AFM investigations of the surface morphology of buffer layers for all-chemical solution Coated Conductors

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Abstract. Crack free layers of $\text{La}_2\text{Zr}_2\text{O}_7$ (LZO) and $\text{CeO}_2$ (CeO) were deposited by chemical solution deposition (CSD) on cube textured Ni 5 at%W substrates. The films were heat treated between 900° and 1020°C, respectively. Besides atomic force microscopy (AFM), several analytical methods were applied to analyse the texture quality of the single buffer layers, enabling us to determine the morphology, microstructure and texture of the films. By X-ray diffraction and EBSD the buffer layers were found to be [100] oriented and strongly biaxially textured. The average grain size of the films was determined to be between 100 and 300 nm, being much smaller than the Ni grain size of 40 µm. In this paper we focus on the investigation of the surface morphology. Various samples of different buffer layers were scanned.

1. Introduction

The production of 2G HTS Coated Conductors by an all chemical solution deposition (CSD) process is considered to be one of the most promising technologies concerning the cost / performance ratio of long length cable and tapes. The conductor architecture is based on a biaxial textured metal tape - Ni or Ni-alloys - as substrate [1], a multilayered LZO (Lanthaniumzirconate) / CeO$_2$ (Ceriumoxide) buffer system or multi-STO (Strontiumtitanatate) buffer layers and superconducting YBCO-layers. Both buffer and YBCO layers were deposited by a low-cost CSD process with a throughput of up to 7 m/h for buffer layers and 12 m/h for the YBCO thin-film.

The deposition of thin oxide layers on top of biaxial textured tapes requires detailed knowledge of the surface topology of the Ni 5at%W-substrate as well as the film growth mechanism for the following layers, as multiple film deposition by CSD leads to an addition of surface defects. A dense, crack-free, smooth and planar buffer is essential for superconducting films with good electrical attributes, which is hard to achieve on long length tape. Special attention is drawn to the LZO and CeO$_2$ buffer growth and morphology.

In this work the conclusions are based on atomic force microscopy (AFM) investigation of the uncovered Ni/W-tape and several continuously produced oxide thin layers. We use Ni-W substrate, because of its good perovskite texture, that is given to the deposited thin films via homoepitaxy.

After the heat treatment XRD and EBSD measurements exhibit a strong in-plane and out-of-plane texture. To reach a high quality YBCO layer not only texture plays an important role, but also the topology of the surface the YBCO-layer is deposited on. So we laid our focus on AFM measurements.

The analysis were carried out in cooperation with the Bonn-Rhein-Sieg University of Applied Sciences.
The AFM micrographs reveal important details on surface quality and properties [2, 3]. For this examples are, single buffer layer grain sizes and surface topography as well as buffer growth behavior over the originally some 18 nm deep substrate grain boundaries. With well grown buffers with minimum misorientations, the installed All-CSD-process seems to be the most prosperous and encouraging facility for the future.

2. Experimental details

For the preparation of All-CSD coated conductors it is necessary to design precursor solutions with adequate wetting and drying behavior for the deposition of high-quality thin films. Composition of the precursor solutions was closely analyzed, as well as viscosity, water content and the wetting angle of contact.

The composition was analyzed by ICP-EOP Spectro Genesis. Viscosity was controlled by rotary viscometry and was carried out in a Brookfield (DV-III) viscosimeter. The water content was determined by Karl-Fischer titration. The investigation on the wetting angle was carried out in a home-made gadget to measure the wetting angle via optical microscopy.

2.1. Substrate preparation. To get a good wettability of the metal substrates, the NiW- tapes at first were degreased in acetone and isopropanol in an ultrasonic bath. Short samples are prepared in tumblers in the ultrasonic bath for 10 min per organic solvent. Longer samples up to 10 m and longer are prepared by continuous cleaning through a dipcoating-like reel-to-reel cleaning process at speeds of 2.5 m/h.

Before and after deposition of each heat treated buffer layer, the samples underwent an ultrasonically cleaning procedure with i-propanol.

2.2. Coating of the layers and furnace reaction. As crystallographically oriented templates cube textured Ni 5at% W tapes were used. The cleaned metal stripes were dip-coated with varying withdrawal speeds on a continuous dipcoating device. The speed of the coating process and the precursor viscosity are the main influencing factors for the thickness of the resulting ceramic layer. As varying layer thicknesses require slightly different heat treatments, it is necessary to accommodate the dwell times to enable the solvent to react and to volatilize out of the layer.

The samples produced on the continuous coating device were dried by throughput of two ovens heated to 60 and 100 °C. After the drying procedure the LZO and CeO precursor films were wound on a quartz tube with 10 cm in diameter, followed by a heat-treatment under forming gas atmosphere of N₂ 5% H₂ between 900 and 1020 °C in a batch furnace.
2.3. Properties of the buffer layers. The surface roughness of the deposited layers was characterized with an Atomic Force Microscope (AFM) (Veeco CPII) The nominal cantilever force was 10 nN and scanning area 10 µm². EBSD investigations were carried out on a Jeol jsm 6400 F SEM. Figure 1 shows, that most of the grain boundaries lay below 10° tilting angle and that the texture component has a quite high value with 97.9 %.

