Effect of boron particle size on microstructure and superconducting properties of in-situ Cu addition MgB2 multifilamentary wire

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Abstract. In previous studies, the secondary (impurity and non-reactive) phase and voids were observed in MgB2 matrix after the heat treatment, and then these are the lowering factors of critical current density ($J_c$) property. In order to improve $J_c$ property by microstructure control of MgB2 matrix, the fine elemental boron powder as the raw material was carried out using the high-speed vibrated milling with tungsten carbide (WC) jar. The average particle size of metal boron powder was decreased from 1.14 μm to 0.20 μm by the high-speed vibrated milling. The various fine particle boron powders as the function of milling time were also prepared, and in-situ Cu addition MgB2 multifilamentary wires using these fine boron powders were fabricated. Critical transition temperature ($T_c$) value of Cu addition MgB2 wire using fine boron powder obtained to about 37 K. No change of the $T_c$ property by the different particle sized boron powders was confirmed. In this paper, the comparisons of microstructure and superconducting properties between the different boron particle sizes were investigated.

1. Introduction

Discovering of MgB2 superconducting compound in 2001 [1], many R&D activities of MgB2 tape and bulk materials were carried out for the practical applications at liquid hydrogen temperature. Because MgB2 has higher critical transition temperature ($T_c$) of 39 K compared with the other metallic superconducting compounds. Furthermore, the features of MgB2 compound are not only higher $T_c$ but also binary chemical composition, lower specific gravity and lower cost. These features will be contributed to the some future electrical power applications. In addition, it is well known that MgB2 compound is one of the lower induced radio-activation superconducting materials, so-called “Low activation superconductor”. Noda et al. reported that induced radio-activity of MgB2 compound remarkably lower than that of Nb-based superconducting materials, and the half-life period of MgB2 compound was estimated within 1 year based on International Thermonuclear Experimental Reactor (ITER) design [2]. MgB2 compound will be one of the attractive superconducting materials to use under the higher neutron irradiation and flux environment applications such as future fusion reactor and high energy accelerator device, and then it will be alternative material of Nb-Ti wire in these field

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applications. In previous study, remarkable critical current density ($J_c$) under the magnetic field at 4.2 K was improved by the Cu addition using Mg$_2$Cu compound and “Low temperature diffusion process” [3]. As the view point of advanced fusion application, microstructure and superconducting properties of Cu addition MgB$_2$/Ta multifilamentary wire using boron-11 isotope powder, which isotope was stable against neutron irradiation, for the restraint of the nuclear heating and transformation by the nuclide conversion were investigated [4]. $T_c$ value of MgB$_2$ wire using $^{11}$B powder was obtained to be 37 K, which was similar value compared with the MgB$_2$ wire using natural boron powder. However, we confirmed that $J_c$ value of MgB$_2$ wire using $^{11}$B isotope powder was lower than that of the sample via natural boron powder. In MgB$_2$ core on lower $J_c$ wire, the much increase of non-reactive boron particle fraction and decrease of current path were observed. A part of $J_c$-B enhancement on MgB$_2$ wire by the microstructure control, we performed the boron powder refining using high-speed vibrated milling in order to increase MgB$_2$ volume fraction with decreasing of non-reactive boron particles. The milling technique is one of the smart methods to improve the $J_c$-B performance due to the fine particle MgB$_2$ grain formation [5-7]. In this paper, the comparisons of microstructure and superconducting property on Cu addition MgB$_2$ wire between the different raw boron particle sizes were investigated.

2. Sample preparations and experimental procedure

Precursor mixture powders were made by metal Mg powder (99.9%), Mg$_2$Cu compound and natural boron powder. The Cu additional composition of precursor powder was fixed to the optimum 3 at% from the previous results of $J_c$-B performances [3, 4, 8, 9].

Boron powder refining was carried out using the high-speed vibrated milling. Boron powder and tungsten carbide (WC) ball were set into the WC jar. When boron powder was set into the WC jar, liquid nitrogen was also used in order to drive off the air gas. The milling time was 1, 3 and 5 minutes. The particle size of refined boron powder was measured using the laser diffraction typed particle size distribution meter (HORIBA LA-920), and average particle size was defined as 50 % of a passing portion cumulative distribution ($D_{50}$). W contamination and B oxidation were evaluated by the Inductively Coupled Plasma-mass spectrometry (ICP-mass) and X-ray diffraction (XRD) analysis.

