Advances in Fabrication of Ag-clad Bi-2223 Superconductors

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***Abstract*—Powder-in-tube (PIT) processing was used to fabricate multifilamentary Ag-clad $Bi_2Sr_2Ca_2Cu_3O_y$ (Bi–2223) superconductors for various electric power applications. Enhancements in the transport current properties of long lengths of multifilament tapes were achieved by increasing the packing density of the precursor powder, improving the mechanical deformation, and adjusting the cooling rate. The dependence of the critical current density on magnetic field and temperature for the optimally processed tapes was measured. J_c was greater than 10^4 (A/cm^2) at 20 K for magnetic field up to 3 T and parallel to the c-axis which is of interest for use in refrigerator cooled magnets. An attempt was made to combine the good alignment of Bi-2223 grains in Ag-sheathed superconducting tapes to obtain high J_c values at high temperature and low field, and good intrinsic pinning of $YBa_2Cu_3O_{7-d}$ (Y-123) thin film to maintain high J_c values in high fields. A new composite multifilament tape was fabricated such that the central part contained Bi-2223 filaments, with the primary function of conducting the transport current. The central Bi-2223 filaments were surrounded by Y-123 thin film to shield the applied magnetic field and protect the Bi-2223 filaments. The J_c values of the composite tape were better than those of an uncoated tape. In the case of 77 K applications, an I_c of about 60 A was obtained in a 150 m long tape and zero applied magnetic field. In-situ strain characteristics of the mono- and multifilament tapes were conducted.**

I. Introduction

Large critical current density (J_c) in superconducting wires and tapes is essential for many practical applications. Material processing still remains the key factor in realizing the potential of high temperature superconductors. The powder-in-tube (PIT) process, which yields a highly textured $(Bi,Pb)_2Sr_2Ca_2Cu_3O_y$ (Bi-2223) superconductor with its c-axis aligned parallel to the tape surface, is an industrially scalable technique for fabricating long-length superconductors [1-5]. A significant progress has been made over the past several years in improving J_c's in wires and tapes to be sufficently high for some commercial applications.

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In zero applied magnetic field, the critical current density of superconducting tape is controlled by its microstructure. Grain boundaries act as barriers to the transfer of transport current between grains. The crystallographic anisotropy of Bi-2223, which exhibits a micaceous or platelike morphology, allows large contact areas, alignment of grains with their c-axis perpendicular to the rolling direction of the tape, easy transfer of current across its grain boundaries, and high J_c values [6-9].

In an applied magnetic field at temperatures above 35 K, $YBa_2Cu_3O_{7-\delta}$ (Y-123) shows much better J_c response than Bi-2223. The irreversibility line (IRL) of high-T_c materials can provide insight into their limitations for various applications. Recent progress in growing single crystals of Bi-2223 provided an opportunity to study its IRL line [10,11]. A sharp irreversibility field (H*) drop was observed between 20 and 40 K. Furthermore, H* was only ≈ 0.3 T at 75 K.

This observation clearly shows very weak intrinsic pinning in Bi-2223 single crystals. Intrinsic pinning in Y-123 single crystal is sufficiently strong to keep H* at 77 K to ≈ 8 T [12,13]. However, the fabrication of Y-123 wires by the PIT technique has not been successful because of YBCO's granularity. Y-123 exhibits greater isotropy than Bi-2223 and its intergranular transport current is poor because of weak links.

In order to fabricate long lengths of superconducting tape with high critical current densities, it is necessary to optimize the conductor uniformity along its length. This broad category includes parameters such as: initial powder properties, deformation processing, and thermomechanical conditions. Recently, we have varied the packing density of the precursor powder, improved the mechanical processing and modified the heat treating schedule; the results are described in this paper.

Furthermore, a new composite multifilament tape was fabricated such that the central part contained Bi-2223 filaments, with the primary function of conducting the transport current. The central Bi-2223 filaments were surrounded by Y-123 thin film to shield the applied magnetic field and protect the Bi-2223 filaments. This new coated tapes showed improved J_c performance in external magnetic field.

