energy as well as its production is also vital for the prevention or reduction of environmental pollution.

It is well known that a significant portion of the generated energy disappears by heat due to the electrical resistance of transmission wires while the energy produced in the dams is carried by electric wires, causing serious energy wastage.

After the discovery of superconductors ensuring the flow of current without electrical resistance, scientists' greatest dream has been to reach superconductors at room temperature. Even though we cannot reach the superconductors at room temperature today, they are used in very important technological areas needing high electrical transport properties at high magnetic fields [1-4]. It is well known that BiSrCaCuO system with the general formula $Bi_2Sr_2Ca_{n-1}Cu_nO_{2n+4+y}$ is abbreviated as BSCCO and includes three different phases expressed as Bi-2201 (n=1), Bi-2212 (n=2) and Bi-2223 (n=3) according to n values showing the number of CuO_2 in its unit crystal cell [5, 6]. YBCO ensuring the pratical use of superconductors in electronic devices is also another important high temperature superconductor [7]. YBaCuO system with the general formula $Y_2Ba_4Ca_{6+n}O_{14+n+\delta}$ is abbreviated as YBCO and includes three different phases such as Y-123 (n=0), Y-247 (n=1) and Y-124 (n=2) as based on both n values showing the corresponding CuO chains in their unit cells and the molar ratios of their metallic atoms [8, 9].

Especially, the discovery of those high-temperature superconductors provides a significant decrease at high spending costs due to the cooling procedure using liquid nitrogen instead of liquid helium in order to achieve superconductivity.

Thus, the main goal of many studies is to improve the basic properties of existing superconductors until new superconductors close to room temperature are discovered. On the other hand, Bi-2212 phase in the BSCCO system

among other high-Tc superconductors is suitable to work because of its advantages such as high thermodynamic stability, relatively cheaper production, and environmentally less harmlessness. It is well known that the doping or adding of chemical elements to the BSCCO system at appropriate rates is a very effective method to improve its T_c, H_c and J_c capacity [10-24]. Moreover, the enhancements in the physical and magnetic properties of high-T_c superconductors strongly depend on parameters such as the amounts of porosity, impurities, and grain orientations in the structure as well as the high-level formation of the desired phases such as Bi-2212 and Bi-2223.

The main goal in the improvement of superconductors is to reach denser granular structures with both regular grain orientation and less porosity, implying the importance of material preparation methods. Some studies clearly show that high critical current density (J_c) values in BSCCO system can be increased by the use of methods such as laser floating zone (LFZ) and the electrically assisted laser floating zone (EALFZ) controlled regularly the orientations of the grains [25-29]. Nevertheless, superconducting samples can also be prepared by using conventional solid-state reaction method due to its simple processes such as mixture of raw materials, the pressing of powders, and sintering steps.

Additionally, the determination of optimal values for the parameters used in the solid-state method such as sintering, pressing, etc. is extremely important in improving the superconducting properties. In previous studies, the researchers focused on the effects of applied pelletization pressure during the material preparation process, showing that the higher J_c values in BSCCO superconductors can be obtained by optimization of the pelletization pressure [30-33]. Another important part of work in the literature is also based on the optimization of the heat treatment process of the BSCCO system to achieve the best Jc values with the creation of new effective pinning centers or with the formation of the high Tc phases at high rates [34-38].

The aim of this study is to examine the effects of the dwell time in the ball milling process to find its optimal values. The basic properties of prepared superconducting samples are investigated by the X-ray diffraction (XRD), the scanning electron microscopy (SEM), resistivity (ρ-T), and magnetic hysteresis loops (*M-H*) in detail.

2 Experimental details

High purity powders of commercial Bi_2O_3 (Panreac, 98+%), $SrCO_3$ (Panreac, 98+%), $CaCO_3$ (Panreac, 98.5+%), and CuO (Panreac, 97+%) were used for the preparation of samples in this study. Polycristalline samples with nominal composition $Bi_{1.8}Sr_2Ca_{1.1}Cu_{2.1}O_y$ were prepared by the standard solid-state reaction methods. Firstly, they were weighed according to the stoichiometric formula of $Bi_{1.8}Sr_2Ca_{1.1}Cu_{2.1}O_y$ with total amount of 12 g. Precursor powders were then poured into a cylinder container with approximately 4 cm in diameter and 5.8 cm in height. The acetone at the appropriate amount was added to the powders to facilitate mixing. In the grinding process, different ball milling times using 1, 4, and 8 h in acetone with five ZrO_2 balls at 1.2 cm diameter were applied to the mixture. The different grinding times in the ball milling process were used only once during sample preparation. In the following stages for the other producing process of the materials, an agate mortar during one hour was used for the grinding of the tablets. The purpose of the ball milling used at the beginning was to better see its effect on the formation of Bi-2212 phase.

