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Non-magnetic Substrates for Low Cost YBCO Coated Conductors

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Abstract—CuNiAl and NiCr alloys were made and processed to non-magnetic, bi-axially textured substrates. Epitaxial YSZ and YBCO were deposited by pulsed laser deposition on the CuNiAl. Samples were cross sectioned and investigated with TEM. It appears that Al forms a thin oxide layer at the CuNiAl surface that acts as a template for the subsequent YSZ layer. The 1.2 μm YBCO layer had a current density of 0.16MA/ at 75K and self field. On the NiCr substrates an Y_2O_3 seed layer was deposited with E-beam evaporation, followed by RF sputter deposition of YSZ and a CeO_2 cap layer. These substrates were coated with a 0.4 μm YBCO layer using a trifluoroacetate process, a very cost effective metal-organic solution process. These samples carried critical currents exceeding 0.7MA/cm² at 77K, self field.

Index Terms—YBaCuO Coated Conductor, Superconductor

I. INTRODUCTION

Development efforts for $YBa_2Cu_3O_{7-x}$ Coated Conductors, or YBCO CCs, have greatly accelerated in recent years. The most obvious reason for this is the excellent self field and in-field performance of the conductor at 50-77K, in particular when compared with the now commercially produced $Bi_2Sr_2Ca_2Cu_3O_x$ conductor. Just as important is the potential of YBCO CC to produce HTS conductors at a much lower cost/kAm than is possible with Bi-2223 conductors, despite the process complexity to produce the YBCO with a high degree of bi-axial texture. Several processes now produce substrates with bi-axially textured buffer layers. The RABITS process, used in this work, is based on epitaxial buffer layer deposition on deformation-textured metal substrates [1-3]. Ion beam-assisted and inclined substrate deposition are based on texturing of the buffer layer during deposition on an untextured substrate [4-6]. For the deposition of high quality YBCO layers a variety of processes are used. LANL demonstrated with pulsed laser ablated YBCO on IBAD buffer layers continuous I_c

exceeding 130A over 1m lengths [6] and, with an adapted deposition method, critical currents of 500A/cm at 75K, self field, in short samples [7]. Electron-beam deposited YBCO based on BaF_2 has demonstrated 200A/cm width [8]. Both low cost and high Ic performance need to be realized in YBCO CC to effectively compete against present and future $Bi_2Sr_2Ca_2Cu_3O_x$ conductors. This makes the development of cost-effective deposition processes quite important. AMSC is therefore in particularly interested in those processing steps where metal-organic deposition (MOD) processes based on cheap solutions can produce buffer or YBCO films with performance at par with similar films obtained through vacuum processing. For example, high J_c values have been obtained at AMSC using deformation textured substrates, vapor deposited epitaxial buffer layers, and a trifluoroacetate (TFA) based YBCO solution process. In 0.4μm thick films J_c reached 1.9 MA/cm² at 77K, SF [2], while 1μm thick films have been made with 1.0 MA/cm² with this TFA YBCO process [3]. High J_c values have also been obtained when the seed layer part of the buffer layer was deposited with a MOD process as well [3]. The substrates that were used were bi-axially textured Ni foils, with a Full Width Half Max (FWHM) value of 8-9°. Ni is considered a good demonstration metal, because it can be made well-textured with a surface that is compatible with epitaxial oxide deposition processes, whether these are physical vapor deposition processes or MOD processes such as have been used for the deposition of Gd_2O_3 seed layers. It has a tenacious bond with NiO, formed during YBCO processing, which reduces the chance of spallation at the Ni interface. However, Ni is ferro-magnetic which is problematic in many potential applications of YBCO CC. It is also quite weak when annealed to induce the cube texture. The use of strong, non-magnetic, oxidation resistant substrates with a sharp cube texture is therefore essential. The alloying of Ni to make it non-magnetic is relatively straightforward: in most cases the Curie decreases more or less linearly with increasing concentration of the alloying component, while the effect of the component is more or less proportional with its valence state. Cu (considered 1+) is the least effective element, but can alloy with Ni over the entire compositional range, and its low cost at high purity. Elements such as Al and Cr are three times more effective. Other elements such as W [9] and V [10] have been used as well. Here we report on the use of CuNiAl and NiCr alloys. The CuNiAl alloy has a small amount of Al to prevent

