The Effect of Sintering Temperature on The Rolled Silver-Sheathed Monofilament Bi,Pb-Sr-Ca-Cu-O Superconducting Wire

Hendrik, P Sebleku, B Siswayanti and A W Pramono

Research Center for Metallurgy and Materials-Indonesian Institute of Sciences (P2MM - LIPI), Kawasan Puspiptek, Serpong, Tangerang 15314, Indonesia

Email: hend027@lipi.go.id

Abstract. The manufacture of high critical temperature (*Tc*) Bi, Pb-Sr-Ca-Cu-O (HTS BPSCCO) superconductor wire fabricated by power-in-tube (PIT) is a multi-step process. The main difficulty is that the value of *Tc* superconductor wire determined by various factors for each step. The objective of this research is to investigate the effect of sintering parameters on the properties of final rolled material. The fabrication process of 1 m rolled-silver sheath monofilament superconductor BPSCCO wire using mechanical deformation process including rolling and drawing has been carried out. The pure silver powders were melted and formed into pure silver (Ag) tube. The tube was 10 mm in diameter with a sheath material: superconductor powders ratio of about 6 : 1. Starting powders, containing the nominal composition of Bi_2-Sr_2-Ca_{m-1}-Cu_m-O_y, were inserted into the pure silver tube and rolled until it reached a diameter of 4 mm. A typical area reduction ratio of about 5% per step has been proposed to prevent microcracking during the cold-drawing process. The process of rolling of the silver tube was subsequently repeated to obtain three samples and then followed by heat-treated at 820 °C, 840 °C, and 860 °C, respectively. The surface morphology was analyzed by using SEM; the crystal structure was studied by using X-RD, whereas the superconductivity was investigated by using temperature dependence resistivity measurement by using four-point probe technique. SEM images showed the porosity of the cross-sectional surface of the samples. The sample with low heating temperature showed porosity more than the one with high temperature. The value of critical temperature (Tc) of the sample with a dwelling time of heating of 8 hours is 70 K. At above 70 K, it shows the behavior of conductor properties. However, the porosity increased as the heating time increased up to 24 hours. The critical temperature was difficult to be identified due to its porosity. According to XRD results, the Bi-2212 phase is prominent in all samples.

Keywords: High-temperature superconductor, powder-in-tube, BPSCCO, Bi-2223, Bi-2212 phase

1. Introduction

Since conventional NbTi and Nb_3Sn low-temperature superconductors dominated the commercial market which was applied to magnetic resonance imaging (MRI) and high-energy physics, the researchers began trying to apply Bi-2223 and REBCO high-temperature superconductors (HTS) for high-resolution NMR, maglev trains, MCZ, as well as magnetic separation of superconducting thin films such as electronic devices and transmission cables for power cables [1-3]. Many attempts in the last ten years have been done to observe the behavior of Bi-2223 / Ag superconducting tape [4-6], while these observations on the superconducting wire were not optimal due to the behavior of the wire drawing to produce porous wire simultaneously with the increase of sintering time. Therefore, in this paper, we will study the behavior of superconducting properties in the form of wire, especially in the Bi-2212/Ag.

The Ag-BPSCCO superconducting wire is very potential to be used to carry electricity for large-scale applications especially power cables. In Ag-BPSCCO superconducting wire, there are several phases system that can be formed. Whereas, the Bi-2223 phase is difficult to make due to its phase formation within a narrow temperature range, other superconducting phase such as Bi-2201, mixture of Bi-2201 and Bi-2212, and Bi-2212 can be easily formed.

To realize the full application of HTS, many issues should be overcome related to powder in tube (PIT) method and mechanical deformation process. To produce Bi-2212 phase/Ag superconducting wire, in-situ PIT method was chosen of which a powder with a nominal of composition of Bi-2223 was filled into Ag-tube and then mechanically deformed into the thin composite wire.

2. Experimental Method

Based on the stoichiometry, to get the phase of $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_2O_{8+\alpha}$, the weight ratio of materials are Bi_2O_3 : $Pb(NO_3)_2$: $SrCO_3$: $CaCO_3$: CuO = 3.32: 1.18: 2.63 :1.0 :2.07. Weighing followed by mortal grinding was carried out. The powder mixture was then inserted into silver tube followed by rolling.