DISCLAIMER

II. EXPERIMENTAL PROCEDURE AND RESULTS

As in our previous study [14], multifilament Ag-clad Bi-2223 tapes were made by the PIT technique with precursor powder having the overall stoichiometry of Bi-2223. The precursor powder contained Pb-added 2212, Ca_2PbO_4, alkaline-earth cuprate and CuO phases. Packing density in the Ag tubes was varied by using precursor powder, including those prepressed into billets, that were inserted into the Ag tubes. The precursor powder was packed into the Ag tubes at a density of $\approx$ 2.3 g/cm^3, while the precursor billets were of two densities: 3.5 g/cm^3 (low packing density) and 4.5 g/cm^3 (high packing density). The powder and prepressed billet Ag tubes were swaged, drawn through a series of dies and then rolled to a final thickness of $\approx$ 200 µm. The standard mechanical processing consisting of > 10% reduction per pass was used in the fabrication of these tapes.

Samples measuring 1.5 m in length were cut from these three tapes and heat treated in 8% oxygen atmosphere. The transport critical current were measured at 77 K, self field with 1 µV/cm criterion. The higher packing density resulted in higher I_c values after heat treatment at 820°C and were maintained uniformly over the entire length [14].

In another set of experiments, we varied the mechanical deformation schedule. The Ag tubes were drawn and rolled according to various reduction ratios per pass. Load cells were mounted on the dies, and pressure exerted on the wires being drawn was monitored. Reduction ratios per pass were optimized on the basis of the die pressure measurements. Microstructures were characterized by scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDS). Figure 1 is a composite showing the typical cross-sectional area of the 37 filament tape at low magnification. For the case of high packing density billet, there was a cross-link between adjacent filaments, especially towards the edges of the tape. This effect was not observed in the case of low density billet.

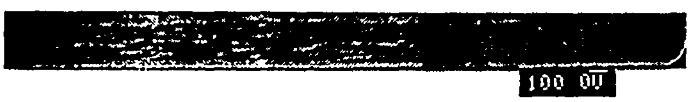

Fig. 1. SEM photomicrograph showing the cross-sectional area of a 37 filament tape (high packing density billet).

Improved mechanical processing of the high density billet showed a pronounced effect on the uniformity of the Ag-superconductor interface. Figure 2 is a composite of several SEM images showing the effect of mechanical deformation on the uniformity of the Ag-superconductor interface. The smoothness of the Ag-superconductor interface is improved. This effect is important for this processing method because the interface plays an important role in controlling the grain morphology and texture of 2223 grains. The more coherent Bi-2223 / Ag interface for the light reduction specimens resulted in higher I_c measurements. The alignment of grains and their junctions have been dicussed in terms of current transport across the grain boundaries by two models: the brick-wall [15], and railway-switch [16]. In the brick-wall model, the c-axis oriented grains form [001] twist boundary along the long face of each grain. The current is assumed to flow predominantly along CuO_2 planes and transfer between grains across large area Josephson junctions. This model describes the low-temperature J_c - H_{app} behavior well. The transport critical current along the c-axis is seen as the bottleneck for current flow in the tape. In the railway model, the current transfer from one grain to another across the small angle [100] tilt boundary. Colonies exhibiting this type of connectivity are responsible for current transfer. In this model, the small angle tilt boundaries are assumed to be the strongest links and therefore responsible for the current transfer. Significant currents can move across the tilt boundaries. The microstructural observations seem to support both models [18].

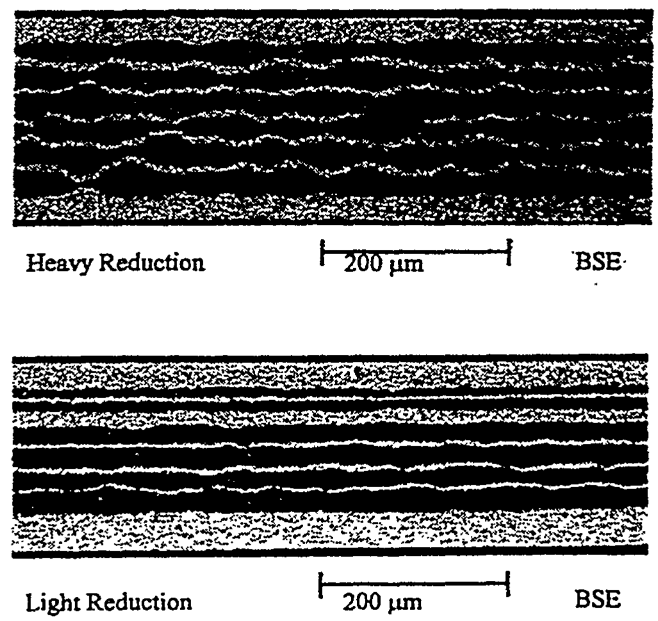

Fig. 2. SEM photomicrogarph showing the longitudinal cross-section of the 37 filament tape (improved mechanical processing for high packing density billet).