The precursor mixture powders were packed into metal Ta tubes. Precursor mono-cored wires were fabricated through the PIT process. Wire drawing was carried out using grooved-roller and hexagonal cassette-roller dies, and the precursor mono-cored wires had a hexagonal diameter of 1.90 mm. The prepared mono-cored wire was cut to short piece wires, and 19 pieces of mono-cored wires were stacked into metal Cu tube. The stacked composite was wire drawn to a final diameter of 1.04 mm. Prepared precursor wires were heat treated using “Low temperature diffusion process” which is various lower temperatures (450-550 °C) during 200 hours under Ar atmosphere [8-10].

$T_c$ value was estimated from magnetization as a function of the temperature using a Quantum Design SQUID magnetometer. The magnetization hysteresis curves as a function of magnetic field under the various temperatures from 4.2 K to 30 K were also measured. The magnetic field from -5 T to +5 T applied to the wire samples. $J_c$ values estimated by the magnetization, so called “Magnetization $J_c$ ($J_{cm}$)”, of all samples were calculated by the “Bean-model” based on the critical state model [11]. The longitudinal microstructure was observed using SEM-EDX and TEM-EDX (Lorenz electron microscope (TECNAI-20F)).

3. Results and Discussions

3.1. Change of particle size distribution on the refined boron powder using high-speed vibrated milling

The particle size distributions of the boron powder after various milling time are shown in figure 1. The particle size distribution of boron powder without milling had two main peaks around 1.5 and 0.5 μm, and average particle size was obtained to 1.07 μm. In the case of the refined boron powder using vibrated milling during 1 minute, the particle size distribution was also shown two main peaks around 0.5 and 0.2 μm, and average particle size was obtained to 0.22 μm, which value was
corresponded to about 1/5 compared with boron powder without milling. These mentioned that the particle size of boron powder could be reduced by the high-speed vibrated milling. According to the XRD analysis of all refined boron powders, the diffraction peaks of B$_2$O$_3$ phase were not observed at all. The liquid N$_2$ into WC jar was effective to restrain the boron oxidation. Figure 2 shows that change of average particle size and W contamination as a function of the vibrated milling time. The average particle size was reduced remarkably during short time (1min). However, average particle size did not change on milling time (3 and 5 min). This suggested that boron refining did not depend on the milling time and shorter milling time promoted to refine boron powder due to the low temperature brittleness by liquid N$_2$. On the other hands, W contamination into refined boron powder was increased with increasing of vibrated milling time from ICP-mass results. From the TEM-EDS mapping, however, W concentration was not clearly observed in powder core filament matrix. The high-speed vibrated milling with liquid N$_2$ was preferable method to refine boron powder.

3.2. Effect of boron particle size on superconducting properties and microstructure of MgB$_2$ wires using atomized boron powder

Figure 3 shows that magnetization curves of MgB$_2$ wires using refined boron powder as a function of the vibrated milling time. These wires were sintered at 525 $^\circ$C for 200 hrs. The magnetic moment was normalized by the total volume of MgB$_2$ core filaments. $T_c$ values of MgB$_2$ wire using refined boron powder were almost obtained to 36.5 K and they were same to the MgB$_2$ wire using commercial boron powder. On the other hands, magnetization moment which was normalized by the total core volume was increased by the boron powder refining. We found that the volume fraction of MgB$_2$ phase was increased by the boron powder refining. This tendency was confirmed on each sintering temperatures (450-550 $^\circ$C), and the maximum magnetization moment was obtained to the sintering temperature of 525 $^\circ$C. Furthermore, we confirmed that magnetization property was improved by the refined boron powder and the optimum milling time, which was obtained to the maximum magnetization, was 3 min. Based on these M-H hysteresis curves, the comparisons of $J_{cm}(B)$ performances as a function of the temperature was shown in figure 4. In each temperature (5-30 K), the $J_{cm}$ properties of MgB$_2$ wire using refined boron powder were higher than that of commercial MgB$_2$ wire (not refined B). $H_{c2}$ values estimated by Kramer plot of refined boron wire were improved.
We thought that $J_{cm}$ and $H_{c2}$ properties enhancements were caused by the increase of volume fraction of MgB$_2$ phase due to the boron powder refining.

4. Conclusions

The high-speed vibrated milling with liquid N$_2$ was suitable to refine boron powder, and particle size was reduced to 1/5 during short time such as 1 minute. And liquid N$_2$ was also effective to restrain the boron oxidization and promote boron powder refining due to the low temperature brittleness. The $J_{cm}$ and $H_{c2}$ properties of MgB$_2$ wire were improved by the boron refining. These were caused by the increase of MgB$_2$ volume fraction.

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