2.1. Mechanical deformation.

A pure silver tube (ø inner tube = 4 mm; ø outer tube = 6 mm) was used for the rolling process. The tube was then filled with a mixture of Bi_2O_3, $Pb(NO_3)_2$, $SrCO_3$, $CaCO_3$ and CuO powders. An area reduction ratio of about 5% to 4 mm in diameter was carried out. The ratio of sheath material to superconductor fixed at 6:1.

2.2. Sintering.

Three samples were rolled and sintered at 820, 840, and 860 °C for about 8 hrs (heating rate 5 °C/min), respectively. Other samples were heat-treated at 860°C, and then the temperature was held for 24 hours. Cooling to room temperature in the furnace was achieved in about 24 hours.

2.3. Resistivity measurement.

To confirm the nature of the superconductivity of superconducting tape, the samples were measured by using a four-point probe (Figure 1). To expose the powder of superconducting wire to an electric current, the sample was mounted in a resin and cut for one side so that the surface of BPSCCO can be attached via four terminals by using Ag paste.

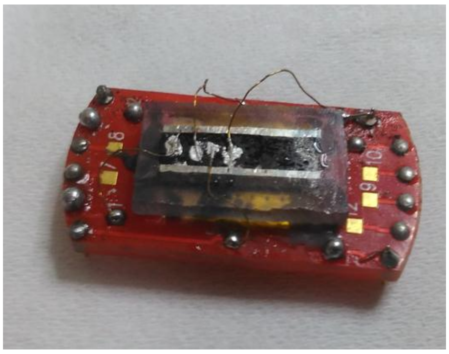

Figure 1. Image of sample Ag-BPSCCO exposed to four points probe

2.4. SEM JEOL model 6390A and XRD.

The morphological analysis of the sample was performed by using scanning electron microscopy (SEM). X-ray diffraction analysis was conducted to investigate the crystal structure of superconducting wire.

3. Results and Discussion

The process of sintering at 840 to 860 ^{o}C temperature range forms two-phase systems which are Bi-2212 and Bi-2223 as also found in the literature [7]. However, there are possible mixture of two phases when the result of experimental research shows it is difficult to make Bi-2212 single phase material. It is indicated by two modes situated on the curve Bi-2201 at Tc zero (20 K) and *Tc* onset (70 K) as observed in Figure 2.

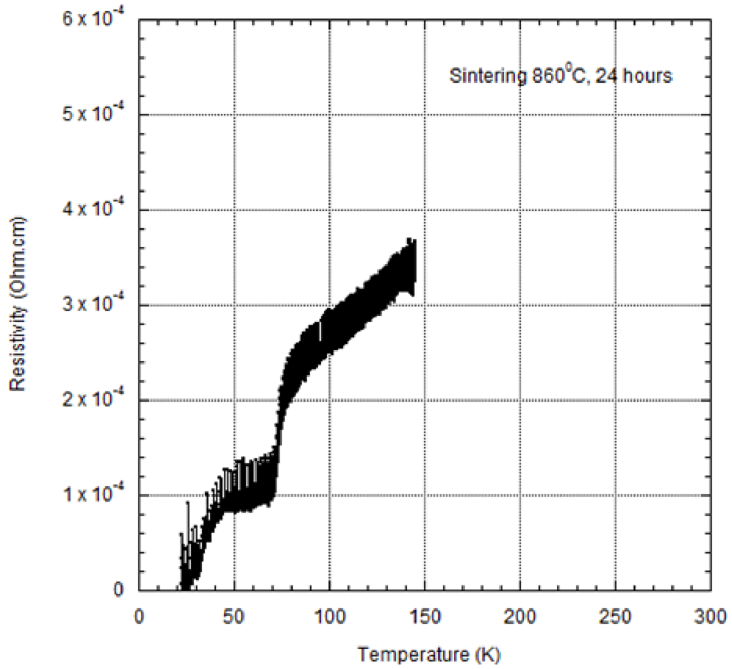

Figure 2. Resistivity dependence on temperature for the sample with sintering temperature at 860°C for 24 hours.

