Enhancement of Phase Formation and Critical Current Density in (Bi,Pb)-2223 Superconductor by Boron Addition and Ball Milling

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Abstract: The effects of crystalline boron addition and ball milling on the phase formation and transport properties of (Bi,Pb)-2223 HTS have been studied. Samples with nominal composition $Bi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3B_xO_y$, x=0 - 0.5, were prepared via a solid state reaction route. Superconducting properties of undoped (reference) and boron-added (Bi,Pb)-2223 compounds were investigated through X-ray diffraction (XRD), scanning electron microscopy (SEM), resistivity and transport critical current density (J_c) measurements. Obtained results have shown that boron additive leads to the acceleration of high-T_c phase formation and enhancement of J_c in (Bi,Pb)-2223 superconductor. The estimated volume fraction of (Bi,Pb)-2223 phase increases from ~25 % for reference specimen to ~75 % for x=0.15. Moreover, strong increase in the J_c was observed for the x=0.15 sample (J_c=340 A/cm^2), compared to a reference sample (J_c=115 A/cm^2). We have studied the effect of high-energy ball milling on J_c in reference and x=0.15 samples. Addition of B in combination with the ball milling leads to the further enhancement of J_c up to 490 A/cm^2, whereas the ball milling of reference specimen causes the marked decrease in both J_c and T_c values. Improvement of superconducting properties in (Bi,Pb)-2223 superconductor can be attributed to the acceleration of high-T_c phase formation along with the enhancement of intergrain coupling due to the elemental boron addition.

Keywords: (Bi,Pb)-2223 phase, Crystalline boron additive, Ball milling, Electrical resistivity, Critical temperature, Critical current density.

1. INTRODUCTION

Since the discovery of $Bi_2Sr_2Ca_2Cu_3O_y$ (Bi-2223) HTS, it has been considered to be one of the most promising materials for large scale applications in superconducting industry. However, due to the very slow formation kinetics of the Bi-2223 phase, hundreds of hours are necessary to prepare an appropriate fraction of Bi-2223 HTS in the final product [1, 2]. Partial substitution of Bi by Pb is the most widely used method to enhance the formation of the Bi-2223 phase [3], but after more than two decades, the formation of pure Bi-2223 phase, which possesses the best possible values of the critical current density (J_c), is still an open subject in the field of Bi-based superconductors [4]. Many factors, including the nominal composition of the precursor powder, thermal processing time and temperature, an element and particle size of dopants, milling treatment of precursor, texturing and synthesis atmosphere significantly influence the phase formation kinetics and J_c of Bi-2223 HTS [5]. Doping studies have demonstrated that substitution or addition of micro - and nanosized dopants is an efficient method to improve the intergrain connectivity and flux pinning capability in Bi-2223 [4, 6-15].

Our previous results show that the partial substitution of lead borate $Pb(BO_2)_2$ for lead oxide PbO as well as the addition of boron-containing compounds into the (Bi,Pb)-2223 HTS promotes the formation of high-T_c phase and leads to the enhancement of transport properties compared to the reference sample [16-18]. The present study was aimed to investigate the

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co-influence of the elemental boron addition and planetary ball milling processing of precursor powder on the formation and transport properties of the (Bi,Pb)-2223 HTS.

