SiC Addition for MgB2 Superconducting Wire by Suspension Spinning

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SiC Addition for MgB_2 Superconducting Wire by Suspension Spinning

Tomoko Goto, Kazuo Watanabe, and Gen Nishijima

***Abstract*—The effect of nanoscale SiC addition on the field dependence of J_c for MgB_2 superconducting wire by suspension spinning was examined to enhance the flux pinning. The suspension spinning of commercially available MgB_2 and SiC powders was examined to fabricate a long superconducting MgB_2 wire by using the spinning medium of poly(vinyl alcohol) (PVA) or polyacrylonitrile (PAN). The as-drawn filaments were uniaxially pressed under 20 MPa at 200°C for 8 h to remove volatile components and connect the MgB_2 grains. The filamentary samples were enveloped by an iron sheet with a pellet of mixed powder of Mg and B to prevent Mg loss, and vacuum-sealed in a fused quartz tube and sintered. The field dependence of J_c for the samples was examined at 4.2 K in magnetic fields up to 14 T. The J_c of the sample was strongly dependent on the spinning medium and sintering condition. Addition of 5 at% SiC in the sample spun by PVA medium attained the maximum J_c value more than 1000 A/cm^2 at 4.2 K by applying the field of 14 T.**

***Index Terms*—Critical current, magnesium diboride, superconducting filaments and wires, suspension spinning.**

I. Introduction

SINCE the discovery of superconductivity in MgB_2 at 39 K, many studies have been made for the fabrication of MgB_2 tapes and wires. Most of the MgB_2 wires are now fabricated by the so-called powder-in tube (PIT) method [1]–[3]. However, the critical current densities of MgB_2 bulk materials are relatively low as compared with conventional A15 compound superconductors. It was reported that doped SiC nanoparticle and reacting at 950°C significantly enhanced the J_c at high magnetic fields [4].

We have developed a fabrication of MgB_2 superconducting filaments by a suspension spinning method to provide a simple manufacturing process for wire making with good process-ability [5]–[7]. Chemical doping in the filamentary MgB_2 superconductors was examined to enhance the flux pinning. Addition of 5 at% SiC in the sample pyrolyzed at 500°C for 30 min and vacuum-sintered at 885°C for 2 h improved the J_c at magnetic fields more than 7 T at 4.2 K and the upper critical field was 14 T [8]. The fabrication of MgB_2 wire by suspension spinning without vacuum sintering was also investigated. The filaments uniaxially pressed under 20 MPa at 200°C for 8 h showed superconductivity above 30 K [9]. In this paper, we examined the spinning and sintering conditions for the SiC added MgB_2 superconducting filaments by suspension spinning to enhance the J_c value.

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II. Experimental

The commercially available MgB_2 powders (98% Alfer Aesar) and nanoscale SiC powders (mixed phase of α and β, 400 nm in diameter) were passed through 350 sieves. The mixed powders with nominal composition of $(MgB_2)_{0.95}(SiC)_{0.05}$ were suspended in poly(vinyl alcohol) (PVA) solution of dimethyl sulfoxide and hexamethylphosphoric triamide (sample 1). The viscous suspension solution was extruded as a filament into a precipitating medium of methyl alcohol and coiled on a winding drum. Poly acrylonitrile (PAN) solution of N, N-dimethylformamide was also used for spinning medium (sample 2). The as-drawn filaments were uniaxially pressed under 20 MPa at 200°C for 8 h to remove volatile components and connect the grains. The filamentary samples with a pellet of mixed Mg and B powders were enveloped by an iron sheet to prevent Mg loss. The samples were sealed in an evacuated quartz tube and heated. Although the as-drawn filaments were round wire, the pressed sample became to be tape-like wire.

The electrical resistivity of the filamentary sample was measured by a standard four-probe technique. Silver pads were formed on the samples by Ag sputter deposition. Silver paint was used to connect the pads of the samples with Ag wires 100 μm in diameter. The sample was cemented to a substrate using epoxy resin (GM-6600 Genus) and set on a measuring holder. External magnetic fields were applied in a direction normal to the filament length using a helium free 15 T superconducting magnet at the High Field Laboratory for Superconducting Materials, Tohoku University. Currents were passed along the direction of the fiber axis and normal to the applied magnetic field. The J_c was defined by the offset method from the point on the I-V curve at which the voltage of 1 μV appeared between voltage terminals separated 2 mm.