Taking into account the applied dwell times, the samples were herein after denoted as A, B, and C, respectively.

After the milling process, the homogenous mixture of powders was pressed into pellets of 2.9 cm diameter by applying a 375 MPa pressure for 2 h at room temperature and then calcined at 750 ^oC for 12 h. The calcined pellets were reground, repressed, and recalcined twice at 820 ^0C for 24 h to start the formation of the superconducting phase. It was obvious from the literature that it was possible to reach larger *M-H* curves in BSCCO system when the pelletizing process was repeated [22, 23]. Thus, these processes based on the milling, sintering, and pressing were repeated two times. Finally, precursor materials were ground and repressed, and annealed at 850 ^0C for 120 h in order to reach a large amount of pure 2212-BSCCO.

The Resistivity and magnetic measurements were carried out on samples using Cryogenic Limited PPMS (from 5 to 300 K) , which can reach the cryogenic temperatures about to 2 K in a closed-loop He system. X-ray powder diffraction analyses to determine the phases present in the samples were performed by using a Rigaku Ultima IV X-ray diffractometer with a constant scan rate (2^o/min)

in the range 2θ = 3–60°. The surface morphologies of the samples were studied by using a Zeiss/Supra 55 Scanning Electron Microscopy (SEM).

3 Results and discussion

3.1 XRD studies

The XRD pattern for all samples is shown in Fig. 1. It is clearly seen from Fig.1 that there are diffraction peaks belonging to Bi-2212 phase at high rates in all samples, meaning that the crystallization process of samples is completed successfully.

On the other hand, different impurity phases in all samples are also observed as depending on the period of grinding. It is worth noting that impurity phases can significantly cause narrow areas in M-H hysteresis loops as well as low intergrain conductivity, which means the deterioration of superconducting properties in samples.

In addition, Fig.1 clearly shows that the intensities and width of the XRD peaks belonging to the characteristic Bi-2212 phases in all samples are almost the same. Note that sharper and narrower diffraction peaks in XRD graphics generally are a sign of high crystallization [39, 40]. Thus, the similar XRD peak intensities seen in all samples reflect that they have the similar crystallinity as well as the formation of same phases.

These results mean that dwell time in the ball-milling system does not significantly affect the growth of Bi-2212 phases. However, one can expect from powers compressed among high-energy collisions of balls during milling that grain' size and crystallization in the structure can change effectively [41].

Despite long grinding processes, the problem of the formation of similar grains can depend on the high amount of balls used in this study since a large number of balls in a small volume can cause only uniform dispersions of oxide particles without any size-reduction, which is the most important effect expected from grinding systems.

On the other hand, automatically obtained lattice parameters a, b, and c are listed in Table 1. It is obvious from table 1 that c parameter for all samples shows the ideal bond length between two Bi-O layers in the Bi-2212 unit cell, meaning that it is suitable to interpret the effect of the different dwell time used in our grinding system on the Bi-2212 phase formations.

3.2 SEM analyses

SEM micrographs for all samples are shown in Fig. 2a-c. It is well known that scanning electron microscope (SEM) gives us important information about the surface morphology such as grain connectivity, the determination of the different phases, and observation of formed pores. Moreover, it is observed from Fig. 2 that all samples have porosity in their structures, leading to the degrade of both crystalinity and electrical conductivity between the superconducting grains. On the other hand, all samples have similar grain orientations and sizes in accordance with XRD measurements, implying that the behavior of their hysteresis curves will be similar even if they have different areas in their M-H loops. It is obvious that all samples have randomly oriented plate-like grain morphology. Also, the formation of randomly oriented grains in samples causes different grain boundaries, showing that the granular structure due to applied different dwell time can be significantly changed.