Several research groups have reported that in Ag-clad Bi-2223 tapes supercurrent is transported through a thin region at the silver-superconductor interface [6, 19-22]. The high current superconducting layers are 2-3 µm thick and next to the silver. Transport J_c values of tapes with identical superconductor cross-sectional area but differing Ag/Bi-2223 interfacial lengths confirm the importance of the interfacial region [21]. The critical current was shown to be proportional to the Ag/Bi-2223 interface perimeter length

(IPL), expressed as a linear function. These results imply that <10% of the Bi-2223 superconductor transports the vast majority of supercurrent in Ag-clad Bi-2223 tapes.

The dependence of the critical current density on magnetic field and temperature for the optimally processed tapes was measured. Magnetic hysteresis measurements were made in a SQUID magnetometer over a temperature range 20-77 K in magnetic fields up to 5 T. The magnetic field was alligned normal to the tape surface, which was the direction of the c-axis for the textured tapes. A 3-cm scan was used and the field was swept in a no-overshoot mode. The Bean model was used to derive J_c values from the magnetic hysteresis loops [23]. Figure 3 shows the magnetization J_c (H_{app}) at several temperatures. J_c was greater than 10^4 (A/cm^2) at 20 K for magnetic field up to 3 T and parallel to the c-axis which is of interest for practical application in refrigerator cooled magnets. At the higher temperatures the sharp initial decline in J_c is followed by exponential decline with increasing magnetic field. The anisotropy of Bi-2223 with increasing temperature and field is pronounced at temperatures above 40 K and fields above 1 T.

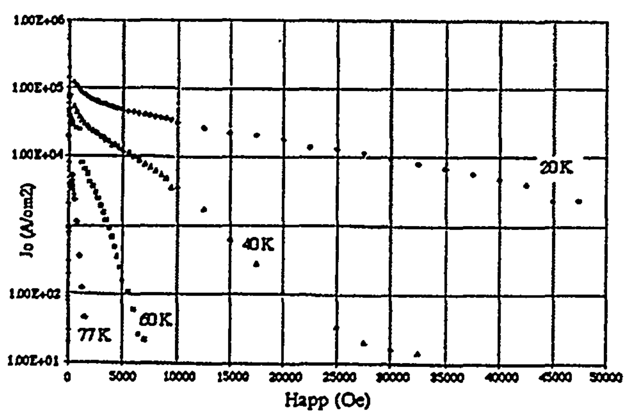

Fig. 3. Magnetization J_c vs magnetic field applied parallel to the c-axis at at different temperatures (SQUID data). J_c greater than 10^4 (A/cm^2) at 20K and 3 T field.

In the case of applications at 77 K, an I_c of ≈ 60 A was obtained in a 150 m long tape and zero applied magnetic field. Figure 4a shows the I-V characteristics of the superconducting tape that carried 42 A in a zero magnetic field. Magnetic field up to 0.4 T was applied along the c-axis. Magnetic field of ≈ 0.2 T (2000 Oe) brings the I_c value from 42 A down to 4 A, and changes the slope around the I_c value [20-22]. These results strongly suggest that ≈ 0.2 T is the "irreversibility field" H* at 77K. Figure 4b shows exponential decline of I_c with H_{app} at 77K (H_{app} < 0.2 T). These results are consistent with the observation that $I_c(0)$ and $I_c(H_{app})$ in Bi-2223 tapes are controlled by different mechanisms: while $I_c(0)$ is controlled by the transfer of current between grains, $I_c(H_{app})$ is controlled by both "intragranular" and "intergranular" effects of flux pinning [22]. Intragranular effects include intrinsic and extrinsic factors. Intrinsic factors arise from large anisotropy of Bi-2223 and very short coherence length along the c-axis. With a magnetic field H_{app} parallel to the c-axis of anisotropic Bi-2223 grains, a stacking of 2D pancake vortices forms in CuO_2 layers and these layers are weakly coupled to each other [23-25]. The interlayer coupling between 2D pancakes becomes weaker with increasing magnetic field. At the certain field, well below the upper critical field, the motion of vortices is strong enough to bring J_c value to zero. J_c remains relatively field independent for fields applied along the CuO_2 planes. Extrinsic factors such as dislocations, surface steps, small particles of secondary phases tend to pin vortices and improve the J_c values.

