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Fabrication and Superconducting Properties of Suspension-spun MgB_2 Filament

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We have developed fabrication of MgB_2 wires by a suspension spinning method and vacuum sintering to provide a simple manufacturing process for wire making with good process-ability. Optimizing the sintering process attained the maximum transport J_c value of more than 10^4 A/cm^2 at 4.2 K and self-field. Because of its easiness of scale up production, the fabrication of MgB_2 wire by the suspension spinning without vacuum sintering was investigated. The as-drawn filaments were hot-pressed under 20 MPa at 200°C for 8 h. The filaments showed superconductivity above 30 K. Indium and SiC additions resulted in an enhancement of the J_c.

Key words: MgB_2 wire, suspension spinning, without vacuum-sintering, critical current density, In and SiC additions

1. INTRODUCTION

Since the discovery of superconductivity in MgB_2 at 39 K, many studies have already been reported on the fabrication of MgB_2 tapes and wires. Most of the MgB_2 tapes and wires are now fabricated by the so-called powder-in-tube (PIT) method. Especially, a high critical current density (J_c) of 10^5 A/cm^2 at 4.2 K and self-field was reported for a Ni-tube without any heat treatment [1]. This process is very attractive from the viewpoint practical application because the fabrication process with no heat treatment leads to a large reduction in fabrication costs for the wires and tapes.

We have developed fabrication of MgB_2 wires by a suspension spinning method and vacuum sintering to provide a simple manufacturing process for wire making with good process-ability. The suspension spinning of commercially available MgB_2 powder was examined to fabricate a long superconducting MgB_2 wire. The as-drawn filaments were pressed and pyrolyzed to remove volatile components. The filaments were then cold-pressed, enveloped by an iron sheet with a pellet of mixed powder of Mg and B, and vacuum-sealed in a fused quartz tube and sintered. Optimizing the sintering process attained the maximum transport J_c value of more than 10^4 A/cm^2 at 4.2 K and self-field [2]. The improvement of J_c at applied magnetic field more than 7 T was detected by doping of 5 at% SiC and the superconductivity at 4.2 K was maintained by applying the field of 14 T [3].

Because of its easiness of scale up production, the fabrication of MgB_2 wire by the suspension spinning without vacuum sintering was investigated in this paper. The effect of In and SiC addition was also examined.

2. EXPERIMENTAL

The suspension spinning of MaB_2 powder was examined to fabricate a long superconducting MgB_2 wire. The commercially available MgB_2 powders (Alfar Aesar) were suspended in a mixed poly(vinyl alcohol) solution of dimethylsulfoxide and hexamethylphosphiric triamide. The viscous suspension solution was extruded as a filament into a precipitating medium of methyl alcohol and coiled on a drum. The as-drawn filaments were hot-pressed under 20 MPa at 200°C for 8 h in air to remove volatile components and to connect the grains.

The electrical resistivity of the filamentary sample was measured by a standard four-probe technique. Silver paint was used to connect silver-sputtered parts of the filament with Ag wires 100 μm in diameter. The sample was embedded in a substrate using epoxy resin (GM-6000 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 filament length 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 2mm.

The magnetization measurements were carried out in a commercial SQUID magnetometer. For the calculation of J_c, the expression J_c (H) = 20 $\Delta M/(b-b^2/3l)$ for a plate in a perpendicular field derived from the standard Bean model was used, where ΔM is the difference of magnetization (in emu/cm^3) measured for ascending and descending applied field, b and l are the sample width and length in cm, respectively.

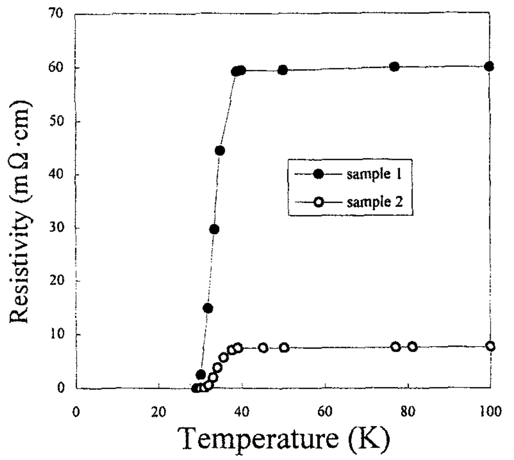


Fig.1. Resistivity as a function of temperature for sample 1 and 2.