oxidation of the CuNi alloy which otherwise can oxidize nearly as fast as plain Cu. Note that small amounts of Al are quite effective, but that larger amounts can easily introduce a Ni(Cu)$_3$Al gamma prime component at 400-800°C . At temperatures below 900°C the external oxidation of an alloy such as CuNiAl was expected to form gamma alumina, which is cubic and has a lattice constant of 0.79 nm. The formation of this surface oxide was therefore expected to be compatible with the deposition of buffer layers such as CeO$_2$ or YSZ as the lattice match is quite good. Ni-13Cr is different, as the hexagonal Cr$_2$O$_3$ would most likely be a poor template for buffer deposition. In the NiCr case processing conditions were selected that would avoid premature oxidation of the substrate. ORNL has demonstrated on Ni-13Cr a J$_c$ of 1.4 MA/cm^2 at 77K, SF, using the e-beam BaF$_2$ process [8]. We were in particular interested to test the compatibility of NiCr with the solution-based trifluoroacetate YBCO process.

II. EXPERIMENTAL APPROACH

CuNiAl alloys with 48wt%Ni and 1-1.5% Al, and Ni with 13wt%Cr, were made by casting. The casts were deformed to tapes using a thermo-mechanical process. The CuNiAl substrates were 150 μm, the NiCr substrates 75 μm thick. The substrates were texture annealed using vacuum processing. The texture was analyzed using a Rigaku XRD system. In all cases the intensity of the (111) reflections in θ–2θ scans were well below 0.5% of the intensity of the (200) reflections. A Siemens (now Brucker-Axis) XRD unit with GADDS detector was used to make (111) and (200) pole figures. These were used to determine the percentages of the various substrate components: percentage with a cube texture, percentage of twins (these do not show up in θ–2θ scans) and percentage of grains with random or undesired texture. FWHM values were determined from the pole figures as well. The FWHM values are a statistical average over a relatively large area encompassing about 8-10,000 grains. The substrates were then prepared for buffer deposition. The CuNiAl was surface-cleaned to remove a thin alumina layer that forms during the texture anneal. For the PLD deposited buffer layers samples were polished to have a well-defined surface. The polished CuNiAl samples were coated with YSZ at LANL using PLD. The sample was heated in vacuum, held at temperature for about 15 minutes, and coated with about 0.7 μm YSZ. Subsequently the substrate was coated with about 1.2 μm YBCO by PLD. The texture annealed NiCr substrates underwent a separate processing step to make their surface compatible with the buffer deposition process. A 20nm thick yttria seed layer was deposited by E-beam deposition. A 0.5 μm thick YSZ and 20nm ceria cap layer were deposited by RF sputtering. Buffer layers on the NiCr were inspected after seed layer deposition and after YSZ and ceria deposition with pole figures and HRSEM. The buffered NiCr sample was then coated with a 0.4 μm thick YBCO layer using the TFA YBCO solution process, similar as described before [2].

The critical current density was measured on bridged
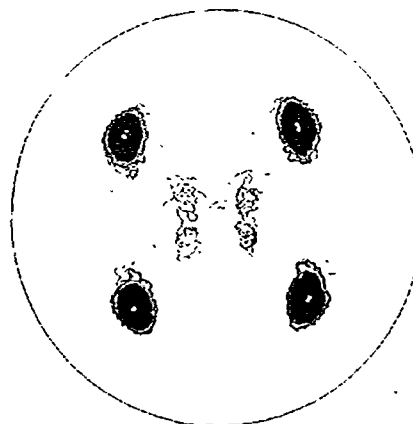
samples at 77K

Fig. 1 (111) pole figure of CuNiAl substrate, enhanced intensity

For the measurement of the thermal expansion some of the cast material was processed to rod. This approach is suitable as the thermal expansion in fcc metals does not depend on lattice direction, and the results from untextured rods are directly applicable to cube textured foils. For comparison a pure Ni rod was measured as well.