The magnetization measurement was carried out in a commercial SQUID magnetometer (Quantum Design MPSM-5). External magnetic fields were applied in a direction normal to the sample length. For the calculation of the magnetic J_c, the expression $J_c\,(H) = 20\,\Delta M\,(b - b^2/3l)$ for a plate in a perpendicular field derived from the modified Beans model was used, where ΔM is the difference of magnetization (emu/cm^3) measured for ascending and descending applied field, b and l are the sample thickness (cm) and length (cm), respectively $(b \ll l)$.

The crystal structure of the samples was studied by X-ray diffractometer by powder method.

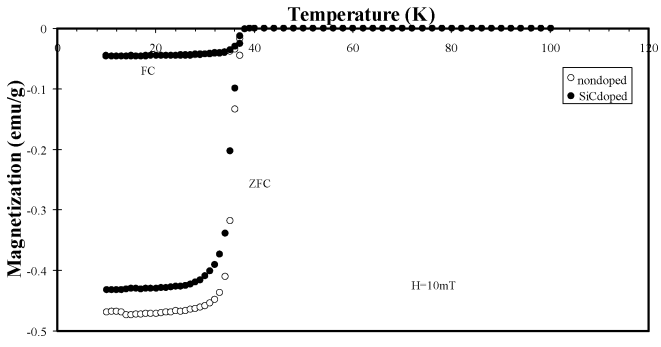

Fig. 1. Magnetization as a function of temperature for the sample 1 uniaxially pressed under 20 MPa at 200°C for 8 h and then vacuum-sintered at 885°C for 2 h.

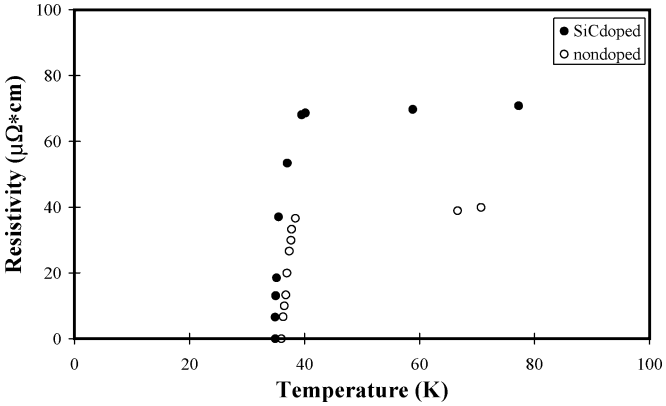

Fig. 2. Resistivity as a function of temperature for the sample 1 uniaxially pressed under 20 MPa at 200°C for 8 h and then vacuum-sintered at 900°C for 3 h.

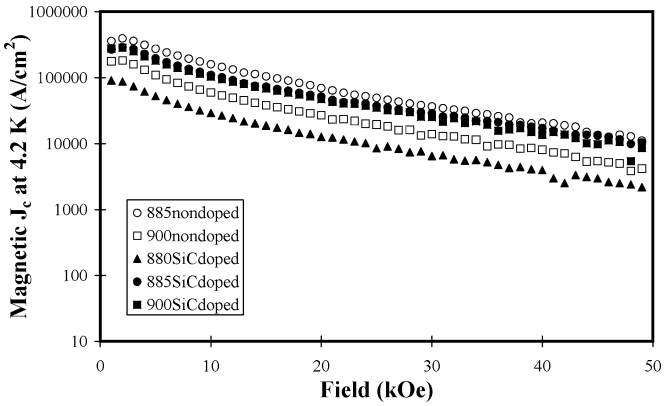

Fig. 3. The field dependence of magnetic J_c at 4.2 K for the sample 1 uniaxially pressed under 20 MPa at 200°C for 8 h and then vacuum-sintered at various temperatures.