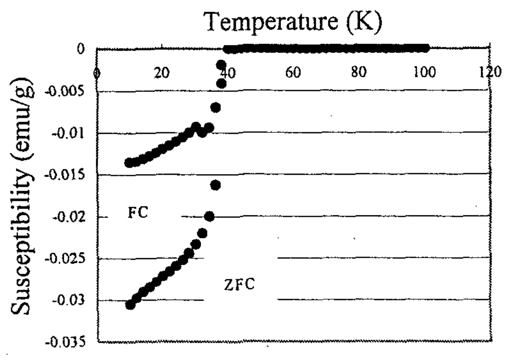


Fig.2 Susceptibility as a function of temperature for sample 2

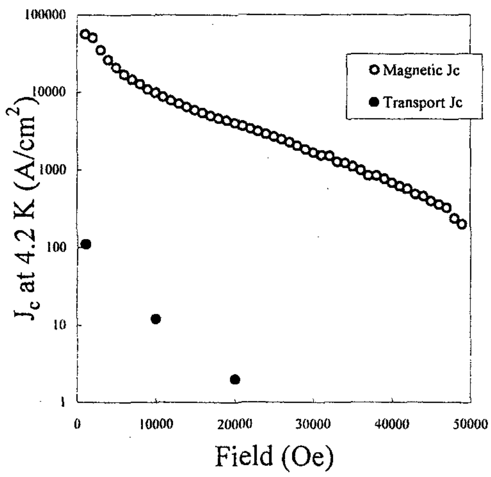


Fig.3 The field dependence of J_c at 4.2 K for sample 2.

3. RESULTS AND DISCUSSION

The as-drawn filaments were uniaxially pressed under 20 MPa at 200°C for 8 h in air to remove volatile components and to connect the grains. The pressed sample (sample 1) was apparently packed densely and packing density of the MgB_2 powders of about 1.4 g/cm^3 was reached in this way. The X-ray diffraction pattern of sample 1 showed the same profile of starting powder and consisted of MgB_2 and slightly MgO phases.

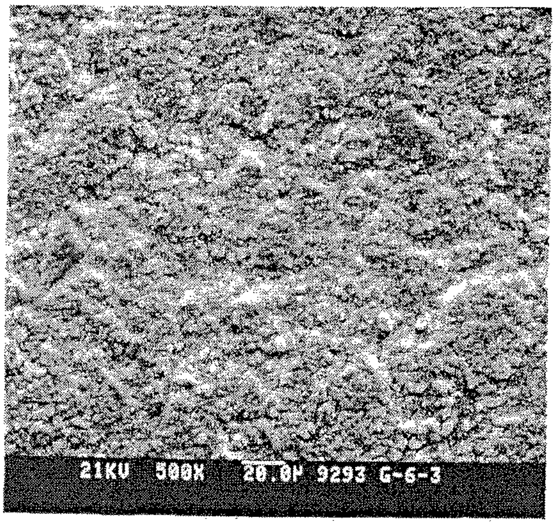
Fig.1 shows the temperature dependence of the electrical resistivity (ρ) for sample 1. The resistivity value abruptly decreases by decreasing the temperature less than 39 K. Sample 1 exhibits superconductivity at zero resistivity temperature of 29 K. The normal-state ρ value is about 60 mΩ·cm at 40 K and 500 times higher than that for the vacuum-sintered filamentary sample of 130 $\mu\Omega$·cm [2]. The transport J_c value at 4.2 K and self-field was low as 20 A/cm^2.

K.Tachikawa et.al found the enhancement of the transport J_c value by Indium addition in MgB_2 tapes due to the improvement of the linkage between MgB_2 grains [4].

