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Fabrication of multi-filamentary Y123 superconductor

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The precursor $Y_1Ba_2Cu_3O_x$ (Y123) filaments were prepared by solution spinning through aqueous poly(vinyl alcohol) solution containing mixed Y, Ba and Cu acetates. The as-drawn filaments were heated to remove volatile components and to generate a superconducting phase. The filamentary superconductors were passed through Ag paste and sintered. By controlling the heating condition, the composite with excellent connection between the filaments and metallic Ag matrix was obtained. The overall transport critical current density (J_e) of more than 2000 A/cm^2 at 77 K and 0 T was achieved for the composite with partially melted Y123 filaments. Although the J_e value decreased by applying a low magnetic field less than 0.1 T, the superconductivity of the composite maintained at 10 T at 77 K.

1. Introduction

For a practical application of high T_c superconducting magnets or cables, the superconducting wires are essentially used for a composite with a number of ceramic fiber cores encased in a metallic matrix. The superconducting is not arrayed in this way only to give the composite a mechanical advantage. It is more based on processing considerations and wanted electrical behavior. For the stabilization of superconductivity on the operating system, the electromotive force and heat, which happens to be generated by flux jump and sudden field change, must be removed from the superconductors immediately through the metallic matrix.

One of the most important steps towards the development of practical high T_c superconductors was the fabrication of polycrystalline, silver sheathed, powder-in tube tapes of Bi-Sr-Ca-Cu-O with high critical current density (J_c). Significant progress has been made for the fabrication of long length of $Bi_2Sr_2Ca_1Cu_2O_x$ and $Bi_2Sr_2Ca_2Cu_3O_x$ conductors. The Ag/Bi based superconductor is effectively a composite in which fiber of high stiffness is surrounded by a metal matrix with considerably higher ductility [1]. The intrinsic properties of Bi based compounds, i.e. the temperature dependence of the irreversibility field, appear to limit them to application at lower temperatures (less than 40 K) or in low fields at higher temperatures. Continuous fabrication of $Y_1Ba_2Cu_3O_x$ (Y123) superconductor coated metal fiber and multi-filamentary wire was studied. However the critical current density (J_c) of the composite is extremely low due to the sintered wires [2].

We have studied a preparation of filamentary superconducting oxides of using a textile fiber spinning technology [3]. The maximum J_c value exceeding 10^5 A/cm^2 at 77 K and 0 T was achieved for the melt-textured filamentary Y123 superconductor [4].

In this paper, we report a fabrication of multi-filamentary Y123 superconductor was studied by using the filamentary samples produced by solution spinning.

2. Experimental procedure

The preparation of a long filamentary Y123 superconductor has been reported elsewhere [5]. Y, Ba and Cu acetates were employed for the starting materials. An aqueous solution was prepared by dissolving poly(vinyl alcohol) with a degree of polymerization of 2450 and saponification degree of 88.0 mol%, propionic acid and 2-hydroxy isobutyric acid. A mixture of the acetates with an atomic ratio of Y : Ba : Cu = 1.2 : 2.1 : 3.1 (filament 1) and Y : Ba : Cu = 1 : 2 : 3 (filament 2) was dissolved in the aqueous solution. We concentrated the resultant solution to obtain a stable viscose homogeneous spinning dope. The dope was deaerated at 80 °C for one day and then extruded as a filament into a hot air zone and coiled on a winding drum. We heated the as-drawn filament to 450 °C at a heating rate of 25 °C/h to remove any volatile components. Then filament (1) was sintered at 900 °C for 15 min followed by furnace cooling in flowing oxygen. Filament (2) was partially melted to enhance the J_c of the sample. The obtained superconducting filaments were passed through

a vessel containing Ag paste (Tokuryoku Kagaku Co. Ltd, Silvest P-255) of fine Ag powder dispersed in a nonaqueous liquid. A bundle of the Ag-coated filament was passed through the vessel again. The composite was heated in flowing oxygen to sinter the coated Ag powder. The critical current (I_c) of the composite was measured by a standard four-probe resistive method. Ag paint was used to connect the composite with Ag electrods 200 μm in diameter for supplying DC currents and 100 μm in diameter for voltage leads. Some composites were embedded in a substrate using phenolic resin (PR-50702, supplied by Sumitomo Bakelite) and set on a critical current measuring holder. External magnetic fields were applied in a direction normal to the filament length using a helium-free 11 T superconducting magnet at the High Field Laboratory for Superconducting Materials, Tohoku University. Current was passed along the direction of the fiber axis and was normal to the applied magnetic field. A criterion of 1 μV/cm was used for I_c measurements. The overall J_e value was estimated the I_c divided by the cross-section of the composite.