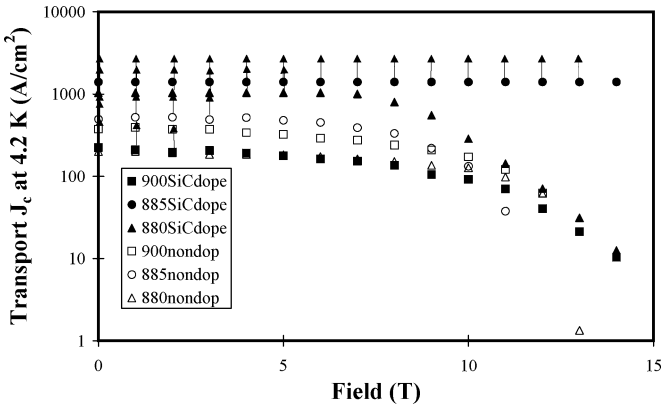

Fig. 4. The field dependence of transport J_c at 4.2 K for the sample 1 uniaxially pressed under 20 MPa at 200°C for 8 h and then vacuum-sintered at various temperatures.

III. Results and Discussion

Sample 1 was spun through PVA spinning medium. The as-drawn filament was uniaxially pressed under 20 MPa at 200°C for 8 h and then vacuum-sintered at 880°C–900°C for 2–3 h. Fig. 1 presents the temperature dependence of the susceptibility for the SiC doped and nondoped samples vacuum-sintered at 885°C for 2 h. Both samples show the T_c of 39 K. Fig. 2 shows the temperature dependence of the resistivity for the SiC doped and nondoped samples. The zero resistivity temperature ($T_c = 35$ K) is slightly low for the doped sample as compared with the nondoped sample with $T_c = 36$ K and higher normal resistivity value of 70 $\mu\Omega \cdot$ cm at 42.0 K was observed for the doped sample. However, the normal resistivity value for the doped sample uniaxially pressed under 20 MPa at 200°C for 8 h was 2 times lower than that for the samples pyrolyzed at 500°C for 30 min and vacuum-sintered at 900°C for 3 h [8].

The field dependence of magnetic J_c for sample 1 uniaxially pressed under 20 MPa at 200°C for 8 h and vacuum-sintered at various temperatures was measured in the applied field up to 5 T and the results for the doped and nondoped samples are shown in Fig. 3. The J_c values for the doped sample sintered at 885°C for 2 h are slightly low as compared with the nondoped sample by applying the fields less than 50 kOe. However, high J_c value of more than 10^4 A/cm^2 was observed for the doped and nondoped samples uniaxially pressed under 20 MPa at 200°C for 8 h and sintered at 885°C for 2 h by applying the field of 49 kOe. The J_c value for the doped sample uniaxially pressed under 20 MPa at 200°C for 8 h was about 4 times higher than that for the doped samples pyrolyzed at 500°C for 30 min (2300 A/cm^2 at 4.2 K by applying the field of 49 kOe) [8].

The field dependence of transport J_c for the samples was examined. As applying the current more than 1 A sometimes burned out the samples, the correct J_c value was not measured in a low field region. The field dependence of transport J_c for sample 1 is shown in Fig. 4. Doping of SiC enhanced the J_c value at high fields. The doped sample 1 uniaxially pressed under 20 MPa at 200°C for 8 h and vacuum-sintered at 885°C for 2 h shows the highest J_c value at high magnetic fields. High J_c value of more than 1000 A/cm^2 is maintained by applying the field of 14 T at 4.2 K. Only the maximum J_c value of 14 A/cm^2 at 4.2 K and 14 T was attained for the SiC doped sample pyrolyzed at 500°C for 30 min and vacuum-sintered at 900°C for 3 h [8]. Thus the significant improvement of the J_c is observed by uniaxial pressing under 20 MPa at 200°C for 8 h. This is considered due to the improvement of densification of the grains by uniaxial pressing.

S. X. Dou *et al.* have successfully introduced pinning site directly into the crystal lattice of MgB_2 grains by SiC nano-particles. They found a very high density of dislocations and massive nano-meter size inclusions inside the grains. These inclusions

