Study of grain boundary properties in Ag-clad Bi$_2$Sr$_2$Ca$_2$Cu$_3$O$_x$ tapes by multi-phase electron backscatter diffraction analysis

A Koblishka-Veneva$^a$, M R Koblishka$^b$

$^a$ Institute of Functional Materials, Saarland University, P.O. Box 151150, D-66041 Saarbrücken, Germany
$^b$ Institute of Experimental Physics, Saarland University, P.O.Box 151150, D-66041 Saarbrücken, Germany

m.koblishka@mx.uni-saarland.de

Abstract. The properties of grain boundaries within Ag-clad (Pb,Bi)$_2$Sr$_2$Ca$_2$Cu$_3$O$_x$ (Bi-2223) tapes are studied by means of electron backscatter diffraction (EBSD). The achieved high image quality of the Kikuchi patterns enables multi-phase EBSD scans including Bi-2223, Bi$_2$Sr$_2$CaCu$_2$O$_x$ (Bi-2212), Bi$_2$Sr$_2$CuO$_x$ (Bi-2201), (Sr,Ca)$_2$Cu$_3$O$_y$ and Ag to be performed. For the EBSD scans a maximum spatial resolution of 30 nm was reached enabling a detailed orientation analysis. The nature of the grain boundaries is discussed on the base of the EBSD data. While the main orientation of the tape is in [0 0 1] direction, a large number of misorientations is detected. These misorientations are visualized using crystal direction (CD) maps. Furthermore, EBSD enables the spatially resolved mapping of the misorientation angles within each phase separately. The influence of these grain boundaries on the current transport properties is discussed.

1. Introduction

The texture achieved during the processing of silver-sheathed (Pb,Bi)$_2$Sr$_2$Ca$_2$Cu$_3$O$_x$ (Bi-2223) tapes is an essential parameter to obtain tapes with high critical currents. The core of monofilamentary Bi-2223 tape consists mainly of Bi-2223, but even the currently best tapes contain about 20% secondary phases [1,2]. Therefore, it is important to analyze the texture of the tapes using a spatially highly resolved technique, which further enables a multi-phase detection. The electron backscatter diffraction (EBSD) technique [3,4] provides here a unique tool for this task, and with the recent development to the EBSD technique concerning the image recording system, a high spatial resolution down to about 50 nm even on oxidic samples can be achieved [5,6]. In recent works, we have demonstrated the usefulness of the EBSD technique on various bulk high-$T_c$ samples with embedded nanoparticles and their interaction with the superconducting matrix. It is now natural to apply this technique also to the Ag-sheathed Bi-2223 tapes, where EBSD should contribute to the understanding of the grain orientation and the resulting grain boundaries.

In this contribution, we analyse the misorientations between the various grains in a monofilamentary Bi-2223 tape by means of EBSD, and discuss the influence of these grain boundaries on the possible current flow.
Figure 1. EBSD IQ map (a) and 5-phase map (b,c) of a Bi-2223 tape (section close to the Ag sheath). The rolling direction (RD) and transverse direction (TD) are indicated. (b,c) analyse the white frame shown in (a); additionally the EBSD-determined grain boundaries are shown.

2. Experimental procedure

Bi-2223 monofilamentary tapes were produced by the standard powder-in-tube method [7]. Monofilamentary Bi-2223 tapes were chosen for the easier surface polishing process, enabling a basic study of the microstructural properties of the Bi-2223 tapes. The critical current $I_c$ was measured to be 27 A (self-field, 77 K), and the critical current density $J_c$ was about 20 kA/cm$^2$. The critical current density of these monofilamentary tapes is not optimized, but this does not influence the basic conclusions of this paper.

A dedicated polishing procedure is required to achieve a high image quality enabling a multi-phase EBSD scan to be performed. For this study, the top surface of the silver-sheathed tape was carefully polished away using 2400 mesh grinding paper. For the surface finish, the tape was subsequently polished using diamond paste (6 μm, 3 μm, 1μm, 1/4 μm grain size), and then a final step using colloidal silica (Stuers OP-S) [8] was applied. Only ethanol was used for cleaning in order to avoid contact with water. This polishing procedure yields a rms roughness of about 5 nm [9], and, the superconducting properties are retained fully as confirmed by magneto-optic imaging performed on such samples [10].
The EBSD system consists of a FEI dual beam workstation (Strata DB 235) equipped with a TSL OIM analysis unit [11]. The Kikuchi patterns are generated at an acceleration voltage of 20 kV, and are recorded by means of a DigiView camera system, allowing a recording speed of the order of 0.1 s/pattern. The time may be slightly longer in the case of a multi-phase scan. To produce a crystallographic orientation map, the electron beam is scanned over a selected surface area and the resulting Kikuchi patterns are indexed and analysed automatically (i.e. the Kikuchi bands are detected by means of the software). An image quality (IQ) parameter and a confidence index (CI) is recorded for each such Kikuchi pattern. A detailed description of the measurement procedure can be found in Refs. [5,6].