Adhesion of Ag powders and connectivity between the filaments and Ag matrix were examined by tensile test of the composite. The tensile test of the composite was measured with an Instron type machine at room temperature. A cross-head speed was 0.0333 mm/s and the gauge length was 5 mm. The apparent stress of the composite was estimated as the load divided by the cross-section of the composite and the apparent strain was as the displacement divided by the gauge length.

3. Results and discussion

3.1. Composite of sintered filaments

Filament (1) was sintered at 900 °C for 15 min and cooled to room temperature at a cooling rate of 100 °C/h in flowing oxygen. A diameter of filament (1) was reduced to 100 μm, in spite of starting from the as-drawn filament of 300 μm in diameter. The I_c of filament (1) was ranged from 20 mA to 50 mA at 77 K and 0 T. Filament (1) was coated by Ag paste. Then five or ten Ag coated filaments were embedded in Ag paste. The composite was heated at various temperatures and times and cooled to room temperatures at a cooling rate of 400 °C/h in flowing oxygen to sinter the Ag powder. The overall J_e at 77 K and 0 T, tensile strength and elongation for the composite heated at various heating conditions are shown in Table I. The reasonable J_e value and strength are observed for the composite heated at 850 °C for 30 min. As compared with the mechanical properties of pure Ag metal, such as tensile strength of 124 MPa and elongation of 48%, the mechanical properties of the composite are poor.

TABLE I The overall J_e at 77 K and 0 T, tensile strength and elongation for the composite heated at various heating conditions and cooled to room temperatures at a cooling rate of 400 °C/h in flowing oxygen

Heating condition	J_e at 77 K and 0 T (A/cm^2)	Tensile strength (MPa)	Elongation (%)
600 °C for 15 min	93	—	—
750 °C for 15 min	200	12	2
750 °C for 30 min	110	27	1
850 °C for 15 min	100	45	3.2
850 °C for 30 min	350	39	1.6
850 °C for 60 min	240	43	5.0
900 °C for 15 min	96	16	1.0

TABLE II The overall J_e at 77 K and 0 T, tensile strength and elongation for the composite heated at 880 °C for various times and rapid-cooled to 850 °C or 800 °C and hold for 15 min and then cooled to room temperature at a cooling rate of 100 °C/h in flowing oxygen

Heating condition	J_e at 77 K and 0 T (A/cm^2)	Tensile strength (MPa)	Elongation (%)
880 °C for 0 min, 850 °C for 15 min	70	42	2.4
880 °C for 5 min, 850 °C for 15 min	210	41	2.6
880 °C for 15 min, 850 °C for 15 min	240	44	2.6
880 °C for 5 min, 800 °C for 15 min	240	45	2.2

High heat treatment of the composite was examined to improve the adhesion of the Ag powder and connectivity between the filaments and Ag matrix. The composite was heated at 880 °C for various times and rapid-cooled to 850 °C or 800 °C and hold for 15 min and then cooled to room temperature at a cooling rate of 100 °C/h in flowing oxygen. The overall J_e at 77 K and 0 T, tensile strength and elongation of the composite are presented in Table II. The interface between the matrix and filaments was considerably improved for the composite heated at 880 °C for 5 min. However, some voids between the filaments and matrix were still observed for the composite. The Ag coating was pyrolyzed up to 450 °C at a slow heating rate of 25 °C/h in air to remove volatile components of the Ag paste. Then the composite was heated at 880 °C for various times and rapidly cooled to 850 °C and hold for 15 min followed by cooling to room temperature at a cooling rate of 100 °C/h in flowing oxygen. Table III lists the overall J_e at 77 K and 0 T, tensile strength and elongation for the composite. The high overall J_e and tensile strength are attained for the composite heated for 5 min. The cross-section of the composite is shown in Fig. 1. The filaments are embedded in metallic Ag matrix and the connection between the filaments and matrix is fairly good.

