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High critical currents in Bi(2223) tapes with Ag and hardened Ag sheaths

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Abstract

High critical current densities up to J_c = 40 000 Acm^{-2} (77 K, 0 T) were obtained in Bi(2223)/Ag short tape samples, produced by conventional powder in tube technique and cold deformation. Towards the elimination of the final pressing and heating treatments of the tapes and with the expectation of an enhanced matrix stability, first samples with dispersion hardened AgMg sheaths were prepared. Their current carrying capacity and mechanical properties and the microstructure of the Bi(2223) filaments were investigated and compared to the conventional prepared tapes. Without pressing treatments critical currents of 8000 - 10 000 Acm^{-2} were achieved using AgMg tubes.

1. Introduction

For Bi(2223)/Ag tapes, high critical transport currents up to J_c = 53 000 Acm^{-2} (77, 0 T) [1] were obtained for short sample pieces, applying final sequences of unidirectional pressing steps for filament densification and heat treatments for phase formation. For longer tape lengths, where unidirectional pressing of the sample is not applicable, the measured transport currents are commonly 3 - 4 times lower. One of the main reasons for this discrepancy is the unsufficient filament densification when pressing is substituted by rolling. The composite combination of Ag, a ductile and very weak matrix, and the Bi(2223) precursor, Bi(2212) + add. phases, being a not fully dense and brittle material, leads to an unfavourable cold deformation behaviour of the tape, which is dominated by the mechanical properties of the filament. As a result the geometry of the tape cross section is not ideal and the interface between the filament and the Ag sheath is too rough. During the heat treatments a significant growth of the tape thickness was observed due to the formation and crystal growth of the Bi(2223) phase and the plastic deformation of the soft Ag matrix [2]. An elimination of these negative tape properties is expected if a harder sheeth material or a matrix reinforcement can be applied. Additional, for future application in high magnetic fields, a reinforced sheath which withstands the Lorentz forces is required. A possible solution of this problem could be the reinforcement of the Ag itself by means of dispersion hardening, also known as internal oxidation, being already industrial applied for e.g. electrical contacts. The advantage of these Ag rich alloys as tape matrix is the preserved permeability for oxygen, a negligible ecrease of the Ag melting point and the heat resistance of the hardening effect. In this work the AgMg element combination was chosen which seems to be the most favourable compound with respect to the chemical influence on the filament and the heat resistivity at the tape annealing temperature of 835 - 840 °C.

We will present in this work first results on the mechanical and electrical properties of Bi(2223)/AgMg tapes compared to the conventional Bi(2223)/Ag tapes, being prepared by exactly the same preparation procedure.

2. Experimental

2.1 *Tape preparation*

The starting material for the Bi(2223) tapes was an oxalate precursor, which was pressed into Ag tubes (6 mm diam., 1 mm wall thickness) after calcination at 800° - 820 °C (50 h) with several intermediate grinding steps. The tubes were cold worked, by swaging to 2 mm diam., drawing to 0.66 mm diam., followed by rolling down to 90 - 100 µm.

Two variations for the rolling deformation steps were applied: A: 200, 2 x 100, 4 x 50, 3 x 20 µm and B: 57 x 10 µm reduction of tape thickness upon rolling.

The final heat treatment of short tape pieces (20 - 40 mm) was performed at 835 - 841 °C for typically 30 + 50 + 50 h (in air) with 2fold intermediate pressing (5 GPa). We used a fixed overall composition of the powder, being $Bi_{1.72}Pb_{0.34}Sr_{1.83}Ca_{1.97}Cu_{3.13}O_x$.

2.2 *Ag Mg preparation*

The AgMg alloys were molten from Ag and Mg pieces in a graphite crucible under Ar

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atmosphere. Afterwards the blocks were swaged to cylinders and supplied with a drilled central hole to form the tube. Three Mg concentrations, 1, 2 and 3 at. % were used. The Bi(2223)/AgMg tapes were cold deformed following scheme B, whith no prior internal oxidation treatment of the sheath.

2.3 *Characterization* of the tapes was performed by means of Vickers microhardness, SEM with EDX and four probe critical current investigations at T = 10 - 77 K and B = 0.2 and 0.4 T.