III. RESULTS

Fig. 1 and 2 show the pole figures of CuNiAl and NiCr, both at enhanced intensity (around 40-70x) to show background reflections. The CuNiAl has a slight tendency to form twins (the four reflections close to the center). These were observed with light microscopy to be short and relatively wide and located at grain boundaries, as is typical for annealing twins. Their percentage was 1-1.5%. The NiCr substrates were essentially free of twins. The FWHM values were 8-9° for the CuNiAl, and 7-8° for the NiCr. Fig. 3 shows the phi scan of the PLD deposited YBCO layer on YSZ/CuNiAl. The FWHM value is 17.8°. The YSZ has a lower FWHM of 13°. The 1.2 μm thick YBCO layer had a J$_c$ of 160kA/cm^2 at 75K.
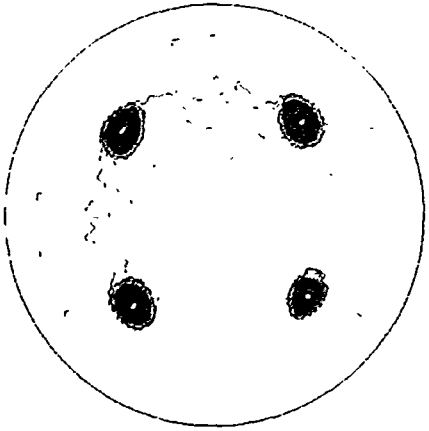
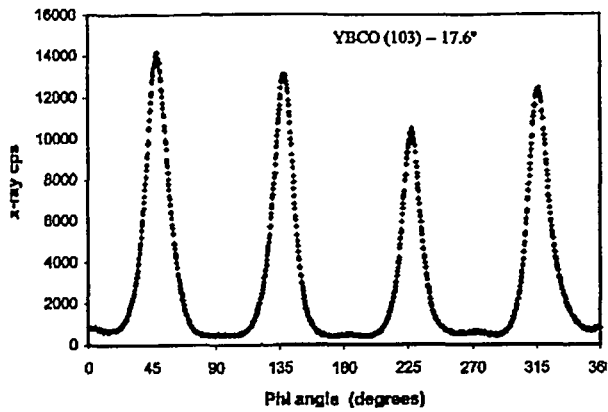Fig. 2 (111) pole figure of NiCr substrate, enhanced intensity

Fig. 3 Phi scan of pulsed laser deposited YBCO on YSZ/CuNiAl

Cross sectional TEM of a YSZ coated CuNiAl sample shows a thin, 20nm thick layer as can be seen in Figures 4 and 5. Chemical analysis of the layer identified it as alumina. Electron diffraction shows very weak reflections that appear to indicate gamma alumina, as shown in Fig. 5.

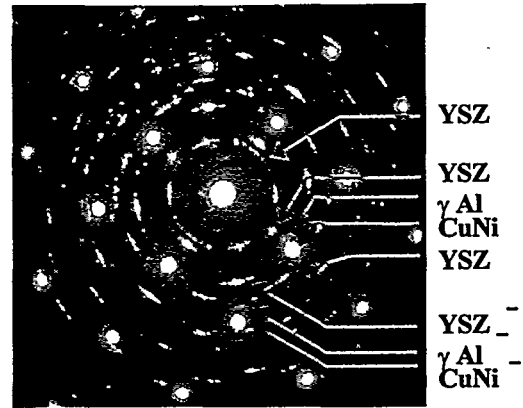Fig. 4 TEM cross section of YSZ/CuNiAl interface, showing thin alumina layer at interface

Fig. 5 Selected area diffraction of YSZ/CuNiAl interface

The usual 45° rotation of the YSZ lattice when deposited on an fcc metal such as Ni did not occur here: the YSZ has a cube-on-cube orientation on the CuNiAl substrate.

The $CeO_2/YSZ/Y_2O_3/Ni$-13Cr stack (E-beam yttria seed layer, sputtered YSZ and ceria cap layer) showed a much sharper texture for the buffer layer sequence. Fig. 6 shows the (111) pole figures of the ceria cap layer of this sample with FWHM values of 6-9°. Two samples that were coated with a 0.4 μm TFA YBCO layer both showed J_c values exceeding 0.7MA/cm² at 77K, SF. No a-axis formation was evident from the XRD pattern, which was very clean as can be seen in Fig. 7.