3. Results and discussion

![Images of EBSD patterns and maps]

**Figure 2.** Inverse pole figure (IPF) maps in [0 0 1] direction (left) and in [1 0 0] direction (rolling direction) on the right side. The colour codes are explained in the stereographic triangles.

Figure 1 presents an image quality (IQ) map which resembles a backscattered electron image. The phase map illustrates the results of the 5-phase EBSD scan performed here. While the majority of the material is found to be Bi-2223 (91.7%), the other phases are present as well. Bi$_2$Sr$_2$CaCu$_2$O$_x$ (Bi-2212) and Bi$_2$Sr$_2$CuO$_x$ (Bi-2201) occur mainly as intergrowths to the Bi-2223 phase, and are often located at the grain boundaries of the Bi-2223 phase. The non-conducting (Sr,Ca)$_{14}$Cu$_{24}$O$_{41}$ particles,
which would be the candidates to provide flux pinning sites within the Bi-2223 grains, are located along the grain boundaries and, hence, are useless for flux pinning. The Ag particles are also located along the grain boundaries, where at least a good electrical contact to the neighboring grain may be provided.

Figure 2 presents the crystallographic orientations found in this section of tape in inverse pole figure (IPF) maps. The left map is in [0 0 1] direction, i.e. the c-axis spread around the sample normal is directly visualized. The dominant orientation is (0 0 1) as expected, but there is a clear misorientation as the colour is ranging between pink and orange. This will be more clear in Fig. 4. The right map gives the orientation in [1 0 0] direction corresponding to the rolling direction (RD). The colour now visualizes the misorientation in the rolling direction, which is also considerable.

A detail of the IPF map is presented in Fig. 3 for the Bi-2223 phase only; contributions of other phases are plotted in grey. Further, the respective unit cells are indicated. Here, it is visible that along large Bi-2223 grains, several small, but misoriented ones are formed. These small grains are acting quasi as a filler material. The resulting misorientation angles are mostly above 20°. As the big grain in the center is not oriented parallel to RD, it is itself an obstacle for the current flow parallel to RD. The boundary through this big grain is also remarkable, as the unit cells plotted are indicating a ‘V’-shape with a misorientation of about 30°.

![Figure 3](image3.png)

**Figure 3.** IPF map for the Bi-2223 phase only; other phases are plotted in grey. Further, the orientation of the unit cells are indicated. For the colour code in the stereographic triangle, see Fig. 2.

Figure 4 presents crystal direction (CD) maps. Here, the colour code is concentrated on 10° around the [0 0 1] direction in (a) and on 10° around the [1 0 0] direction in (b). This way of presenting the data
clearly reveals that only 22% of the Bi-2223 is oriented within this limit, whereas even 10% of the Bi-2223 grains have an orientation close to (1 0 0). Fully oriented grains do practically not exist. This implies that a current flowing through the tape parallel to RD has to face a large amount of obstacles formed by high-angle grain boundaries. Of course, in the present multifilamentary tapes, the achieved texture is much better as in the present sample, but the main problem of secondary phases within a filament still remains; the filaments are practically shells of Bi-2223 enclosing the secondary phases [1]. It will especially be a problem to control the intergrowths of Bi-2212 and Bi-2201, which are in principle of the same type, but have one or two CuO_2 planes less.

![Gray Scale Map Type: <none>](image1)

**Color Coded Map Type: Crystal Direction**

| Direction | Min | Max | Total | Partition |
|-----------|-----|-----|-------|-----------|
| 0°        | 1°  | 0°  | 0.221 | 0.224     |

**BSCCO 2223**

[001]

![Gray Scale Map Type: <none>](image2)

**Color Coded Map Type: Crystal Direction**

| Direction | Min | Max | Total | Partition |
|-----------|-----|-----|-------|-----------|
| 1°        | 0°  | 1°  | 0.107 | 0.107     |

**BSCCO 2223**

[100]

**Boundaries: <none>**

**Figure 4.** Crystal direction (CD) maps. The left map gives all grains within 10° of the [0 0 1] direction, and the right map all grains within 10° of the [1 0 0] direction. All other grains are plotted in white.