2. MATERIALS AND METHODS

Boron added (Bi,Pb)-2223 HTSs with the nominal compositions $Bi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3B_xO_y$, x=0, 0.1, 0.15, 0.3, 0.5 were prepared by the solid state reaction method. Appropriate amounts of highly pure Bi_2O_3, PbO, $SrCO_3$, $CaCO_3$, CuO and elemental B **(**purity of 95%**)** were mixed and sintered at 850 ^oC for 40 h in air with intermediate manual grindings using agate mortar and pestle. The resulting materials were pressed into pellets of 10 mm in diameter and 1.5 mm thickness under hydrostatic pressure of 29 MPa. The pellets were annealed at 840 ^oC for 30 h in air, then cooled to room temperature in the furnace. The synthesized compounds were characterized by powder XRD analysis using the Dron-3M diffractometer (CuK_α radiation). The resistivity as a function of temperature, $\rho(T)$, and transport critical current were measured by a standard four-probe method. We have tested the influence of dry ball milling processing of precursor powders (before being pressed into pellets) on the microstructure and transport properties of the reference (x=0) and boron added (x=0.15) samples. Partly reacted precursor powders (850 ^oC for 40 h) without and with boron additive (x=0.15) were ball milled in a planetary mill (Fritsch Pulverisette 7 Premium line) for 1 h at rotating speed of 200 rpm, in air. Eighty agate balls 5 mm in diameter were used as a grinding medium. After milling, the powders were pressed into pellets and annealed at 835 ^oC for 30 h in air. The surface morphology of the samples was examined using a SEM (VEGA TS5130MM). The dc current of 5 mA was used to determine the $\rho(T)$ dependence of the samples. The typical sample size for J_c measurements was ~9×0.5×0.5 mm^3. J_c values were measured at the liquid nitrogen temperature in the self field, with a criterion of 1 μV/cm.

3. RESULTS AND DISCUSSIONS

XRD patterns of the reference and boron added specimens synthesized from manually-ground precursors (denoted as as-prepared samples) are shown in Figure **1**. The dominance of the low-T_c 2212 phase over the high-T_c 2223 phase was observed in the reference sample. With the incorporation of boron, the 2223 phase is markedly enhances, whereas the 2212 phase fraction decreases. The trace amount of the very low-T_c 2201 phase appears at doping level of x=0.3 and intensifies with increasing a dopant content to x=0.5. Moreover, XRD pattern for x=0.5 shows a broad diffuse halo indicating the formation of amorphous phase for the high doping level. The volume fractions (V) of the (Bi,Pb)-2223 phase were estimated from XRD intensity ratios of the (Bi,Pb)-2223 and Bi-2212 phases using the following equation [19]: $V=\{ I_{H(00\underline{10})}/[I_{H(00\underline{10})} + I_{L(008)}]\} \times 100$ [%], where $I_{H(00\underline{10})}$ is the XRD peak intensity of (00$\underline{10}$) in the Bi-2223 phase and $I_{L(008)}$ is the peak intensity of (008) in the Bi-2212 phase. The calculated volume fraction of (Bi,Pb)-2223 phase increases from ~25 % for reference specimen to ~75 % for x=0.15 in a relatively short sintering time of 70 h, which indicates that boron additive accelerates the solid state reaction rate and hence the (Bi,Pb)-2223 formation. Figure **2** represents the temperature dependence of resistivity for the as-prepared (Bi,Pb)-2223 samples. The linear temperature dependence of the resistivity in the normal state region of $\rho(T)$ transitions observed for all the samples indicates that the current flows preferentially along the conducting CuO_2 planes (ab-planes) of the oriented grains in these compositions. Onset temperature of superconducting transition is near 112 K for the reference sample and remains unchanged with increasing the boron content. For the undoped specimen zero resistivity is reached at T_c^{off}=102.5 K. T_c^{off} decreases to 100.5 K with the addition of x=0.1 boron, and then increases slightly up to 101.5 K at x=0.15. A further addition of B (x>0.15) leads to gradual decrease of T_c^{off}, that can be attributed to the appearance and increase of the secondary 2201 phase and deterioration of crystallinity for x=0.5, in accordance with the XRD results. Figure **3** represents the XRD patterns of reference and boron-added samples, which were prepared using ball-milling treatment of powder: the partly reacted (850 ^oC for 40) precursor powders were ball-milled prior to pressing into pellets, then pelletized, and sintered at 835 ^oC for 30 h (denoted as ball-milled). XRD data shown in Figures **1** and **3** indicate that the ball-milling does not substantially affect the phase assemblage in the reference and boron-added (x=0.15) samples. Resistivity versus temperature curves for ball-milled reference and x=0.15 samples are presented in Figure **4**. The relatively higher T_c^{off}=102 K and lower normal state resistivity of ball-milled B-added sample with respect to the as prepared B-added sample, x=0.15, (Figure **2**) may be attributed to the improvement of intergrain connectivity. At the same time, ball-milling of reference sample leads to the decrease of T_c^{off} from 102.5 K to 98 K and increase in normal state resistivity. Therefore, unlike the boron added sample, ball-milling treatment degrades the connectivity between grains in the boron free (reference) sample. Figure **5** illustrates