Intergranular effects include only extrinsic factors such as grain alignment, presence of secondary or amorphous phases along the grain boundaries, microcracks, etc. These factors tend to produce weak links along the boundaries and reduce the transport current.

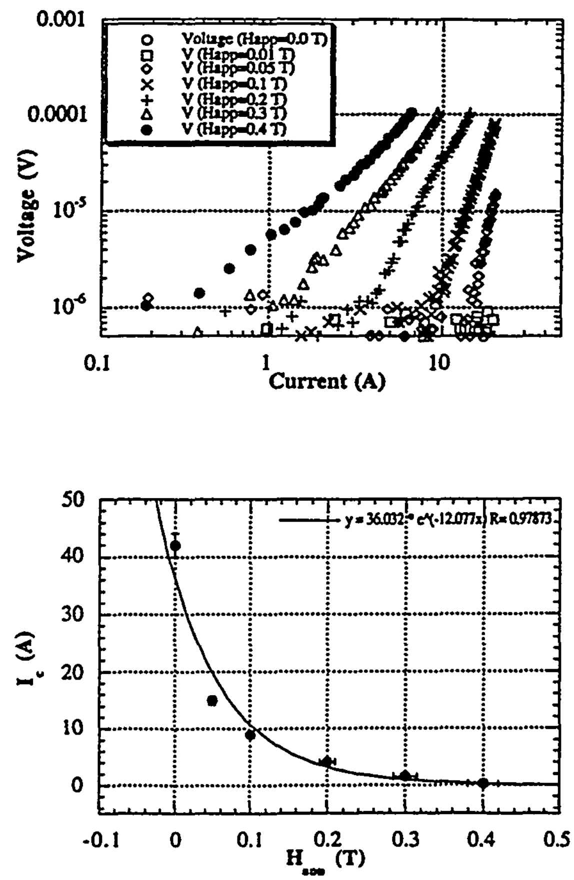

Fig. 4. a) Voltage - current characteristics for the multifilament tape at 77 K and magnetic field along the c-axis; b) exponential dependence of I_c as a function of the applied field at 77 K.

The concept of 2D pancake vortices suggest that optimal pinning sites will be defects approaching interlayer spacing for pancake vortices. The creation of artificial pinning centers by heavy ion irradiation [26,27], or introduction of columnar inclusions of non-superconducting material that extends over the entire sample in the direction of applied field [28,29] are approaches in enhancing the flux pinning. The interaction of vortices with correlated defects increases the pinning energy and effectively increases the resistance to the motion of vortices.

The heavy ion irradiation produces amorphous tracks through the sample thickness with size on the order of the coherence length along the a,b planes (< 50 Å). The critical current densities in magnetic fields are strongly influenced by the irradiation defects [27]. The decay of both transport and magnetization currents in external magnetic fields are less pronounced after irradiation.

A chemical approach to the formation of columnar defects by using nanorods of MgO was developed [28,29]. Measurements of the critical current density as a function of temperature and field demonstrated that MgO nanorods enhanced J_c at elevetaed temperatures and magnetic fields. Also, a shift in the irreversibility line towards higher temperatures was observed in samples with MgO nanorods.

An attempt was made to combine the good alignment of Bi-2223 grains in Ag-sheathed superconducting tapes to obtain high J_c values at high temperature and low field, and good intrinsic pinning of $YBa_2Cu_3O_{7-\delta}$ (Y-123) thin film to maintain high J_c values in high fields [30]. A new composite multifilament tape was fabricated such that the central part contained Bi-2223 filaments, with the primary function of conducting the transport current. The central Bi-2223 filaments were surrounded by Y-123 thin film to shield the applied magnetic field and protect the Bi-2223 filaments.

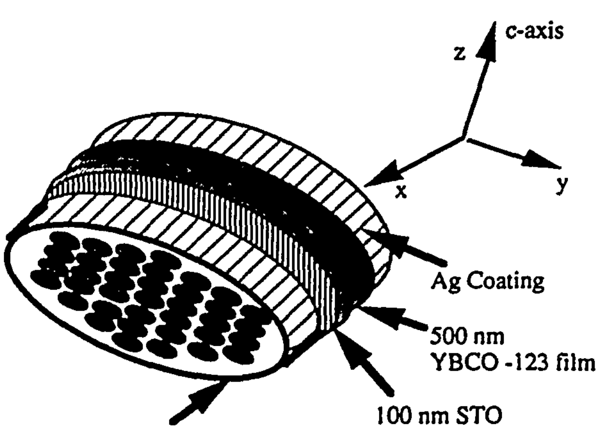

Fig. 5 Schematic diagram of Y-123-shielded Bi-2223 multifilament tape.

