Ambient temperature dependence on superconducting properties of MgB\textsubscript{2} wires synthesized with low temperature diffusion process

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Abstract. In the previous study, we investigated the Cu addition using the Mg\textsubscript{2}Cu compound and the reconsidering of heat treatment through lower temperatures and longer time treatments, the so called “Low temperature diffusion process”. The critical current density (\(J_c\)) under the low magnetic field was enhanced by the “Low temperature diffusion process” compared to the conventional PIT process. In this study, transport critical current (\(I_c\))-temperature (\(T\))-magnetic field (\(B\)) measurements of the Cu added low temperature processed MgB\textsubscript{2} wires were carried out in order to investigate the possibility of high temperature operation. In the \(I_c-T-B\) measurement, the samples were cooled by helium (He) gas and its temperature was controlled by the output of the external heater. The transport \(I_c\) values of the mono-cored and 19 multifilamentary wires under the 20 K and 0 T were obtained to be above 100 A. Furthermore, the transport \(I_c\) degradation by the ambient temperature and bending strain was also investigated for a large scaled low temperature diffusion processed MgB\textsubscript{2} cable.

1. Introduction
The MgB\textsubscript{2} compound was discovered superconducting in 2001. The features of MgB\textsubscript{2} are a high \(T_c\) of 39 K, a simple binary chemical composition and relatively low material cost. Many R&D activities regarding MgB\textsubscript{2} tape and bulk materials were carried out for practical applications at liquid hydrogen temperature (20 K). Furthermore, it is well known from the simulation result that the radiation-induced property change in MgB\textsubscript{2} is lower than that of Nb-based superconductors such as Nb-Ti, Nb\textsubscript{3}Sn and Nb\textsubscript{3}Al [1]. The MgB\textsubscript{2} compound is one of the “Low activation superconductors” and will be attractive superconducting material to use under neutron irradiation environment applications such as fusion reactors and high energy accelerator devices. However, the \(J_c\) values of MgB\textsubscript{2} are poor in comparison with Nb-based superconductors. A new process to enhance the \(J_c\) property is required for the practical application.

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Recently, a newly Cu added powder in tube (PIT) process using Mg$_2$Cu as the Mg and Cu source materials during MgB$_2$ formation was developed [2-4]. We found that $J_c$ values under low magnetic field increased remarkably due to the small amounts of Cu addition using the Mg$_2$Cu compound. It is caused by the decomposition of Mg$_2$Cu; the reaction of Mg with B followed by the formation of high $T_c$ MgB$_2$ diffusion phase [4]. On the other hand, the metal sheath material of the PIT wire was also investigated in view of neutron irradiation environment applications [5]. In the metal sheathed materials, low activation, workability, mechanical strength, non-magnetization and non-corrosion properties are required. In these points, we thought that the Ta and Cu metals were suitable materials [5]. We also developed the new heat treatment process using low temperature and long time which was named “Low temperature diffusion” [6]. At the same time, a 100 m classed mono-cored Ta sheathed wire having a Cu stabilizer was successfully fabricated in order to investigate the workability of metal Ta sheathed sample [6]. The $J_c$ value of MgB$_2$/Ta/Cu mono-cored wire was significantly increased by the low temperature diffusion process and it was higher compared to commercial Nb-Ti wire at 4.2 K and magnetic field below 3 T, suggesting that MgB$_2$/Ta/Cu wires synthesized with low temperature diffusion process was able to be one an alternative superconducting materials with respect to Nb-Ti wire.

One of authors, S. Yamada, investigates a new scenario, which is a co-generation system of electricity and hydrogen fuel on the fusion power plant for the compatibility to the hydrogen society in future [7]. The Energy Supergrid, i.e. the transportation of electricity and hydrogen fuel through the energy pipeline, was proposed in USA [8]. At present, World Energy Transmission System in EU is also discussed to assess the global network of the superconducting power cable and fuel pipeline [9]. Based on this background, Hybrid Energy Transfer Line (HETL) consisting of a 10 kA class superconducting cable and spacing for liquid hydrogen was designed by the fusion technology [7]. The superconducting wire of HETL is operated in the temperature range of 17 K- 24 K, so that high $T_c$ oxide and MgB$_2$ will be influential candidate superconducting materials of the HETL. It is known that high $T_c$ oxide superconductors such as BSCCO and YBCO have excellent $J_c$ performance in liquid hydrogen temperature of 20 K. However, they are much more expensive and have weak mechanical properties compared with the MgB$_2$ wire.