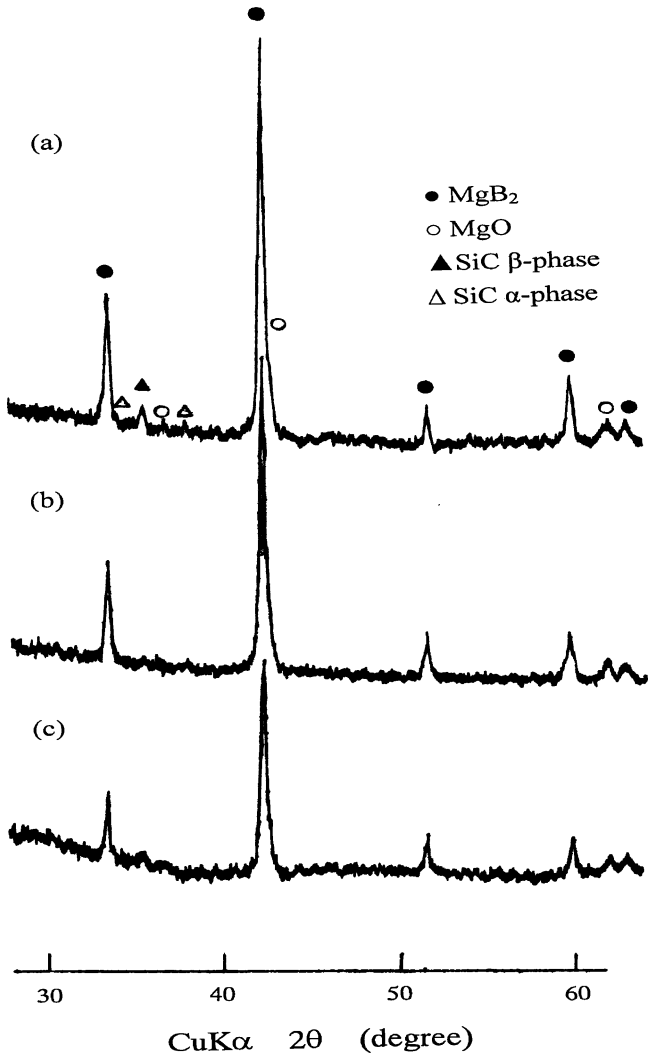

Fig. 5. X-ray diffraction pattern of the SiC doped sample 1 uniaxially pressed under 20 MPa at 200°C for 8 h and sintered at various vacuum-sintering conditions. (a) 880°C for 2 h, (b) 885°C for 2 h, (c) 900°C for 3 h.

were most likely the Mg_2Si as the major impurity phase picked up by the X-ray diffraction pattern [4].

X-ray diffraction patterns of the 5 at% SiC doped sample 1 uniaxially pressed under 20 MPa at 200°C for 8 h and sintered at various temperatures are shown in Fig. 5. The structure of the sample sintered at 880°C for 2 h is mixed phase of MgB_2, MgO and SiC. The SiC phase disappeared by sintering at 885°C for 2 h. Weak broad peak at $2\theta = 35$ degree was slightly detected in the X-ray diffraction pattern of the sample sintered at 900°C for 3 h. On the other hand, the structure of 5 at% SiC doped sample pyrolyzed at 500°C for 30 min and sintered at 900°C for 3 h was a mixture of MgB_2, MgO, Mg_2Si and MgB_4 phases [8]. This is considered that SiC reacted with MgB_2 and turned to fine inclusions without clear crystal structure by sintering at 885°C for 2 h for the sample uniaxially pressed under 20 MPa at 200°C for 8 h. These fine inclusions acted as effective pinning center, responsible for the enhanced flux pinning.

The doped sample uniaxially pressed under 20 MPa at 200°C for 8 h and sintered at 885°C for 2 h consisted of relatively rough structure with grain size of 100–500 nm as shown in Fig. 6.

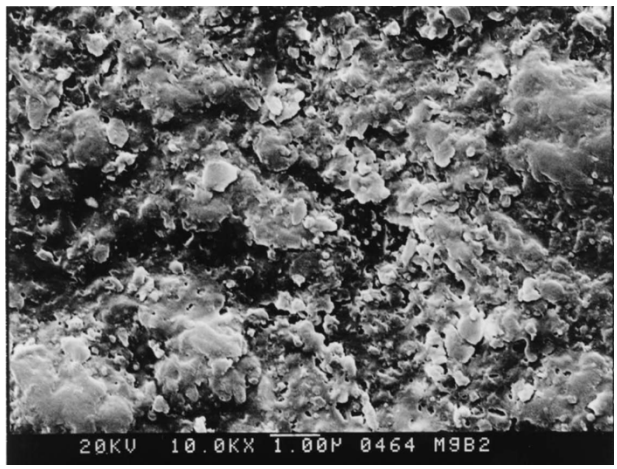

Fig. 6. Scanning electron microscopy image of cross-section of the SiC doped sample 1 uniaxially pressed under 20 MPa at 200°C for 8 h and then vacuum-sintered at 885°C for 2 h.