The as-rolled tape was cut into 4-cm lengths and Y-123 thin films were grown by off-axis magnetron sputter deposition [31]. A 100-nm-thick layer of $SrTiO_3$ (STO) was deposited as a buffer. $Y_{1.1}Ba_2Cu_3O_{7-d}$ was sputtered in a 200-mtorr gas mixture of argon and oxygen. The substrates were at ≈ 700°C. Figure 5 shows the experimental arrangement. Coated and uncoated tapes were heat treated according to the Bi-2223 schedule.

The magnetic susceptibility measurement as a function of temperature for Y-123 thin film in the as-coated tape at H_{app} = 100 Oe parallel to the c-axis showed the transition temperature $T_c \approx 72$ K along with the broad transition region [30]. The lower T_c was possibly due to the surface structure of thin film grown on Ag-sheathed Bi-2223 tape. The texture of the Ag substrate should have an effect on the lattice of Y-123. The reason for growing STO as a buffer layer was to lower the lattice mismatch between Y-123 and Ag. Also, it is possible that the oxygen deficiency in the as-coated state caused lowering of the T_c.

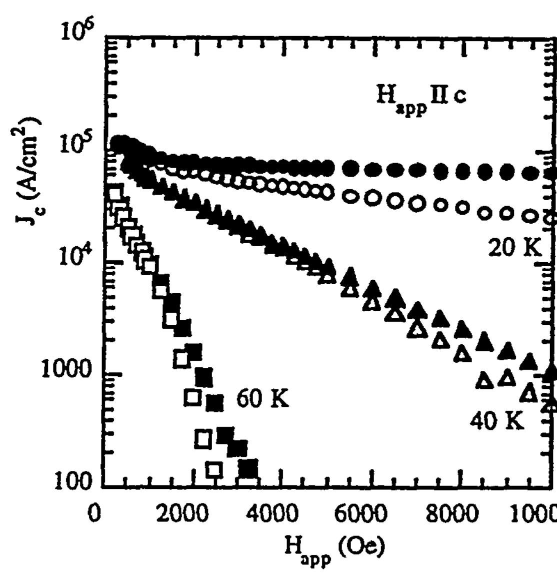

Fig. 6. Magnetization J_c vs magnetic field applied parallel to the c-axis at 20, 40, and 60 K: solid triangles represent Bi-2223 tape coated with Y-123 thin film and heat treated; open squares represent uncoated, and heat-treated reference tape.

Figure 6 shows the J_c dependence of a heat-treated Bi-2223 tape coated with Y-123 thin film on H_{app} and T, the reference tape in the figure was uncoated but heat treated under the same conditions. A magnetic field was applied parallel to the c-axis for all measurements performed at 20, 40, and 60 K. The increase in J_c is attributed to the magnetic contribution of the Y-123 thin film to the Bi-2223 grains.

Its platelike morphology should enhance surface pinning. The increase is more pronounced at lower temperatures and at higher fields, a finding that is similar to the effect of the enhanced pinning by columnar defects in the Bi-2212 system [28]. In our case, the effect is shifted to lower temperatures because of the critical temperature of Y-123 thin film. Also, the magnitude of the effect is affected by the misorientation of the Y-123 grains. However, the results of our work show that this approach can be used to enhance J_c response of Bi-2223 phase at higher temperatures and in higher magnetic fields. Moreover, from the processing point of view, the results show that heat treating Y-123 thin film according to the Bi-2223 tape schedule is compatible with and beneficial for Y-123.

Poor mechanical properties have seriously hampered the commercial application of high T_c superconductors. During fabrication and service, the conductors are subjected to axial and bending stresses. In operation, the material is subjected to additional stresses by temperature gradients and magnetic fields. In large and/or high-field magnets, electromagnetic hoop stresses could even reach the ultimate strength of the material. These stresses can cause microstructural damage in the conductors and thereby degrade current transport properties. Although silver is widely used as a sheath material, its mechanical properties are not adequate to withstand the stresses developed during fabrication and service. Therefore, techniques such as adding silver to the superconductor powder, using alloy sheath material as an alternative to silver and fabricating multifilament conductors have been developed to improve the strain tolerance characteristics of the conductors [32-36].