the relationship between the transport J_c values (77 K, zero field) and the amount of boron additive for the as-prepared and ball-milled samples. The value of J_c is 115 A/cm^2 for as-prepared reference sample, which is consistent with the literature values for bulk, non-textured (Bi,Pb)-2223 superconductors [9-11, 20]. Significantly enhanced J_c values were observed for the as-prepared B-added samples (x=0.1 – 0.3), especially with x=0.15 boron additive (340 A/cm^2). This J_c value is about 3 times higher than for an as-prepared reference compound processed for the same total sintering time of 70 h. Enhancement of J_c seems to result from the increase of the (Bi,Pb)-2223 volume fraction and enhancement of the intergrain coupling in optimally doped sample. In agreement with the XRD and resistivity data shown in Figures **1** and **2**, decrease of J_c values for a higher level of B-doping (x≥0.3) imply the worsening of coupling at grain boundaries due to formation of 2201 phase and marked deterioration of the crystallinity for x=0.5. Figure **5** reveals that ball milling treatment leads to the further enhancement of J_c up to 490 A/cm^2 for the doped sample with x=0.15. Contrary to the B-added sample, ball-milling processing leads to the strong decrease of J_c in reference sample. Figure **6** illustrates the surface SEM micrographs of the as-prepared and ball-milled samples with x=0 and 0.15. Obviously, the boron-added samples show the slightly larger grain size than that of reference samples. Both of the improved morphology and accelerated formation of the 2223 phase lead to an enhancement of the current-carrying capacity in boron-added samples. The morphology of the as-prepared and ball-milled boron-added samples is almost identical, but ball milling treatment seems to produce a more homogeneous distribution of doped particles which results in an improved intergrain connectivity and better self-field J_c performance.

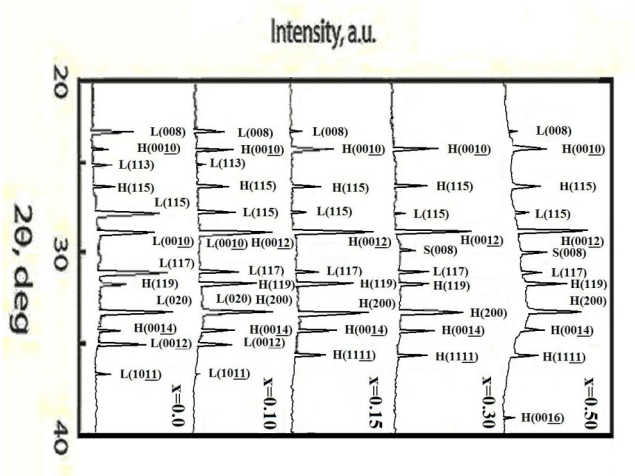

Figure 1: XRD patterns of as-prepared $Bi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3B_xO_y$ samples with various boron content. The diffraction peaks indexed H(hkl), L(hkl) and S(hkl) represent the Bi-based 222, 2212 and 2201 phases, respectively.