3.2. Composite of partially melted filaments

A partial melting of filament (2) was made to enhance the overall J_e of the composite. Filament (2)

TABLE III The overall J_e at 77 K and 0 T, tensile strength and elongation for the composite. The coated Ag was pyrolyzed up to 450 °C at a slow heating rate of 25 °C/h in air to remove volatile components of the Ag paste. Then the composite was heated at 880 °C for various times and rapidly cooled to 850 °C and hold for 15 min followed by cooling to room temperature at a cooling rate of 100 °C/h in flowing oxygen

Heating condition	J_e at 77 K and 0 T (A/cm^2)	Tensile strength (MPa)	Elongation (%)
880 °C for 5 min	240	65	2.8
880 °C for 15 min	160	46	2.0
880 °C for 30 min	140	45	3.0

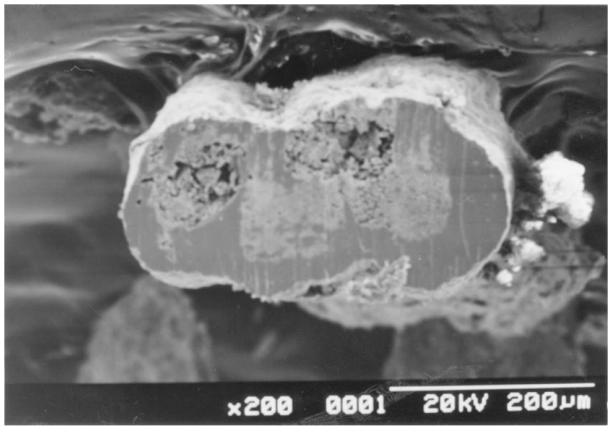

Figure 1 The cross-section of the composite with sintered filamentary superconductors.

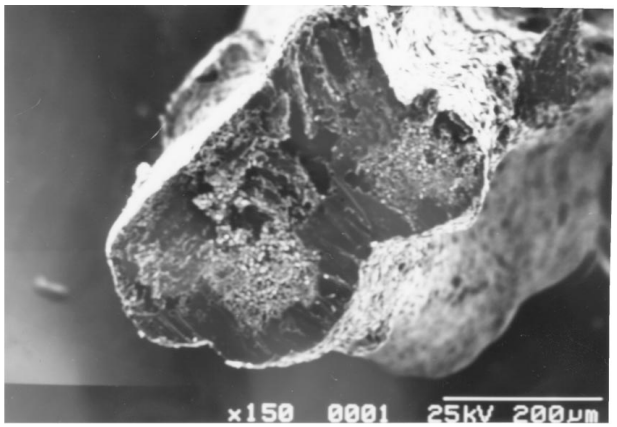

Figure 2 The cross-section of the composite with partially melted filamentary superconductors.

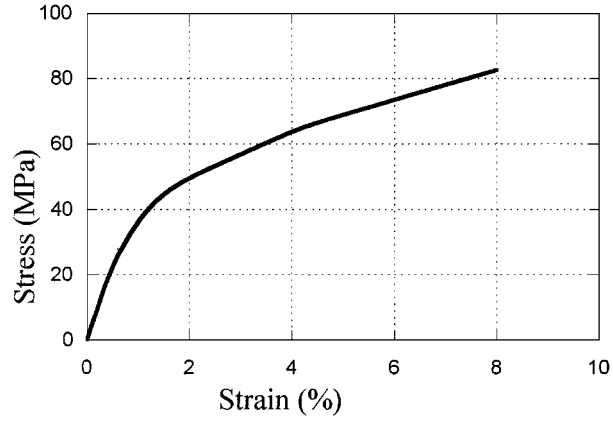

Figure 3 Apparent stress and strain curve for the composite with partially melted filamentary superconductors.

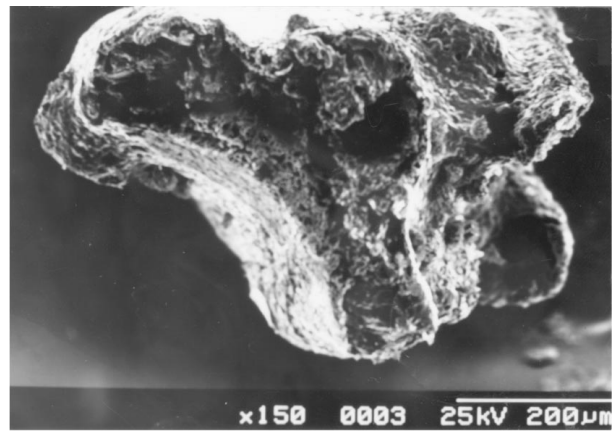

Figure 4 Fracture surface of the composite with partially melted filamentary superconductors after tensile test.