![Figure 1. Grain boundaries and texture component of LZO analysed via EBSD](image1)

The XRD texture analyses were carried out on a Brukker D4 endeavour with theta-2theta scans. Figure 2 presents a LZO single layer with a thickness of 140 nm and a strong (001) cubic texture.

![Figure 2. X-ray measurement of a single LZO layer of 140 nm thickness and (001) texture. The peak height is about 4500 counts and the peak angle lays at 33.3°.](image2)

3. Results and discussion

AFM investigations were carried out on single, double and triple coated ceramic buffer films. The scanning area of the films was from 1x1 to 40x40µm². An AFM line scan on a 40x40 µm² area of the uncoated Ni/W surface shows an average elevation of 140 nm and a maximum height difference of 60 nm. The RMS roughness of the scanned area is 8.404 nm. In Fig. 3 a typical grain boundary between two Ni crystallites is marked. According to the line scan it has a depth of 20 nm and a spatial width of 800 to 1200 nm.
Figure 3. AFM generated profile measurement on a blank NiW-substrate. On closer examination is the depth and width of a grain boundary and the average height as well as the RMS roughness. Figs. 4 a) and b) represent the surface topology of different Ni/W-tapes covered with a single LZO layer. After the deposition the RMS roughness are between 8 and 12 nm for the 10x10 µm² areas. The biggest differences in height were detected between two crystallites, as close to the deep boundary between to grains a region is situated with a bead parallel to the boundary. The height of this bead explicitly above the area forming the flat crystallite plain, resulting in a problem for the conducting properties of thin films, as a relatively great difference in height has to be surmounted within a few micrometers.

Figure 4. Topography of two different LZO samples as 2D and 3D images. The upper row represents the single deposited LZO layer and in the second row there are images of the double coated LZO layer. Images a) and b) have scanning areas of 10x10 µm² with average RMS values of 8 to 12 nm and c) an d) have scanning areas of 1*1 µm² with an RMS value of 2 nm. The 1x1 µm-scan in fig 4 c reveals that the first LZO layer generates a relatively rough topography with large crystallites protuding out of the coating volume. With the second LZO layer represented in fig. 4 d) the vacant capacity between the crystallites is filled, resulting in decreased relative roughness: Related to a film thickness of 140 nm after the first deposition, the RMS roughness of 8 nm corresponds to 5.7 % of the film volume. After the second deposition the RMS roughness rises to 12 nm, which corresponds to 4.3 % related to a layer thickness of 280 nm.

After the deposition of LZO a third layer consisting of CeO₂ is superimposed, which is necessary to ensure the growth of high-quality films as the lattice constant of cerium oxide matches slightly better to the lattice constant of YBCO than LZO does.

The investigation of the CeO₂-surface reveals a smooth an homogenous surface. The ceria crystallites only rise a little out of the surface, resulting in a low RMS-value of 8 nm and a low height difference in the scanned area. While the average RMS value contains the corrugation and grain
boundaries of the substrate we selected a representative area of 2.5*2.5 µm² in size. There the average height is 2.7 to 3.3 nm, with RMS values of about 1.1 nm, as can be seen in Figure 5.

Both scanning areas contain grain boundaries underneath the deposited buffer films. It is clearly visible that the sharp grain boundaries detected on the Ni/W-substrate surface were filled buffer layer oxides during the dip-coating process resulting in a wave-like surface structure of the buffered tape without sharp grain boundaries.

Figure 5. Topography of two CeO samples, as 2D and 3D images with scanning-areas of 10*10 µm². The upper row represents a quite good CeO-film with a RMS value of 10 nm and the second row shows an improved CeO-film with a quite small maximum amplitude of 38.6 nm and a roughness of 8 nm. The fourth image represents roughness analyses of scanning areas of 2.5*2.5 µm² in size. These areas were cut out of the 10*10 µm² areas.

4. Conclusions

High quality CeO and LZO buffer layers were chemically deposited on cube textured NiW-substrates with very good c-axis alignment of the unit cells and a smooth and crack free surface.

At present we understand the established microstructure as a result of a growth mechanism during the annealing of the film. The quality of the underlying NiW-substrate is essential for the growth mechanism because it provides the biaxial texture that is transferred to the LZO and CeO₂ buffer layer. To reach epitaxial growth the substrate must be atomically planar and the film has to grow in a layer-by-layer mode yielding the same texture and grain size in the film as in the substrate. The texture is transferred very well, but obviously this is not the case for the grain sizes because the LZO grains have dimensions about 100 nm and the Ni-substrate has grain sizes about 40 µm [5].

All layers had very fine (00l) textures, although the roughness within one grain is about 1 nm, after the final CeO₂-film, while the overall roughness is about 10 nm. So the roughness is reduced to facilitate the occurrence of epitaxial growth and to transfer the texture through all buffers into the superconducting YBCO film. Moreover several coating steps reduce the depth of grain boundaries and fill the vacant volumes that are represented through the maximum amplitude of the AFM-measurements. This provides an excellent long length all-solution buffer which is ready to be coated in a CSD-trifluoroacetates-YBCO process.
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