3. Results

3.1 *Mechanical properties*

The cold deformation of the tapes worked very well for all three used Mg concentrations. The Vickers microhardness H_v (measured on the tape surface) was controlled through the whole final annealing (ann.) and pressing (pr) procedure as shown in fig.1. The starting values of the AgMg alloys were 100 - 120 Kp mm^{-2} compared to $\approx$ 70 kp mm^{-2} for pure Ag. The internal oxidation of the AgMg sheath was performed during the first heat treatment step and is expected to be completed after 1 - 2 h at 840 °C. The observed Hv values of about 55 - 85 Kp mm^{-2} were well below the expected hardness of about 120 kp mm^{-2}. The explanation is that temperatures above 700 °C reduce the hardness with prolonged annealing time due to Mg diffusion, coarsening of the MgO particles and creating MgO intergrain layers, as was confirmed by optical microscopy [4]. Regarding this, the observed H_v values after 50 h/840 °C heat treatment are reasonable and in accordance with the commonly observed behaviour of AgMg alloys.

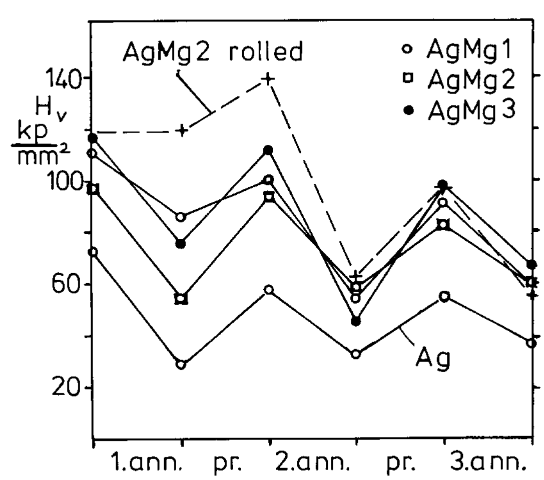

Figure 1 Vickers microhardness H_v of the Ag, AgMg sheaths of Bi(2223) tapes during the final heat-pressing treatment (see text).

This interpretation is confirmed regarding tapes where pressing was replaced by rolling. After the shorter annealing time of 24 h the optimum H_v value of 120 Kp mm^{-2} was observed, whereas the further treatments led also to a reduced final matrix hardness, comparable to the case of pressed samples. After each pressing treatment a work hardening effect was observed. Nevertheless the final hardness of the Ag Mg sheath in pressed tapes of 60 - 70 Kp mm^{-2} was more than 50 % higher compared to pure Ag.

Regarding the change of the tape thickness during the final treatments (see fig. 2) a clear advantage using hardened Ag was observed, where the tape thickness reduces by 10 - 15 µm to a final thickness of 90 - 100 µm. A reduced densification effect was observed for the reference tape with Ag matrix. Therefore internal oxidized AgMg obviously withstands better the filament expansion during reaction and therefore supports the densification of the filament. The optical micrograph of an axial cut of a Bi(2223)/AgMg tape shows a quite regular filament thickness (fig. 3) illustrating our general observation of a very positive influence of the hardened sheath on the final filament geometry after cold deformation, especially avoiding sausaging.

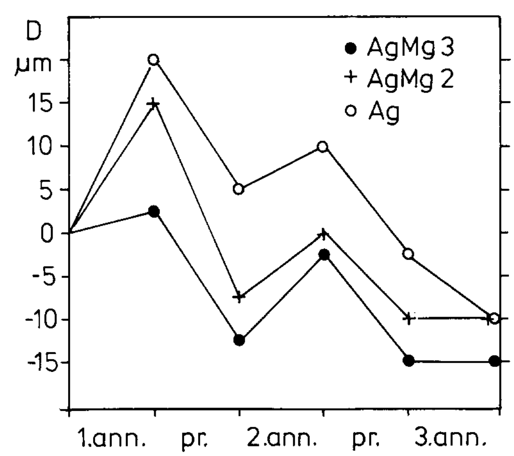

Figure 2 Variation of the tape thicknesses during the final heat-pressing treatments.

3.2 *Microstructure*

The microstructure of the tapes was investigated by means of SEM, EDX and optical microscopy. The internal oxidation of AgMg led to strongly reduced final grain sizes in the matrix material of typically 2 - 10 µm being one order of magnitude smaller than for pure Ag (50 - 100 µm). For 3 at. % Mg content well developed nucleation of MgO at the grain boundaries was observed while for a lower Mg content mainly spherical submicron MgO inclusions were found by SEM.