In this study, the $I_c$-T-B performances of Cu added MgB$_2$/Ta/Cu mono-cored and multifilamentary wires were investigated in order to verify the possibility of the large superconducting cable such as the HETL application. In addition, $I_c$ degradations with applying bending strain of the sample wires were investigated.

### 2. Sample preparation and experimental procedure

#### 2.1. Fabrication of MgB$_2$/Ta/Cu mono-cored and multifilamentary wires

The Cu added MgB$_2$ precursor powder was made by metallic Mg (99.9%, -200μm) and amorphous B (99.9%, -submicron) powders. The Cu addition was carried out by using Mg$_2$Cu. The Mg$_2$Cu compound was only crushed mechanically into fine powder using a ball-milling process for 3 hours. The Cu addition was fixed to be 1.0 at% due to the results of $J_c$-B measurement at 4.2 K [4-5]. These source powders were mixed in the following ratio;

$$0.98 \text{ Mg} : 0.01\text{ Mg}_2\text{Cu} : 1.99\text{ B} \quad (1\text{at%Cu} \text{addition}).$$

The MgB$_2$/Ta/Cu mono-cored composite was made by the inserting a Ta tube (purity of 99.99%, OD:10 mm and ID:6 mm) into Cu tube (purity of 99.99%, OD:14 mm and ID:10 mm). Before the powder packing, this composite sheath was annealed at 900°C in Ar for the softening of the Ta sheath. The prepared powder mixture was tightly packed into the metal Ta/Cu composite sheath. This MgB$_2$/Ta/Cu composite was processed wire deformation using drawing dies with a 10 % reduction. The composite mono-cored MgB$_2$ wires finally had a diameter of 1.04 mm.
The MgB<sub>2</sub>/Ta/Cu 19 multifilamentary wire was also fabricated. At first, the wire drawing of mono-cored MgB<sub>2</sub>/Ta wire was carried out using grooved roller and cassette roller dies, and this wire finally had a diameter of 2.0 mm. A part of mono-cored MgB<sub>2</sub>/Ta wires was cut into short pieces, and 19 short pieces were stacked into a Cu tube (OD:14 mm and ID: 10 mm). This composite was drawn using grooved roller and drawing dies. The final diameter of the MgB<sub>2</sub>/Ta/Cu 19 multifilamentary wire is about 1.04 mm. These mono-cored and multifilamentary wires were heat treated by the low temperature diffusion process which was at 475°C for 200 hours in Ar atmosphere[6]. The typical photograph of the cross-sectional area on the MgB<sub>2</sub>/Ta/Cu 19 multifilamentary wire is shown in fig.1.

2.2. I<sub>c</sub>-T-B measurement

To evaluate the I<sub>c</sub>-T-B performances, the 15 Tesla split type superconducting magnet at Tsukuba Magnet Laboratory of National Institute for Materials Science (TML-NIMS) was used. The Variable Temperature Insert (VTI) system was inserted into the superconducting magnet bore. The schematic diagram of the superconducting magnet which was inserted into the VTI is shown in fig.2. In this system, the temperature of the sample was controlled by a flow of cold helium gas. The Liquid He is drawn from the cryostat of the superconducting magnet through the pickup line and a needle valve, and led down to the heat exchanger. At the heat exchanger, cold helium gas is heated to the setting VTI temperature using a non-inductive heater, and then flowed into the sample space. A small heater was preferably also mounted on the sample holder to give close temperature control and to optimize settling times. The detailed cold He gas temperature is mainly controlled by the liquid He flow rate according to the opening of the needle valve, the small heater of the VTI and the two cernox thermometers attached sample probe and VTI system. The thermometer attached to the sample probe was installed into the sample holder. In this study, sample temperature was defined as the indicated value of the cernox thermometer attached to the sample probe. The sample cooling gas was compulsorily exhausted from the vacuum line of the VTI by pumping.