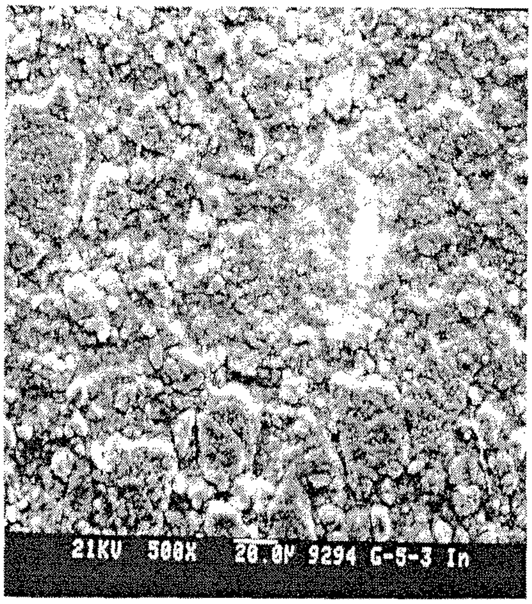
The addition of Indium in the present wire was examined to enhance the connectivity of the MgB_2 grains. The suspension spinning of the mixed powder MgB_2 and In of 0.5 at%, 5 at% and 10 at% were made. The as-drawn filaments were uniaxially pressed under 20 MPa at 200°C for 8 h in air to remove volatile components and to connect the grains. The temperature dependence of the resistivity for the pressed filamentary sample of MgB_2 with 0.5 at% In (sample 2) is shown in Fig.1. Sample 2 exhibits superconductivity at zero resistivity temperature of 31 K. The ρ value at 40 K is 7.5 mΩ·cm and one order of magnitude lower than that for sample 1. The transport J_c value at 4.2 K and self-field for sample 2 was 120 A/cm^2. SQUID measurement of sample 2 shows the sample exhibits superconductivity with diamagnetic response due to Meissner effect below 39 K as shown in Fig. 2. The effect of In contents on the ρ and J_c values at 4.2 K and self field was also examined. However, the improvement of the ρ and J_c values was not observed by In addition of more than 0.5 at %. This is due to the rough structure of MgB_2 grains by In addition.

The field dependence of the J_c at 4.2 K for sample 2 was measured by electrical resistivity and magnetization measurements and the results are shown in Fig.3. The transport J_c of sample 2 was disappeared by applying the field more than 3 T. On the other hand, the magnetic J_c value at 4.2 K was 5.7 x10^4 A/cm^2 and 2 x10^2 A/cm^2 at 0.1 T and 4.9 T, respectively. This attributed that the poor connectivity of MgB_2 grains is dominant for the present sample.

The filamentary samples were characterized by scanning electron microscopy and the SEM image of the cross-section of samples is shown in Fig.4. Fine grains packed densely in sample 1, while sample 2 consists of relatively rough structure. The Indium content in sample 2 was examined by EDX analysis. The In element distributed homogeneously in the cross-section of the sample. So far the J_c value of the wire is low and the



Sample 1



Sample 2

Fig.4 Scanning electron microscopy images of the cross-section of sample 1 and 2.

improvement of the connectivity for MgB_2 grains is needed.

Recently, it is reported significantly enhancement of the magnetic J_c and flux pinning of MgB_2 system superconductors doped with nano particle SiC by *in-situ* reaction method at high temperature of 950 °C [5]. In order to sinter the present filamentary sample at higher temperature, we examined the vacuum sintering with a pellet of Mg and B powder [3]. The suspension spinning of the mixed MgB_2 and 5at% nanoscale SiC powders (400nm) was made. The as-drawn filament was pressed and pyrolyzed at 500 °C for 30 min. The samples were then cold-pressed, enveloped by an iron sheet with a pellet of Mg and B powders, and vacuum-sealed in a fused

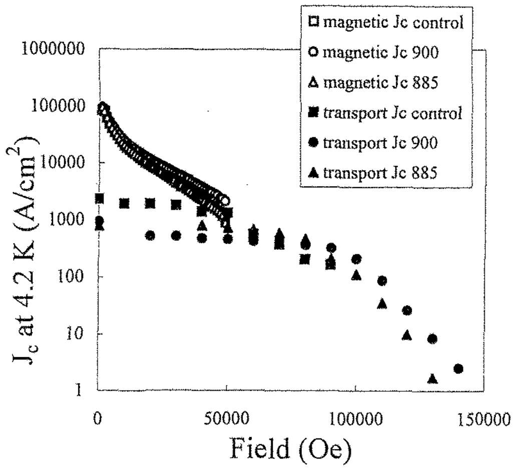


Fig.5 The field dependence of transport J_c and magnetic J_c for the SiC doped sample vacuum sintered at 885°C and 900°C.

quartz tube and sintered at 885°C for 2 h and 900°C for 3 h. The field dependence of J_c for the sample doped with SiC was dependent on the sintering condition. Figure 5 shows the field dependence of transport J_c and magnetic J_c for sample doped with 5 at% SiC and sintered at 885°C for 2 h and 900°C for 3 h respectively. The J_c value for the undoped sample (control) was also shown. While the enhancement of the transport J_c at the field ranging from 6 T to 8 T is observed for the sample sintered at 885 °C for 2 h, significant improvement of the transport J_c at high field more than 9 T is observed for the sample sintered at 900 °C for 3 h. The superconductivity at 4.2 K for the sample was maintained by applying the field of 14 T.