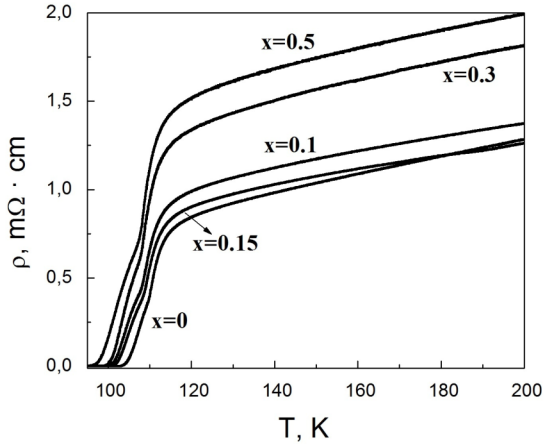

Figure 2: Resistivity versus temperature curves for as-prepared $Bi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3B_xO_y$ samples.

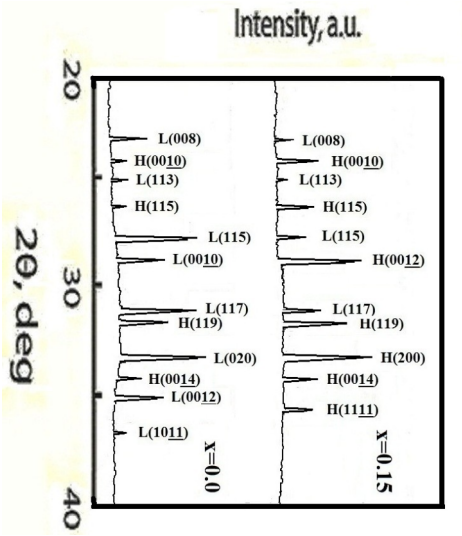

Figure 3: XRD patterns of ball-milled samples. The diffraction peaks indexed H(hkl), L(hkl) and S(hkl) represent the Bi-based 2223, 2212 and 2201 phases, respectively.

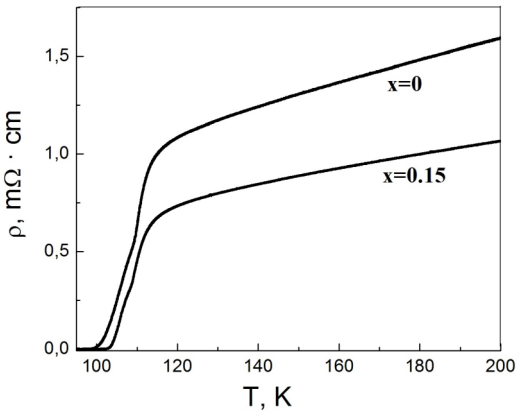

Figure 4: Resistivity versus temperature curves for ball-milled $Bi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3B_xO_y$ samples.

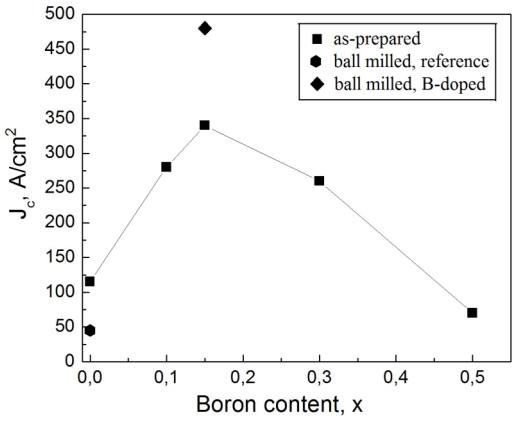

Figure 5: Relationship between the critical current density and boron content for as-prepared and ball-milled samples.

4. CONCLUSIONS

The effects of the boron addition and dry planetary ball milling on the phase formation, microstructure and transport properties of $Bi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3O_y$ HTS have been investigated. The incorporation of elemental boron into the (Bi,Pb)-2223 system accelerates the formation of high-T_c phase and leads to the significant increase of the current-carrying capability compared to the boron-free reference sample. It has been shown that the transport J_c of B-added (Bi,Pb)-2223 can be further enhanced by using a planetary ball milling treatment of precursor powder.