3.3 Electrical measurements

Figure 3 shows the electrical resistivity versus temperature curves for all the samples, from 5 to 150 K. It is clearly seen from Fig. 3 that all samples have high transition temperature width (ΔT_c) range even if they show the trace of nearly one step transition between their T_c (onset) and T_c (offset) critical temperatures (see Table1). Transition temperature in type II superconductors strongly depends on the degree of crystalinty, defects, and porosity in the structure. Thus, low T_c (offset) values seen in samples indicate that there are important problems in samples due to negative parameters such as poorly connected grains, high porosity, and irregular grain orientations. The results show that sample A has the best electrical and physical properties compared to other samples, showing that a good grinding process with the increased dwell time has not been performed sufficiently although a homogenous mixture is provided, presumably due to the high number of used grinding balls.

3.4 Magnetic properties

The magnetic-hysteresis cycles, between applied fields of ± 2 T, for all the samples at 10 K, are presented in Fig. 4. As seen from Fig. 4, all samples show characteristic diamagnetic properties of HT_c superconductors. However, the magnetization significantly decreases with the increasing of dwell time. It is seen that sample A including the ball milling time of 1h has the best superconducting properties compared to other samples due to its largest hyteresis loops. The superconducting properties in samples start to deteriorate due to remarkably decreasing widths of hysteresis curves with long dwell time. On the other hand, it is well known that the average size of a crystal can be easily calculated by the Debye-Scherrer equation [42].

Moreover, the full width half maximum (FWHM) values, which can be obtained by taking half value of the peak range in XRD patterns, are necessary to calculate the grain size. It has been clearly seen in Fig. 2 that the parameters such as XRD peak positions, peak range, and peak intensity for Bi-2212 phase in all samples do not change considerably, meaning that all samples have similar grain size. Thus, the narrow areas of *M-H* hysteresis loops seen in both sample B and C should be related to the formation of impurity phases in different types, weaker conductivity between grains, and the amount of voids instead of the formation of different grain sizes.

The J_C values of the samples were calculated at T= 10 K, using the Bean's model [43]:

$$J_C = 30 \frac{\Delta M}{d}$$

where J_C is the magnetization current density in ampéres per square centimeter of a sample. $\Delta M = M_+ - M_-$ is measured in electromagnetic units per cubic centimeter, d is the diameter of cylindrical samples.

Figure 5 shows critical current density (J_c) values calculated according to the *M-H* loops for all samples. As seen from Fig. 5, Jc values significantly decrease with increasing dwell time. The low J_c values observed in sample B and C are based on their narrow *M-H* loops, which means that energy stored in the ball milling and pressing stages during material preparation in these samples is very

low. On the other hand, different grain sizes in the structure can cause different pore-sizes due to the regularities of the grains, implying low J_c values. It is obvious that our samples produced in this work have similar grain sizes, causing similar intergranular properties together with similar porosity in samples. However, very low J_c values are observed in both samples B and C, indicating that different grain boundaries and weak links between grains are as effective as the formation of pores and its distribution.

4 Conclusions

In this study, we have observed the effect of different dwell times (1, 4, and 8 h) in ball-milling procedure on the physical and magnetic properties of Bi-2212 phase. XRD data and SEM results clearly show that all samples have the high volume ratio of Bi-2212 phase, indicating that homogenous mixing is achieved by the applied method. However, there are significant differences in both values of critical current density and fields of M-H curves seen in samples due to the ball milling processes, indicating that optimization of the ball milling is also required to get the best superconductivity properties.

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Figure captions

Figure 1. XRD patterns of the A, B and C samples. The symbols indicate the different phases: **+** Bi-2212;* $Bi_4Sr_2Ca_2Cu_4O_{14+x}$; Δ $Bi_2Ca_2O_4$; ♦ Bi_2CaO_4

Figure 2. SEM micrographs obtained in the surfaces of a) A; b) B; and c) C samples

Figure 3. Electrical resistivity as a function of temperature curves for all the samples.

Figure 4. Magnetization hysteresis curves for all samples measured at 10K and ±2 T external applied magnetic field.

Figure 5. Calculated critical current densities for all the samples at 10K as a function of applied field.

Table captions

Table 1. Lattice parameters and resistivity measurement results for the samples

Samples	a(Å)	b(Å)	c(Å)	T_c^{onset} (K)	T_c^{offset} (K)	R (mohm-cm) at 150 K
A	5.414	5.395	30.793	78.5	50.8	1.73
B	5.4	5.4	30.785	80.2	49	2.61
C	5.399	5.399	30.778	70.7	44.4	3.15