Microstructure and superconducting properties of Cu addition MgB₂ multifilamentary wires using boron isotope powder as the boron source material


Abstract

The natural boron has two kinds of isotopes which are existed 20 wt% boron-10 (¹⁰B) and 80 wt% boron-11 (¹¹B). ¹¹B isotope is stable against the neutron irradiation without nuclear transformation. For an advanced fusion application, in-situ PIT process using ¹¹B isotope powder as the boron source material was desirable to enhance low induced radio-activity of MgB₂ superconducting wire. We tried to fabricate the in-situ Cu addition MgB₂/Ta/Cu multifilamentary wire using boron isotope powder as the boron source material. In this study, superconductivity and microstructure of MgB₂ wire using boron isotope powder were investigated. Tc value of MgB₂ wire using ¹¹B powder was shown to about 37 K by the low temperature diffusion reaction, and its value was higher than commercial natural powder. However, Jc value of MgB₂ wire using ¹¹B isotope powder was remarkably lower than that of commercial natural powder. This is caused by the forming of much impurity phase and decrease of MgB₂ volume fraction due to the remaining of much non-reactive ¹¹B isotope powder.

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Keywords: MgB₂; Cu addition; low activation; ¹¹B isotope; superconductivity

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1. Introduction

MgB$_2$ superconducting compound was discovered in 2001 [1]. The features of the MgB$_2$ compound are a higher critical temperature ($T_c$) of 39 K, simple binary chemical composition, lower specific gravity and relatively low cost material. For future applications, we investigated the possibility of applying MgB$_2$ superconducting wire in an advanced nuclear fusion power plant system. Because radio-activity property of MgB$_2$ compound is remarkably lower compared with Nb-based superconductors shown in fig.1. Fig.1 shows that simulation result for the decay time behavior of induced radioactivity of various superconducting wires in the toroidal field (TF) coils of an ITER design [2]. The superconducting magnets of an advanced fusion reactor will be fabricated by the Nb-based superconductors such as Nb-Ti, Nb$_3$Sn and Nb$_2$Al due to the excellent high magnetic field property. However, Nb-based superconductors require a longer cooling time than tens of thousand years, if the 10 mSv/h for the remote-handling recycling level is satisfied [2]. The activity that is maintained during a long time cooling is caused by the formation of long-lived nuclides such as $^{84}$Nb ($t_{1/2}=2.0\times10^4$ years). The longer cooling time would be given to the serious impact to the maintenance and radioactive waste treatment schedules. A dose rate of around 10 mSv/h–100 mSv/h is kept at the magnet near the vacuum wall for a long time. This means that thicker shielding is necessary in front of the magnet if Nb-based superconductors are used for the long operation. Therefore, all components of fusion reactor were suitable to compose with low activation materials and they were preferable to reduce various long-lived radio-nuclide elements such as Nb, Ni and Ag and so on wherever possible.

MgB$_2$ and V based superconductors were remarkably shorter decay time than Nb-based superconducting wires and their half-life are within 1 year shown in fig.1 [2]. We thought that MgB$_2$ compound will be desirable as one of the candidate materials of “low activation superconducting magnet” for a fusion reactor operated near D-T core plasma. The merits of applying to MgB$_2$ superconducting wire in an advanced nuclear fusion power plant system are lower induced radioactivity and higher efficiency of the cryogenic system due to the higher critical temperature ($T_c$) property. In the fusion reactor, the Poloidal field (PF) and feedback coils require a larger coil radius to correct the position of the plasma. These coils are operated near core D-T plasma, and the lowering of heat load by the nuclear heat generation will be contributed with the stable operation. Although there are many merits of MgB$_2$ wire for

![Fig.1 The decay time behavior of induced radioactivity of various superconducting wires in the TF coils of an ITER design [2]](image-url)
fusion application, present critical current density ($J_c$)-$B$ performance of MgB$_2$ wire is obviously lower than commercial Nb-Ti wire. We studied about the Cu addition using Mg$_2$Cu compound in the MgB$_2$ phase with the purpose of improving $J_c$ without lowering $T_c$ property, and $J_c$ property of MgB$_2$ wire via low temperature diffusion was drastically improved under the low magnetic field region [3-6].