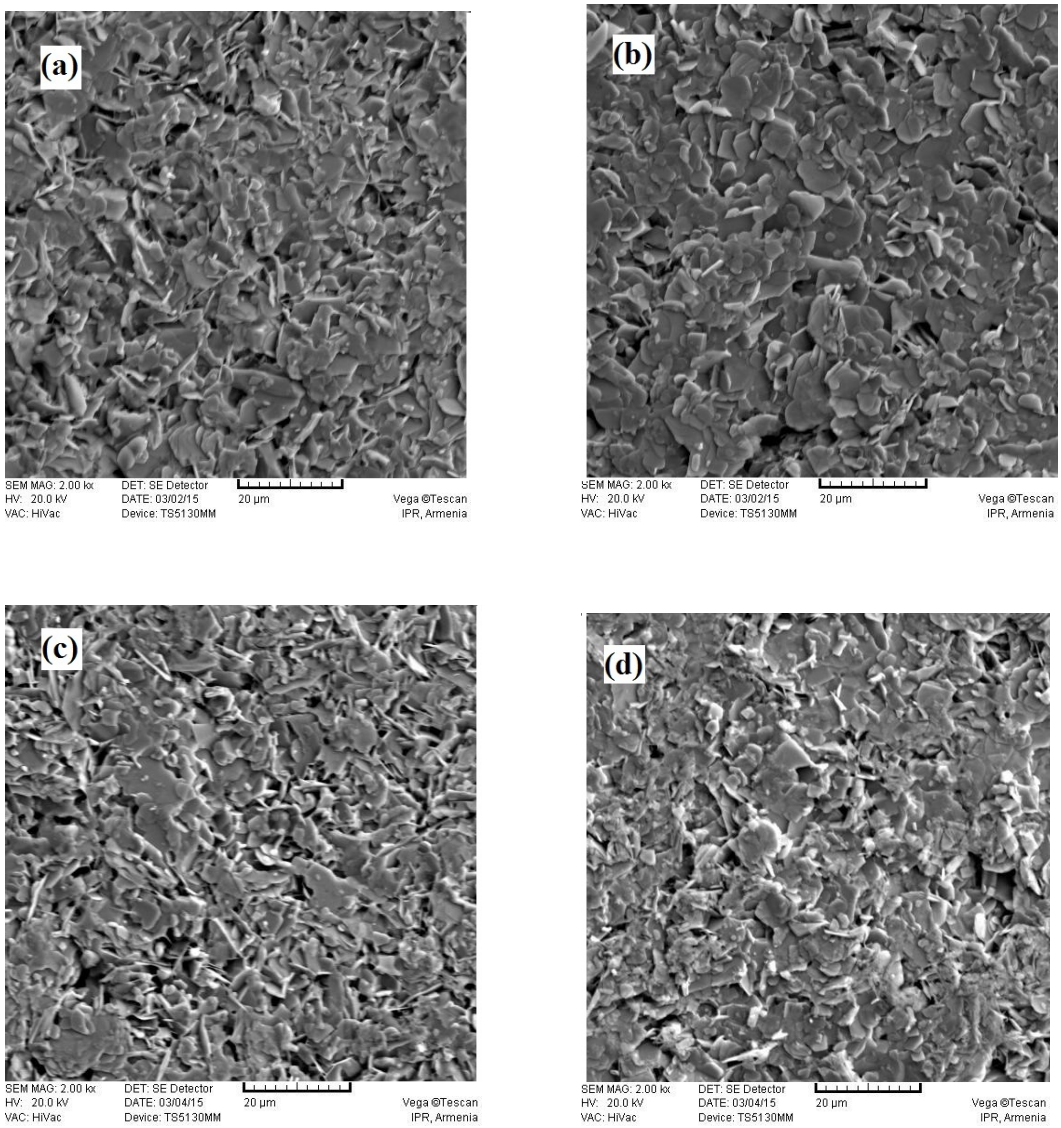

Figure 6: Surface SEM images of the as-prepared and ball-milled samples. (**a**): as-prepared (x=0), (**b**): as-prepared (x=0.15), (**c**): ball-milled (x=0), (**d**): ball-milled (x=0.15).

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