The use of Ag-2at%Mg alloy as an outer sheath in multifilament tapes provided excellent mechanical properties [37]. The use of AgMg outer tube combined two advantages: having a pure Ag sheath in contact with Bi-2223 filaments avoiding possible chemical reaction, and a ductile, deformable outer tube of AgMg which becomes strengthened through the MgO dispersions formed during the heat treatment of the tape. However, the critical current reached only 80% of the value for reference tape with pure Ag sheath. The indication was that the oxygen exchange between flaments was modified which affected the formation of 2223 phase and therefore the transport current values.

Several multifilament Bi-2223 tapes were made using pure Ag and Ag-alloy (AgMg or AgMn) sheaths [38]. The slope n of the V-I curves in a double logarithmic plot was taken as an indication of the tape quality. The microscopic defects such as microcracks in the filaments should force the current to flow locally through the Ag sheath reducing the n value [39]. The n value in tapes made using Ag-alloy was 14-15 compared with n value of 20-22 for tapes with pure Ag [38]. These transport measurement results strongly suggest that the J_c reduction in Bi-2223 multifilament tapes with Ag-alloy sheaths occurs due to the local microscopic defects in individual filaments.

To evaluate the strain tolerance characteristics of the Bi-2223 tapes, in situ bending tests were conducted. Figure 7 shows bending characteristics of mono and multifilament conductors. It shows that ε_{irr} for the monofilament conductor increases with decrease in superconductor fill factor. In multifilament tapes, the added Ag increases the strain tolerance of the tape which is consistent with reported I_c values for bending strains [40,41]. The improvement in mechanical tolerance for bending in multifilament tapes is possibly due in part to the better grain alignment of Bi-2223 grains. However, for applying these tapes to large scale devices, the appropriate data for strain degradation should be determined by the axial tensile tests, if the primary stress in the devices is a hoop stress [40].

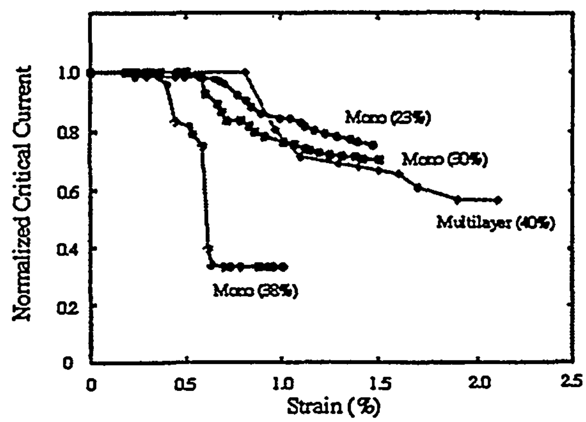

Fig. 7 Strain tolerance of Ag-clad Bi-2223 mono- and multifialment tapes

III. CONCLUSIONS

Transport current properties in multifilament Ag-clad Bi-2223 superconducting tapes were improved by varying the mechanical and thermal parameters during tape processing. The packing density of precursor powder, improved mechanical deformation and cooling rate all had a pronounced effect on the critical current of the superconducting tapes. The dependence of the critical current density on magnetic field and temperature for the optimally processed tapes was measured. J_c was greater than 10^4 (A/cm^2) at 20 K for magnetic field up to 3 T and parallel to the c-axis which is of interest for use in refrigerator cooled magnets. An attempt was made to combine the good alignment of Bi-2223 grains in Ag-sheathed superconducting tapes to obtain high J_c values at high temperature and low field, and good intrinsic pinning of $YBa_2Cu_3O_{7-\delta}$ (Y-123) thin film to maintain high J_c values in high fields. A new composite multifilament tape was fabricated such that the central part contained Bi-2223 filaments, with the primary function of conducting the transport current. The central Bi-2223 filaments were surrounded by Y-123 thin film to shield the applied magnetic field and protect the Bi-2223 filaments. The increase in J_c is attributed to the magnetic contribution of the Y-123 thin film to the Bi-2223 grains. In the case of 77 K applications, an I_c of about 60 A was obtained in a 150 m long tape and zero applied magnetic field. In-situ strain characteristics of the mono- and multifilament tapes were conducted.

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