Transport I<sub>c</sub>-B measurement as the function of the sample temperature was carried out using the DC four probe method (voltage tap distance: 10 mm). The I<sub>c</sub> criterion was defined to be 1 μV/cm and the core J<sub>c</sub> value was calculated as the I<sub>c</sub> divided by the MgB<sub>2</sub> core area. For the 4.2 K comparisons between He gas cooling and liquid cooling, I<sub>c</sub> measurement was also carried out into the liquid He in order to verify the soundness of the VTI system. The I<sub>c</sub> degradation by the bending strain of mono-cored and multifilamentary wires were also investigated. The bending test was performed by setting short samples on some aluminum bending holders with different curvatures at room temperature for a time of 60 seconds. The bending strain (ε) was defined by the following equation (I):

\[
\varepsilon = \frac{\Delta L}{L_0}
\]

Fig.1 Typical photograph of the cross-sectional area on the MgB<sub>2</sub>/Ta/Cu 19 multifilamentary wire.
\[ \varepsilon = \left( \frac{D}{2R} \right) \times 100 \] ---- (I)

where \( D \) is the overall diameter of the wire, and \( R \) is the bending radius of the bending holder. After the bending, the sample was removed from the holder, to measure the \( I_c \)-B property at 20 K.

3. Results and Discussions

3.1. Verification of the \( I_c \)-T-B measurement system

Fig.3 shows the comparisons of the transport \( I_c \) values between He gas cooling and liquid He cooling on the mono-cored wires. In this case, the sample temperature was set to 4.2 K. In the both cooling cases, we confirmed that the transport \( I_c \) showed mostly the same value under magnetic fields above 6 T, indicating that the \( I_c \)-B measurement using the He gas cooling was a suitable method. No observation of different cooling powers between gas and liquid cooling process were found in this study. In the lower magnetic field of 4 T, however, the difference in the values was clearly observed, suggesting that it was caused by the heat invasion from current lead and sample probe. In this study, the sample temperature was set to 4.2 K, 10 K, 15 K and 20 K. The current sweep was carried out after the sample temperature stabilization. For example, typical I-V curves as a function of the magnetic field of the mono-cored wire under the gas cooling of 15 K and the change of sample temperature during \( I_c \) measurement is shown in fig. 4. In the case of a setting temperature of 15 K on the VTI system, the sample temperature was 14.8 K. On the magnetic field of 4 T, a transport \( I_c \) value was defined as about 25 A and the sample temperature was kept at 14.8 K during superconducting state. In the case of magnetic field of 2 T, the transport \( I_c \) value was higher than that of 4 T and the sample temperature was elevated to 15.0 K at a transport current of 60 A in spite of the superconducting state, suggesting that the increase in the sample temperature was caused by the heat invasion from the current lead of the sample probe. We confirmed that the size of the sample temperature increase was
estimated to be about 0.2 K and that the sample temperature was close to the VTI setting temperature control in each measurement conditions. Thus, we found a highly certificated sample temperature using helium gas cooling by the linkage between heat exchanger with inductive heater and two cernox thermometers attached to the VTI and sample probe.

3.2. $I_c$-T-B properties of MgB$_2$/Ta/Cu mono-cored and multifilamentary wires

The typical $I_c$-T-B properties of Cu added MgB$_2$/Ta/Cu mono-cored and multifilamenatry wires are shown in figures 5 and 6. The $I_c$-B properties using liquid He cooling are also shown in figures 5 and 6 for the comparison. We observed that $I_c$-B was decreased with increasing sample temperature in the both cases. In the case of a sample temperature of 20 K, a transport $I_c$ value of 50 A was obtained in magnetic field above 1 T. This $I_c$ value corresponds to a core $J_c$ value of 300 A/mm$^2$, estimated from the MgB$_2$ core area for mono-cored wire. Similarly, the core $J_c$ value was about 1000 A/mm$^2$ in the case of the multifilamentary wire. These suggested that the multifilament configuration was effective to improve the core $J_c$ property. Furthermore, it was noticed that the core $J_c$ value at 4.2 K under 1 T of the Cu added MgB$_2$ multifilametary wires was about 5000 A/mm$^2$, and its value was almost equivalent to the conventional Nb–Ti wire, suggesting that Cu added MgB$_2$ wire are an alternative material to Nb–Ti wires for a low activation superconducting magnet system in the low magnetic field region of an advanced fusion application. From the results of the $I_c$-T-B measurement of the multifilamentary wire we found that the Cu added MgB$_2$/Ta/Cu wire was able to apply to the conductor of the HETL due to the satisfied core $J_c$ specification of the HETL [7]. We now approach the optimization of the wire configuration such as Ta barrier and Cu ratios for the further $J_c$ improvement of MgB$_2$/Ta/Cu wire.