Figure 2 depicts our measurements of electrical resistance versus temperature. It points out that after long sintering time of 860°C sintering temperature there is a growing of Bi-2201 together with Bi-2212. This phenomenon suggests that it is likely Bi-2201 phase is a main phase. We also observed the data points in the ρ-T curve of a sample sintered for 24 hours (Figure 2) is wider than the one in the curve ρ-T of a sample sintered for 8 hours (Figure 3). Several factors can cause this discrepancy. Since the sample was cooled down from room temperature to 1.5 K, the surface cooled faster than the middle of superconductor, hence it is possible to get wider data points and the transition temperature.

Figure 3 depicts our measurements of electrical resistance versus temperature in the three sample wires. It is noted that with regard to 8 hours sintering time sample having a lower sintering temperature of 820°C exhibited no superconductivity behavior. In contrast to the one with lower sintering time, both samples with higher sintering temperature of 840°C and 860°C tend to shift toward superconducting phase having Tc onset 80 K and 85 K, respectively. The obtained value for the sample sintering temperature of 840°C and 860°C is in good agreement with literature data [8-10]. This phenomenon suggested conductor-superconductivity transition behavior occurred at 840°C.

Figures 4 and 5 revealed that the surface morphology of the cross section of wire samples after they have been cut. The image points out that the samples sintered for 8 hours (Figure 4) have less open channel than the sample sintered for 24 hours (Figure 5). The grains of the sample which was sintered for 24 hours are smaller and brighter in contrast to those sample sintered for 8 hours (Figure 4). Sintering in the air atmosphere can be accompanied by the formation of gas bubbles since the bubbles formed in the PIT process are intrinsic in the wire. Several authors claim that the use of overpressure may reduce the gas agglomeration as a source of current flow damage. Hence, it is proposed that the intermediate process like rolling and heat treatment is beneficial to produce the superconducting grains. In addition, sintering under vacuum environment can be used to prevent gas bubble formation.

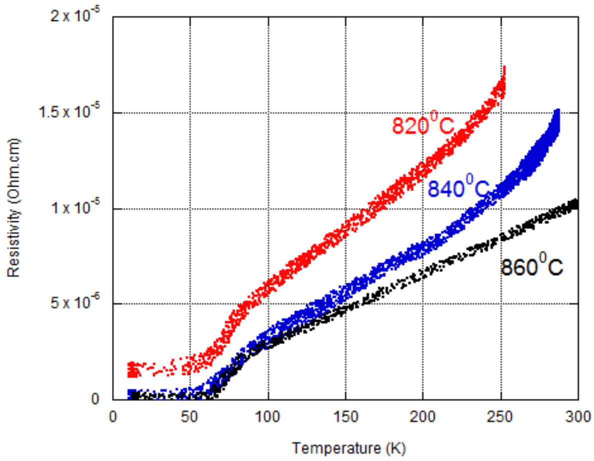

Figure 3. Resistivity dependence on temperature for samples with sintering temperature at 820°C, 840°C and 860°C for 8 hours.

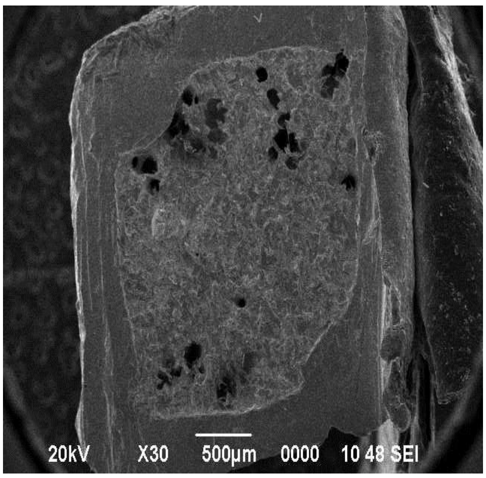

Figure 4. The surface morphology of cut sample of Ag-BPSCCO wire with 8-hour sintering time.