The as-drawn filaments with mixed MgB_2 and 5 at% nanoscale SiC powders were uniaxially pressed under 20 MPa at 200°C for 8 h in air to remove volatile components and to connect the grains. Then the samples were vacuum-sintered at 900 °C for 3 h and 885 °C for 2 h and 880 °C for 2h, respectively. Figure 6 presents the field dependence of magnetic J_c for the samples sintered at 900 °C, 885 °C and 880°C. The J_c value for the sample (control), which was pyrolyzed at 500°C for 0.5h and vacuum sintered at 885°C and 900°C, was also shown in Fig.6. Press heating at 200°C of the sample with SiC doping resulted in significant enhancement of the flux pinning. For example, high magnetic J_c values at 4.2 K such as 2.75×10^5 A/cm^2 and 1×10^4 A/cm^2 at 0.2 T and 4.9 T, respectively were attained for the sample after sintered at 885°C. The X-ray diffraction pattern of the samples was measured. The structure of the sample annealed at 885°C was a mixture of MgB_2, MgO, Mg_2Si and MgB_4 phases. The improvement of the flux pinning for the sample was considered due to the combination of counter-balanced Si and C co-substitution for B as reported as S.X.Dou et.al [5].

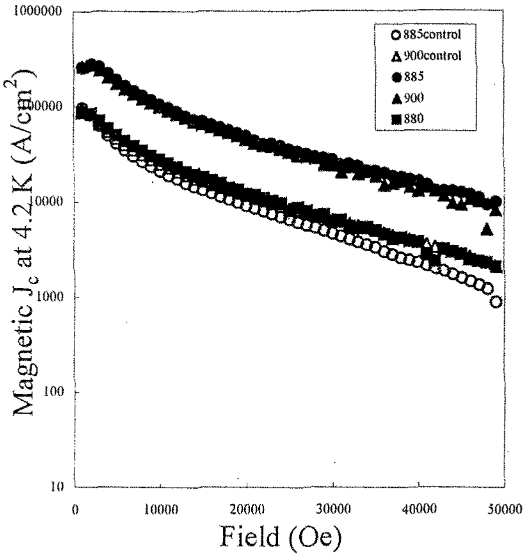


Fig.6. The field dependence of magnetic J_c for the SiC doped sample press heated and then vacuum sintered at 880°C, 885°C and 900°C.

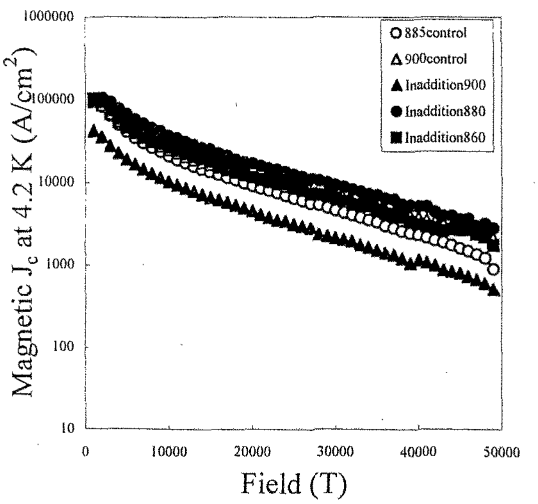


Fig.7. The field dependence of magnetic J_c for the In and SiC doped sample press heated and then vacuum sintered at 860°C, 880°C and 900°C.

The suspension spinning of the mixed powder MgB_2 and 0.5 at% In and 5at% nanoscale SiC was made. The as-drawn filaments were uniaxially pressed under 20 MPa at 200°C for 8 h in air to remove volatile components and to connect the grains. The samples were vacuum-sintered at 900 °C for 3 h and 880 °C for 2 h and 860 °C for 2h, respectively. Figure 7 presents the field dependence of magnetic J_c for the samples sintered at 900 °C, 880 °C and 860°C. The control data are also shown in Fig.7. An enhancement of the flux pinning was observed by press heating at 200°C with 0.5at% In addition and SiC doping. Although the J_c value decreased by In addition, the optimum annealing temperature decreased to 880°C.

4. CONCLUSION

The suspension spinning of commercially available MaB_2 powder and without vacuum sintering was examined to fabricate a long superconducting MgB_2 wire for its easiness of scale up production. The as-drawn filaments were hot-pressed under 20 MPa and 200°C for 8 h. The filaments showed superconductivity at 29 K with transport J_c value of 20 A/cm^2 at 4.2 K and self-field. Indium addition resulted in an enhancement of T_c value to 31K and J_c value to 120 A/cm^2.

Significant enhancement of the flux pinning was observed after vacuum heating at 885°C for the SiC doped sample.

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