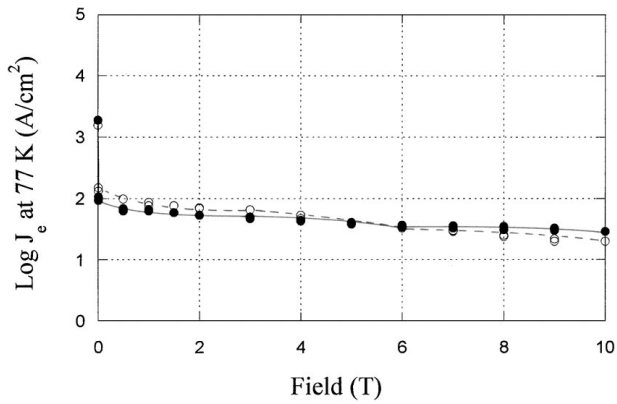

Figure 5 J_e vs. applied field for two samples of the composite with partially melted filamentary superconductors.

was partially melted at 1040 °C for 30 min and was cooled to 920 °C at a cooling rate of 60 °C/h, subjected to slow cooling to 860 °C at a cooling rate of 30 °C/h and then furnace-cooled in flowing oxygen [5]. The I_c of the filament is ranged from 0.3 A to 0.6 A. The filaments were coated by Ag paste. A bundle of the Ag-coated filaments was passed again through the vessel of Ag paste. The composite was pyrolyzed to 450 °C at a heating rate of 25 °C/h to remove volatile components of the Ag paste. Then the composite was heated at 880 °C for 5 min and rapidly cooled to 850 °C and hold for 15 min and cooled to room temperature at a cooling rate of 100 °C/h. The overall J_e of the composite was improved to 2100 A/cm^2 at 77 K and 0 T. The cross-section of the composite is shown in Fig. 2. Although the examination of the strain dependence of I_e for the composite is necessary [6], the tensile test of the composite was examined to check the sintering of Ag powder and connection between the filaments and matrix. The typical apparent stress-strain curve of the composite is shown in Fig. 3. The apparent tensile strength of 82 MPa and elongation of 8.0% due to the mechanical property of Ag metal are detected. Fracture surface of the composite after tensile test is shown in Fig. 4. A ductile deformation of the matrix metal is observed.

The overall J_e for the composite was examined in magnetic fields up to 10 T at 77 K. Fig. 5 shows the field dependence of the J_e for two samples of the composite. Although the J_e values of both samples reduced by applying a low field, a drop of the J_e in the magnetic field ranging from 0.5 T to 10 T is small. The field dependence of J_c for Y123 superconductor is sensitive for the weak link at the grain boundary, which affects J_c in low fields, while a pinning effect appears in a high field region. The magnetic field dependence of transport J_c for the filamentary Y123 superconductor produced by solution-spinning method and partial melting process was examined. The field dependence of the J_c for the filamentary sample with randomly

aligned texture normal to the fiber axis was better than that for the sample with aligned texture to the direction of the filament diameter. The J_c value of more than 10^2 A/cm^2 at 10 T was maintained for the filamentary Y123 [7]. The degradation of J_e in low field for the present composite indicates weak-link behavior of the filamentary superconductors. Although the J_e value of the composite decreased rapidly with the applied field, it is worth noting that the both samples maintained superconductivity up to 10 T at 77 K ($J_e =$ 29 A/cm^2 at 77 K and 10 T).

4. Conclusion

Fabrication of the multi-filamentary Y123 superconductors was examined. Filamentary Y123 superconductors were prepared by solution spinning and sintering or partial melting process. The filamentary superconductors were passed through the vessel of Ag paste. A bundle of the Ag coated filaments was passed again through the Ag paste. The composite was sintered under various heating conditions in flowing oxygen. By controlling the sintering condition, the composite with excellent connectivity between metallic Ag matrix and superconducting filaments was obtained. The high overall J_e of more than 2000 A/cm^2 at 77 K and 0 T was attained for the composite with partially melted filaments. Although the J_e value of the composite decreases with applying a low field, it is worth noting that the composite maintained superconductivity up to 10 T at 77 K.

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