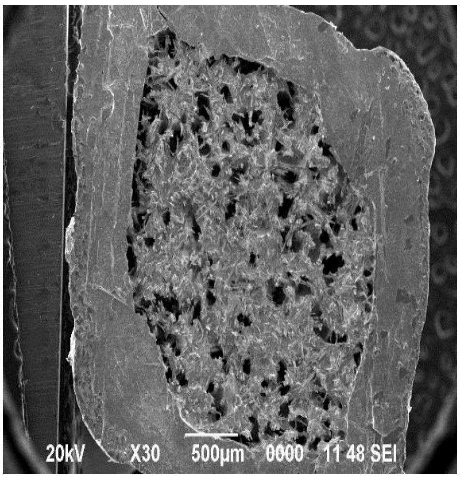

Figure 5. The surface morphology of cut sample of Ag-BPSCCO wire with 24-hour sintering time.

Figure 6 shows the XRD patterns of samples with a sintering temperature of 820, 840, and 860°C. Almost single phase of Bi-2212 compounds was detected for sample sintered at 820, 840, and 860°C, along with the small bump at 20° that related to resin during sample preparation.

It is interesting to note that there are various transitions of Bi-2212 with respect to the change in sintering temperature and sintering time. This phenomenon suggests the sample structure of Bi-2212 is disoriented and higher temperature leads to ordering of aligned structure in the copper-oxygen plane. By increasing temperature up to 860°C, upon melting and solidification of the typical of the superconducting grains of Bi-2212 are optimally formed and thus aligned crystallographically in the copper-oxygen plane so that its resistivity reached zero resistance.

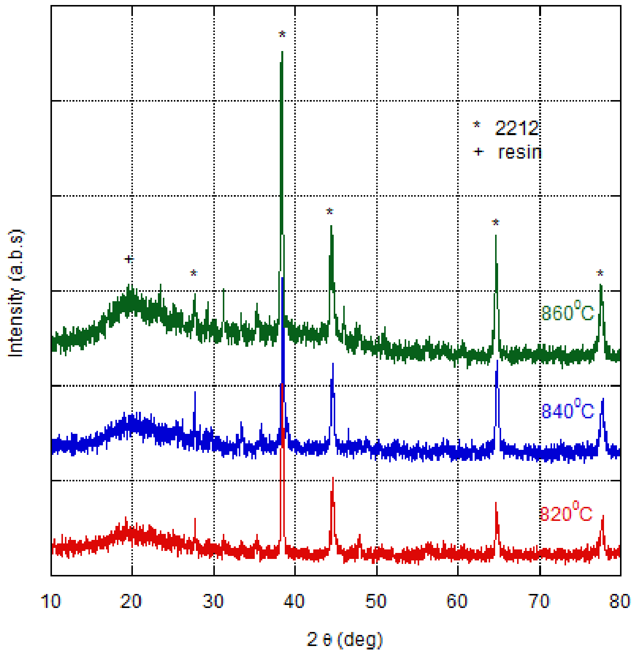

Figure 6. XRD result of sample of Ag-BPSCCO with a sintering temperature of 820°C, 840°C and 860°C for 8 hours.

4. Conclusion

Four samples of Bi,Pb-Sr-Ca-Cu-O silver-sheathed monofilament superconducting wire with the diameter of 4 mm and a length of 1 meter has been fabricated by the rolling process. Those samples were analyzed by the superconducting properties to provide a favorable path for further processing in the manufacture of superconducting wire. The effect of sintering temperature on crystalline phase formation and resistivity has been studied. The sample sintered at 860°C for 24 hours showed the dominant existence of Bi-2212 phase. Nevertheless, the sample sintered at 820°C for 8 hours has not yet formed a superconducting phase. It was clear that sample sintered at 840°C and 860°C showed that the critical temperature (Tc) shifted to zero resistance (0 ohms) indicated the Bi-2212 phase as also confirmed by X-RD data. All samples sintered for 8 hours are less porous than the one sintered for 24 hours.

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