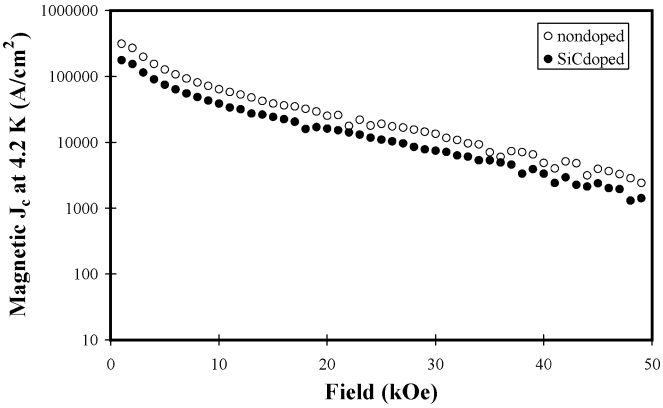

Fig. 7. The field dependence of magnetic J_c at 4.2 K for the sample 2 uniaxially pressed under 20 MPa at 200°C for 8 h and then vacuum-sintered at 880°C for 2 h.

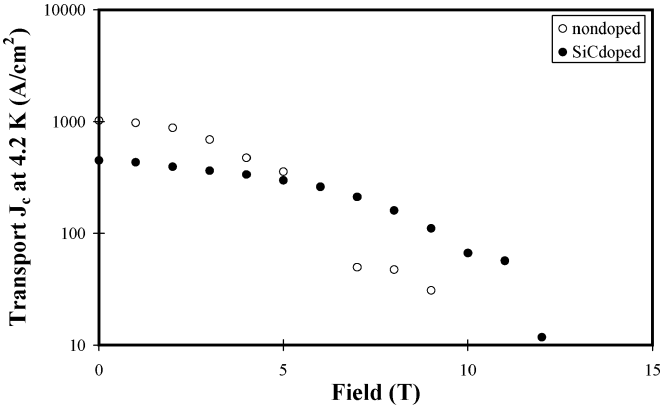

Fig. 8. The field dependence of transport J_c at 4.2 K for the sample 2 uniaxially pressed under 20 MPa at 200°C for 8 h and then vacuum-sintered at 880°C for 2 h.

Densely stacked MgB_2 grains with grain size of 50–100 nm were observed in the cross-section of the nondoped sample.

The suspension spinning of MgB_2 through PAN was also made (sample 2). The optimum sintering condition was at 860–880°C for 2 h for sample 2. The normal resistivity value at room temperature for sample 2 was two orders of magnitude higher than that for sample 1. Figs. 7 and 8 show the field

dependence of magnetic J_c and transport J_c for the doped and undoped sample 2, respectively. A magnetic J_c value of more than 10^3 A/cm^2 is observed for the doped and nondoped samples by applying the field of 49 kOe. Although the transport J_c value for the nondoped sample is higher than that for the doped sample by applying the fields up to 6 T, the upper critical field improved from 9 T to 12 T by SiC doping. However, both J_c values for sample 2 are lower than that for sample 1. It is considered that PVA spinning medium resulted in the higher densification of MgB_2 grains in the precursor filament than that for PAN spinning medium. This shows the densification of the MgB_2 grains is also dependant on the spinning conditions.

IV. Conclusion

We examined the effect of nanoscale SiC additions on the field dependence of transport and magnetic J_c for 5 at% SiC doped MgB_2 filamentary sample. The sample was fabricated by suspension spinning through PVA and PAN spinning medium. The precursor filament was uniaxially pressed under 20 MPa at 200 °C for 8 h and vacuum-sealed in a fused quartz tube with a pellet of mixed Mg and B powders to prevent Mg loss and sintered. The J_c of the sample was strongly dependent on the spinning medium and sintering condition. Addition of SiC for the sample spun by PVA medium enhanced the pinning force and attained the maximum transport J_c value of more than 1000 A/cm^2 at 4.2 K and 14 T.

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