The coefficient of thermal expansion (CTE) of CuNiAl was measured to be 13.8x10⁻⁶/K at room temperature, while the cte of Ni was very close to that of NiCr, at 12.6x10⁻⁶/K at RT. At higher temperatures the CTE for NiCr and Ni diverged somewhat.

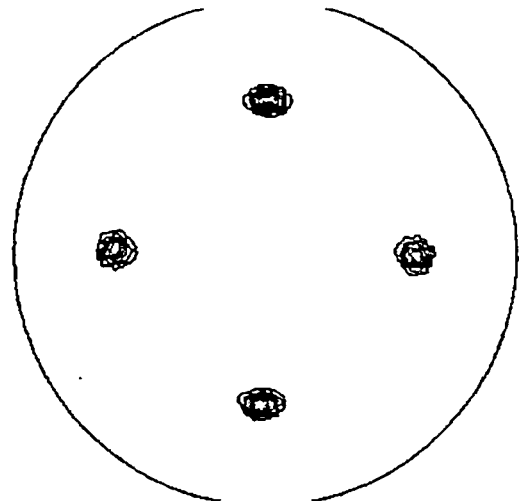Fig. 6 (111) pole figure of CeO_2 on YSZ/Y2)3/Ni-13Cr
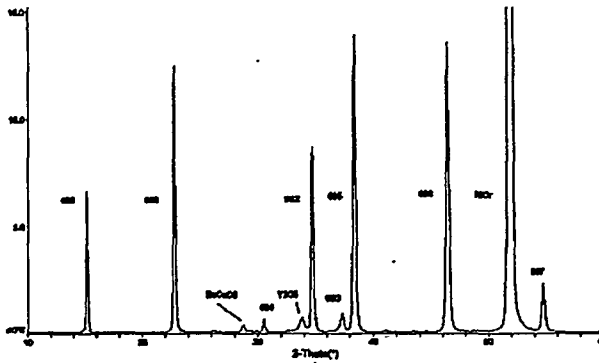
Fig. 8 XRD scan for trifluoroacetate solution derived 0.4 μm thick YBCO layer on CeO₂/YSZ/Y₂O₃/NiCr substrate

Note that the CTE for Ni is slightly lower than generally quoted in the literature, which might be due to differences in purity, and the fact that some quoted CTE values are averaged over the 20-100°C temperature range.

IV. DISCUSSION

The most obvious difference between the two substrates is the presence of a strong oxide former in CuNiAl. It is not clear when exactly the alumina layer formed: before or after buffer deposition. Gamma alumina is hard to detect with XRD as it has a defect spinel structure, in which 21.3 Al atoms are randomly distributed over 16 octahedral and 8 tetrahedral sites. However, the post-deposition scenario does not seem likely as the cube-on-cube orientation of a YSZ layer on a clean, metallic CuNiAl surface would cause an unusual amount of lattice strain, much more than a 45° in-plane orientated YSZ layer would. The pre-deposition formation of a thin interface oxide such as cubic gamma alumina is comparable with the pulsed laser deposited YSZ on Ni, as was recently demonstrated by Lee et.al. Here too a thin oxide layer, in this case (100) NiO, is expected to facilitate the cube-on-cube deposition. The reported J_c value of 0.5 MA/cm² at 77K, in a very thin PLD YBCO layer (250 nm) was considered high for a single buffer layer but lower than normal due to a certain degree of 45° mis-orientation of the YBCO film, as is also the case for the YBCO/YSZ/CuNiAl substrate in this work. The experiment with CuNiAl also demonstrated that substrates with coefficients of thermal expansion higher than that of Ni can still be a viable choice. In this case the difference is about 1.2x10⁻⁶/K, which will introduce an additional compressive strain on both buffer and YBCO layer of around 0.1%, assuming that the substrate does not yield.

The sputter deposited CeO₂/YSZ/CeO₂/CuNiAl yields a much better overall texture, with no deterioration of the texture with increasing buffer layer thickness. The buffered

Ni-13Cr samples showed the best texture of all, with a FWHM value of 6-9° being very close to that of the substrate. J_c results are lower than for corresponding Ni based samples, most likely due to a somewhat higher surface roughness of the alloy substrate.

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