MgB$_2$ wire was fabricated by the in-situ Powder In-Tube (PIT) process via diffusion reaction between magnesium (Mg) and boron (B). The boron mainly has two kinds of isotopes which are boron-10 ($^{10}$B) and boron-11 ($^{11}$B). The natural isotope abundance of boron is mainly 20 wt% of $^{10}$B and 80 wt% of $^{11}$B. The $^{10}$B pellets are used to neutron absorption material of nuclear fission reactor because it has large nuclear reaction cross-section. Generally, $^{10}$B isotope is transformed to Li and He by the neutron irradiation. $^{11}$B isotope is stable against the neutron irradiation without nuclear transformation. We thought that in-situ PIT process using $^{11}$B isotope powder as the boron source material was suitable to enhance radio-activity of the low activation MgB$_2$ superconducting wire for fusion application. In this study, we tried to fabricate in-situ PIT processed Cu addition MgB$_2$ multifilamentary wire using the $^{11}$B isotope powder as the boron source material. Superconductivity and microstructure of Cu addition MgB$_2$ wire were investigated.

2. Sample preparation and experiment procedure

Precursor mixture powders were made by metal Mg powder (99.9%), Mg$_2$Cu compound and $^{11}$B isotope powder (98% under @Cambridge Isotope Laboratories, Inc.). It was difficult to disperse the Mg$_2$Cu compound homogeneously into precursor powder due to hard and brittle material and large grain size, so that the Mg$_2$Cu compound was only crushed mechanically to fine powder using a ball-milling process for 3 hours. The Cu additional composition of precursor powder was adjusted to optimum 3 at%Cu from the previous results of $J_c$-$B$ performances [3-6]. Precursor mixture powder using low purity commercial boron powder (99%) was also made for comparisons. The precursor mixture powders were tightly packed into metal Ta tubes (purity of 99.99%) with 10 mm outer diameter and 6 mm inner diameter. At first, precursor mono-cored wires were fabricated through the PIT process. Wire drawing was carried out using grooved-roller and cassette-roller dies, and the precursor wires finally had a diameter of about 2.00 mm. The prepared mono-cored wire was cut to short piece wires, and they were

![Fig.2 Typical SEM image of cross-sectional area in Cu addition MgB$_2$/Ta/ Cu 19 filament wire using $^{11}$B isotope powder as the boron source material](image)
stacked into metal Cu tube (OD:14 mm and ID: 10 mm). The number of stacked mono-cored wires was 19 pieces. This stacked composite was wire drawn to a final diameter of 1.04 mm. Intermediate annealing was not required between wire draws. Typical SEM image of cross-sectional area in MgB$_2$/Ta/Cu filamentary wire via PIT process using $^{11}$B isotope powder is shown in fig.2. Prepared multifilamentary precursor wires were heat treated using Low temperature diffusion process which is various lower temperatures (450-550 °C) during 200 hours in an Ar atmosphere [5-6].

After heat treatment, $T_c$ value was estimated from magnetization measurement with a Quantum Design SQUID magnetometer. $T_c$ value was defined by the onset of the transition of magnetization. The transport critical current $I_c$ (4.2K) were measured using a DC four-probe method. Transport $I_c$ (4.2K) measurement was made under magnetic fields up to 10 T using a superconducting magnet. $J_c$ values of all samples were defined as the transport $I_c$ values divided by the cross-sectional area of the powder-filled core, and these $J_c$ values were the so-called “core $J_c$”. And then the microstructure of longitudinal direction on the wire sample was observed with a Scanning Electron Microscope and Energy Dispersive X-ray spectrometer (SEM-EDX).

3. Results and discussions

3.1. $T_c$ property of MgB$_2$/Ta/Cu multifilamentary wire using $^{11}$B isotope powder

Figs. 3(a) and (b) are summarized that $T_c$ property of Cu addition MgB$_2$ multifilamentary wire using $^{11}$B isotope powder as the boron source material. Typical magnetization (M)-temperature (T) curves by zero-field cooling method as a function of sintering temperature in MgB$_2$ wires using $^{11}$B isotope powder is shown in fig.3 (a). $T_c$ value was defined by the onset of the transition on M-T curve. $T_c$ value was obviously increased by the elevating of heat treatment temperature; 29.5 K, 36.9 K, 37.8 K, 38.0 K and 38.0 K for 450 °C, 475 °C, 500 °C, 525 °C and 550 °C, respectively. This was suggested that high $T_c$ MgB$_2$ phase via PIT method using $^{11}$B isotope powder promoted to form at the heat treatment above 500 °C. The comparisons of $T_c$ property as a function of sintering temperature between $^{11}$B isotope powder and low purity B powder is shown in fig.3 (b). In the case of low diffusion process, there is plateau region of optimum $T_c$ property above 500 °C in the Cu addition samples. $T_c$ property of $^{11}$B