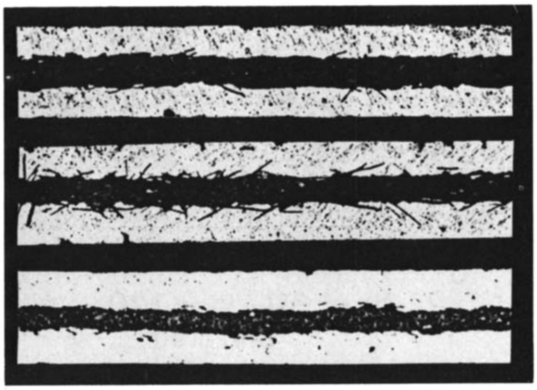

Figure 3 Optical micrographs of longitudinal cuts of Bi(2223) tapes with (from top) Ag sheath (pressed), AgMg1 (pressed), AgMg2 (rolled).

The filament surface was investigated removing the Ag by etching. The filament surface (see fig. 4) typically shows the occurrance of misoriented out of plane standing Bi(2223) grains on the textured oxide material. These grains can also be observed in the micrograph of fig. 3 as oxide channels in the Ag matrix. Hensel et a. [4] claimed this effect as one contribution for a transport current limitation since these crystals penetrate the oriented Bi(2223) layer. In tapes with pure Ag sheaths we found a qualitative relation between a reduced number of misoriented grains and enhanced transport currents.

For AgMg sheaths being pressed we observed a significant enhancement of the occurrance of these misoriented grains, which is a nonfavourable situation for high transport currents (fig. 4). It seems that the much smaller grain sizes in the sheath offer more grain boundaries at the sheath-filament interface to allow oxide grains to grow into the tape matrix obviously supported by microcracks through pressing, since this effect is not observed for tapes deformed only by rolling (see fig. 3).

3.3 *Texture of the Bi(2223) phase:*

The quality of the oriented Bi(2223) phase growth was characterized by means of X ray rocking curves measured on filament surfaces (removed silver) for the axial direction (avoiding broadening from radial filament curvature). For Ag sheathed tapes we typically measure a misorientation of 8° (half width, omega scan) compared to the value of 10° obtained for a Ag Mg sheathed tape.

Therefore with the applied tape preparation method no effect of the AgMg sheath towards an improved quality of the oriented Bi(2223) grain growth was observed. Electron microscopy also confirms the Bi(2223) grain alignment inside the filament but no enhancement of the quality of grain alignment (see fig. 4).

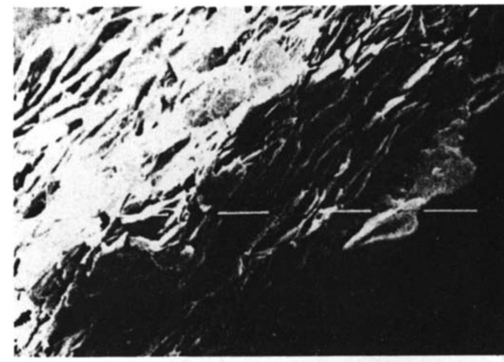

Figure 4 SEM pictures of a brocken Bi(2223) filament edge (upper) and filament surface (lower) of a Bi(2223)/AgMg2 tape (pressed).

3.4 *Transport currents*

Critical currents were measured by means of a standard four probe method. With two intermediate pressing steps during the final annealing at 841 °C, 30 + 50 + 50 h and performing the tape deformation scheme A we obtained up to J_c = 40 000 A cm^{-2} (77 K, 0 T). But the scattering was still unsatisfactory (see. fig. 5) with a strong contribution of changing filament cross section along the tape. If in a tape currents above 30 000 Acm^{-2} occured, all other samples usually carry at least 20 000 Acm^{-2}. When using the deformation scheme B, supressing crack formation through small deformation steps, we observed a very stable filament cross section area along the tape (see fig. 3) but changed annealing conditions were necessary. An optimization was found for reduced temperature of 835 °C and longer time 50 + 70 + 70 h with J_c up to 27 000 Acm^{-2}. We attribute this change to an enhanced filament densification and presumably reduced effective O_2 partial pressure in the filament. The J_c vs. B and T behaviour, not shown in this contribution, was similar to the results of other authors.