It was found that $I_c$ degradation as the function of the magnetic field in the multifilamentary wire was lower than that of mono-cored wire. This is caused by the reduction of the MgB$_2$ core diameter. The relationship between critical magnetic field ($H_c$) and sample temperature in the mono-cored and multifilamentary wires was investigated. The $H_c$ value was estimated by the conventional Kramer formula. The $H_c$ value of the multifilamentary wire was higher than that of mono-cored wire, corresponding that it was 12.2 T. These suggested that multifilament was one of the effective methods to improve core $J_c$ property. We summarized that MgB$_2$/Ta/Cu wire synthesized with the low
temperature diffusion was one of the desirable materials for the large superconducting cable under the 20 K operation such as HETL conductor.

Fig. 5 Typical $I_c$-T-B properties of Cu addition MgB$_2$/Ta/Cu mono-cored wire.

Fig. 6 Typical $I_c$-T-B properties of Cu addition MgB$_2$/Ta/Cu multifilamentary wires.

Fig. 8 The $I_c$ degradations as a function of the applied bending strain at 20 K of the MgB$_2$/Ta/Cu mono-cored and multifilamentary wire.

3.3. $I_c$ degradation by bending strain in the MgB$_2$/Ta/Cu mono-cored and multifilamentary wires

It was clearly found that Cu added MgB$_2$/Ta/Cu wires synthesized with low temperature diffusion already had excellent $I_c$-T-B performance, suggesting that it is a suitable conductor material for the
HETL. The mechanical properties are also important factors for the practical and HETL applications. The bending test, which is one of the typical mechanical tests, was simply carried out using the bending holder. At the loading of the bending stress, the bending strain was introduced to the wire samples with fitting the curvature of bending holder for 60 seconds. The bending strain was estimated from the wire diameter and the curvature of the holder, and the bending strain was estimated as described in the equation (I).

As bended, the wire sample was removed from the holder without back bent straight. If the bending strain was within the elastic limit, wire sample was straight. The wire sample after the applied to bending stress was connected to sample probe of $I_c$-T-B measurement system. Fig.7 shows typical I-V curves obtained at 20 K under 2 T in the multifilamentary wire after the bending test. The $I_c$ value, bench mark of $I_c$ value in the case of non bending stress applying, was picked up from the figs.5 and 6. We confirmed that a $I_c$ degradation by the bending strain occurred in the mono-cored and multifilamentary wires. However, it was notice that transport current was confirmed in spite of the applying higher bending strain above 2.0 % in the both wires. It suggested that metal Ta matrix is one of the suitable high mechanical strength materials in the MgB$_2$ wire. The gradient of the transport $I_c$ after the applying bending strain was decreased with increasing the bending strain, suggesting that MgB$_2$ grain connectivity was weaken by the bending strain. The $I_c$ degradation of the mono-cored sample was also measured. The $I_c$ degradations as a function of the applied bending strain at 20 K under the various magnetic field was shown in fig.8. It was clearly found that the $I_c$ degradation of the multifilamentary wire was smaller than that of mono-cored wire, suggesting that multifilament are the most effective method to restrict the $I_c$ degradation. In order to restrict the $I_c$ degradation of 50 %, bending strain was admitted to 0.7 % on the mono-cored wire. In the case of the multifilamentary wire, the acceptability of the bending strain was obtained to be 1.8 %. Furthermore, the magnetic filed dependence of the $I_c$ degradation was also investigated. We confirmed that $I_c$ degradation did not change regardless of the applied magnetic field, suggesting that the $I_c$ degradation did not depend on the applied magnetic field.

4. Conclusions
$I_c$-T-B performances of Cu addition MgB$_2$/Ta/Cu mono-cored and multifilamentary wires were investigated in order to verify the possibility of the large superconducting cable for the 20 K operation such as HETL.

- The core $J_c$ value at 4.2 K under 1 T of the Cu addition MgB$_2$ multifilametary wires was 5000 A/mm$^2$, suggesting that Cu addition MgB$_2$ wire was able to be an alternative material for Nb–Ti wires.
- In the case of the sample temperature of 20 K, transport $I_c$ value of 50 A was obtained at magnetic field above 1 T. The Cu addition MgB$_2$/Ta/Cu wire was one of the desirable materials for the large superconducting cable under the 20 K operation such as HETL conductor.
- It was clearly found that the $I_c$ degradation of the multifilamentary wire was smaller than that of mono-cored wire, the acceptability of the bending strain was obtained to be 1.8 % in the $I_c$ degradation of 50 %.

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