![Fig.3 $T_c$ property of Cu addition MgB$_2$ multifilamentary wire using $^{11}$B isotope powder as the boron source material. Fig.3 a) is M-T curves of MgB$_2$ wires and b) is $T_c$ value as a function of heat treatment temperature.](image-url)
isotope sample was higher compared with each amount of Cu addition sample. On the other hand, D. K Finnemore et al, have reported that $T_c$ value of MgB$_2$ bulk sample made by $^{10}$B isotope powder was obtained to 40.2 K and its value was higher than that of commercial boron powder [7]. We found that MgB$_2$ phase formed by $^{11}$B isotope powder had higher $T_c$ property as well as $^{10}$B powder.

3.2. Transport current property of MgB$_2$/Ta/Cu multifilamentary wire using $^{11}$B isotope powder

Fig.4 shows that core $J_c$ property at 4.2 K as a function of sintering temperature in MgB$_2$ multifilamentary wires using $^{11}$B isotope powder. The core $J_c$ under magnetic fields of 1 and 2 T was increased by elevating sintered temperature. The highest core $J_c$ values were obtained to the 500 °C, and their value were estimated 1,700 (4.2 K@1 T) and 800 (4.2 K@2 T) A/mm$^2$, respectively. This suggested that optimum sintering condition was 500 °C for 200 hours in this study. Typical core $J_c$-B performance of MgB$_2$ multifilamentary wire using $^{11}$B isotope powder is also shown in fig.5. The $J_c$-B data of MgB$_2$ wire using commercial B powder was also plotted for comparisons. All samples were sintered at 500 °C for 200 hours. In the case of $T_c$ property, MgB$_2$ formed by $^{11}$B isotope powder has higher property compared with commercial B powder sample. However, core $J_c$-B property of $^{11}$B isotope powder was lower than that of commercial B powder. We thought that lowering of $J_c$-B property using $^{11}$B isotope powder was caused by the volume fraction decrease of MgB$_2$ phase into wire filaments.

3.3. Microstructure observation of longitudinal section in MgB$_2$ wires using $^{11}$B isotope powder

Fig.6 shows that typical SEM images of longitudinal section in MgB$_2$ wires using commercial B and $^{11}$B isotope powders. In the case of both commercial B and $^{11}$B isotope powder samples, MgB$_2$ phase was mainly formed. Furthermore, small particle sized MgCu$_2$ phase and non-reactive boron were also observed. MgCu$_2$ phase was formed by phase transformation from Mg$_2$Cu phase. However, obvious difference of non-reactive boron was confirmed. From the SEM image analysis, non-reactive boron area fraction of commercial boron sample was estimated to be about 7 %. In the case of $^{11}$B isotope sample, non-reactive boron area fraction was also estimated to be 19 % and it was about twice as much as commercial boron powder sample. We thought that increase of non-reactive boron fraction induced to
decrease of MgB₂ volume fraction and current path. It was proven that lowering of \( J_c \)-B property shown in fig.6 was caused by the MgB₂ volume fraction. These suggested that decrease of non-reactive boron volume fraction was required to improve \( J_c \) property of \(^{11}\)B isotope powder sample, and then fine grained \(^{11}\)B powder using ball-milling was one of the effective methods for further \( J_c \) enhancement.

4. Conclusions

MgB₂ multifilamentary wire using \(^{11}\)B isotope powder as boron source material was made. \( T_c \) value of the \(^{11}\)B isotope powder sample heated at 500 °C for 200 h was obtained to about 37 K and was higher than that of commercial B powder as boron source. \( J_c \)-B performance of MgB₂ wire formed by \(^{11}\)B isotope powder was decreased compared with that of commercial B powder. This will be caused by the decrease of MgB₂ volume fraction. From the SEM observation, we found that the non-reactive boron area fraction was estimated to be 19 % and it was also about twice as much as commercial boron powder sample.

Acknowledgements

This work was mainly supported by the NIFS program budget (NIFS11UFFF036), in part by the Grant-in-Aid for Young Scientists (B#21760240) from the Ministry of Education, Culture, Sports, Science and Technology (MEXT). We would like to thank for Tsukuba Magnet Laboratory of National Institute for Materials Science for providing with the chance to use high magnetic field magnet facilities for the \( I_c \) measurement. And also thanks for the Instrument Center of Institute for Molecular Science for proving with the use SQUID magnetometers for \( T_c \) measurements.

References