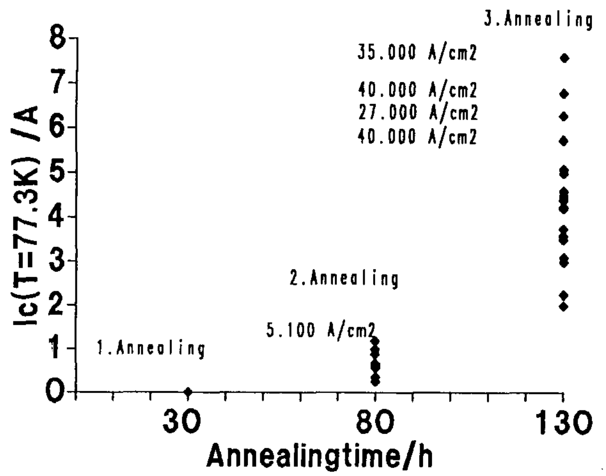

Figure 5 Variation of critical currents of Bi(2223)/Ag tapes with final annealing pressing treatment

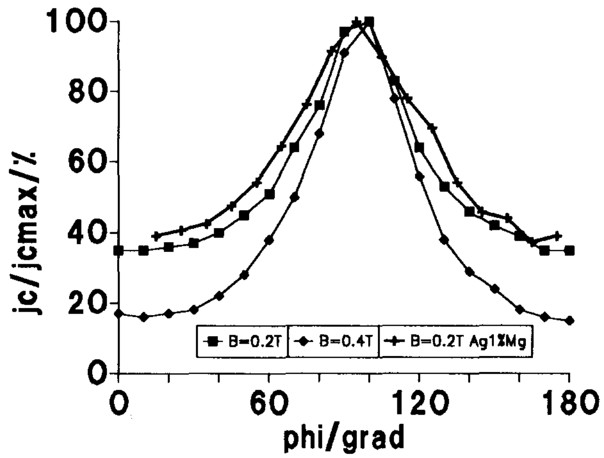

Figure 6 Orientation dependence of J_c for a Bi(2223)/Ag and Bi(2223)/AgMg1 tape in a magnetic field of 0.2 and 0.4 T (at 90°, B parallel tape surface).

For Bi(2223)/AgMg tapes, worked down after scheme B, we observed further decreased annealing temperatures scaling with the Mg content, being 836 °C for AgMg1 and 833 °C for AgMg2. In this case too the consequences of an enhanced filament density seems to be the reason, but also an influence of a changed oxygen permeability of the Ag-MgO composite cannot be excluded regarding the much smaller oxygen permeability of MgO.

Nevertheless transport currents (short pressed tapes) up to J_c = 22 000 Acm^{-2} (77 K, 0 T) for AgMg1 and J_c = 16 000 Acm^{-2} for AgMg2 sheathed tapes were found until now.

In tapes being deformed only by rolling, 8000-10 000 A cm^{-2} (77 K, 0 T) were measured for the first few samples, which is very promising for long length samples.

The dependence of J_c on the orientation of the tape in a magnetic field of B = 0.2 T and B = 0.4 T was measured (fig. 6). No significant change, except a small broadening of the maximum being in agreement with the texture analysis was observed using hardened Ag, indicating a similar grain alignment, texture and current path in these tapes.

4. Conclusions

We have obtained high critical current densites in Bi(2223)/Ag tapes but with still unsatisfactory reproducibility over a series of samples. The reason for this is the sum of nonoptimized sample properties as texture quality, microstructure, filament densification. and grain interconnection.

The use of internal oxidized AgMg as hardened sheath material was applied successfully. The enhanced hardness could partly withstand the heat treatment of the tape, stabilizes the tape composite, and is expected to have the potential for further improvements, especially through optimizing the internal oxidation process.

The improved mechanical tape properties and the observed J_c = 8000 - 10 000 Acm^{-2} in tapes deformed by rolling only, are very promising for further J_c enhancements. Especially the Mg additions obviously have no negative chemical influence on the filament. So the application of AgMg sheath was found fo be very suitable substituting pressing steps by rolling, being necessary for long tape lengths but modified tape preparation techniques need to be developed to combine the mechanical advantage of internal oxidized AgMg alloys with the high critical currents of pressed short samples, which is the central scope of our running activities.

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