The map presented is Fig. 5 is a phase map (Bi-2223 indicated in red, other phases are black). Grain boundaries towards other phases are disregarded in this analysis. Boundary misorientations are shown between 1° and 5° (yellow), 5° and 10° (green) and between 10° and 30° (blue). Regarding the boundaries only found within the Bi-2223 phase, many small angle boundaries (misorientations smaller than > 5° shown in yellow, ~20%) are detected, but the majority of the boundaries is found between 10° and 30° (31.5%) with a total length of 6.38 mm, while the sum of both small-angle boundaries is 6.2 mm. Note that also the boundary within the large grain as discussed in Fig. 3 falls
into this limit. This figure confirms the results of Fig. 4, indicating that the misorientations within the Bi-2223 phase are not so small as one might expect from e.g., the brick-wall model for the grains within Bi-2223 tapes. The situation corresponds is more similar to the railway-switch model [12], but essentially, much more complex due to the presence of the secondary phase particles. Therefore, the grain boundaries in Bi-2223 tapes are serious obstacles for the current flow, not only due to the high misorientation angles, but also due to the secondary phase particles which can be either insulating ((Sr,Ca)14Cu24O81) or electrically conducting (Ag). Especially the first case is a bad one, as then the entire area of the particle is posing a border for the current flow.

![Misorientation map of Bi-2223 phase](image)

**Figure 5.** Misorientations within the Bi-2223 phase. The underlying map is a phase map (Bi-2223 drawn in red).

If one regards the same section of tape with a tolerance of 20° around the [0 0 1] direction, then 61.7% of the Bi-2223 grains fall in this limit. Here, we are now closer to obtain at least a good current path through the sample, but still, a fully superconducting current path is not available. This is the reason for the strong field dependence of the transport currents as observed in the Bi-2223 tapes. In current state-of-the-art multifilamentary tapes (see e.g. Ref. [1]), the amount of grains not oriented in the RD direction is considerably reduced, but the intrinsic tendency of Bi-2223 grains to form subgrains and intergrowths cannot easily be controlled during the tape processing. In order to increase the amount of intragranular current density, one should find a way to employ the existing secondary phases as flux
pinning sites within the grains, but it is an essential task to keep the grain boundaries free from these obstacles. One possibility to improve the flux pinning could be the addition of specific nanoparticles; another approach could be the addition of a oxide to the precursor powder which influences the formation of the (Sr,Ca)$_{14}$Cu$_{25}$O$_{41}$ particles.

In summary, we have presented an EBSD analysis of a monofilamentary Bi-2223 tape. EBSD scans could be performed with 5 phases, and a resolution of ~70 nm. The phase map reveals that the secondary phases are located mainly along the grain boundaries. The detected misorientation angles within the Bi-2223 phase are found to be mainly of the order of 10° to 30°.

**Acknowledgements** We would like to thank T. Qu and Z. Han (Tsinghua University, Beijing) for the tape samples. This work was performed with financial support of DFG project no. MU959/12.

**References**
[1] Holesinger T G, Kennison J A, Liao S, Yuan Y, Jiang J, Cai X Y, Hellstrom E E, Larbalestier D C, Baurceanu R M, Maroni V A and Huang Y 2005 *IEEE Trans. Appl. Supercond.* **15** 2514
[2] Yao P, Zeng R, Liu H K and Dou S X 2000 *Physica C* **337** 174
[3] Baba-Kishi K Z 2002 *J. Mat. Sci.* **37** 1715
[4] Humphreys F J 2004 *Scripta Materialia* **51** 771
[5] Koblishka-Veneva A, Koblishka M R, Hari Babu N, Cardwell D A, Shlyk L and Krabbes G 2006 *Supercond. Sci. Technol.* **19** S562
[6] Koblishka-Veneva A, Koblishka M R, Ogasawara K, Mücklich F and Murakami M, J. *Superconductivity* **18** (2005) 469
[7] Qu T M, Han Z and Flükiger R 2006 *Physica C* **444** 71
[8] OP-S, Struers Co., Denmark
[9] Koblishka-Veneva A and Koblishka M R 2004 in: Magneto-Optical Imaging, ed. T. H. Johansen, D. V. Shantsev, Kluwer Acad. Press p. 242
[10] Koblishka M R, Wang W G, Seif B, Skov-Hansen P, Vase P and Andersen N H 2001 *IEEE Trans. Appl. Supercond.* **11** 3242
[11] Orientation Imaging Microscopy software version V4.0, user manual, TexSEM Laboratories (TSL), Draper, UT.
[12] Hensel B, Grasso G and Flükiger R 1995 *Phys. Rev. B* **51** 15456