Rotatable magnetron sputtering of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin films on single crystal substrates

I. De Roeck^a, G. De Winter^a, R. Van Paemel^f, R. De Gryse^a, J. Denul^b, I. Van Driessche^c, E. Bruneef, S. Hoste^c, E. Georgiopoulos^d, C. Andreouli^e, A. Tsetsekou^f

^aGhent University, Department of Solid State Sciences, Krijgslaan 281/S1, 9000 Gent, Belgium; ^bBekaert, Bekaertweg 2/6030, 8550 Zwevegem; ^cGhent University, Dep. Of Inorg. And Phys. Chemistry, Krijgslaan 281/S3, 9000 Gent, Belgium; ^dDept. of Mineral Resources Engineering, Technical Univ. Crete, University Campus, 73100 Chania, Greece; ^eCERECO S.A., 72nd km of Athens-Lamia Ntl Road, P.O.Box 146, 34100 Chalkida, Greece; ^fNational Techn. Univ. of Athens, Dept. of Mining Engineering and Metallurgy, Iroon Polytechniou 9, 157 80, Athens, Greece.

Abstract - The development of sputter deposition of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO) from rotatable targets is an important step in the realisation of a large scale deposition process for YBCO coated conductor. Rotatable magnetron sputter sources are standard equipment in large scale sputter industry and the power input in a rotatable magnetron can be much higher compared to a planar magnetron with the same race track area. This results in improved discharge characteristics and increased deposition speed. For this work a flame sprayed and a plasma sprayed cylindrical YBCO target was used to deposit YBCO thin films on single crystal MgO. C-axis oriented YBCO thin films were obtained at lower pressure than is normally used for YBCO sputter deposition, resulting in a higher deposition speed. The influence of deposition parameters (such as sputter pressure, oxygen partial pressure, substrate temperature, ...) on the properties of the YBCO layer was investigated and the possibilities for large scale deposition were explored.

Keywords: large scale, rotatable magnetron, YBCO deposition, single crystal

I. Introduction

Magnetron sputtering is one of the most easily scalable techniques to develop $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO) coated conductor, since it is used in industrial environments for web coating of meters wide and kilometres long substrate foil. Several problems encountered with sputter deposition of YBCO are related to the fact that the input power and the discharge voltage are too low. The use of a rotatable sputter

magnetron with cylindrical YBCO target for the sputter deposition of YBCO has multiple benefits. First of all, the power input can be considerably higher than for a non-moving target with the same racetrack area, because of better heat dissipation. Consequently the deposition speed can be higher and the discharge characteristics are improved. The material usage of the target is also much more efficient in the case of sputtering from a rotatable target. We already demonstrated that

with rotatable magnetron sputtering c-axis oriented YBCO can be obtained on Ytria Stabilized Zirconia (YSZ) buffered hastelloy tape [1]. In this work we will further investigate the sputter deposition from YBCO rotatable targets and optimise the sputter conditions of YBCO on single crystal MgO substrates.

II. Experimental procedure

The used rotatable sputter magnetron has been described elsewhere [1]. For this series of experiments two different YBCO targets were used. Both had a length of 32 cm, a diameter of 15 cm and a stoichiometric 1:2:3 composition. The first one is a flame sprayed YBCO target with Ag addition and no post-annealing. The other one is a plasma sprayed YBCO target without Ag addition, but it was annealed. Further details have been described elsewhere [2][3][4]. For the YBCO deposition, the chamber was evacuated to a base pressure of below 10^{-3} Pa. Ar and O₂ was admitted up to a total sputter pressure of 2 - 5 Pa. MgO (100) was used as substrate at temperatures between 630°C and 720°C during deposition and cooled down in $8 \cdot 10^{-4}$ Pa oxygen after deposition. The orientation and the c-axis parameter of the thin film were analysed by X-ray diffraction (XRD) patterns and the critical temperature (T_c) was determined from the resistance-temperature curve measured with a Variable Temperature Cryostat from Cryogenic.

III. Results and discussion

a. Influence of sputter parameters

Fig. 1 shows four XRD $\theta/2\theta$ spectra for samples deposited respectively at 630°C, 655°C, 680°C and 720°C using a plasma

sprayed target. The sputter pressure was 5 Pa, the sputter power 2 kW and the Ar/O₂ ratio was 3/1. Layers deposited at 680°C and 720°C mainly consist of c-axis oriented YBCO. A axis peaks become visible for the layer deposited at 655°C and for deposition at 630°C the YBCO peaks are very small with (103) as main orientation. Deposition at 680°C gives the best result : no green phase Y₂BaCuO₅, no a-axis peaks, but for a substrate temperature of 720°C, decomposition peaks were already observed, as well as some (103) orientation and green phase peaks. Table I gives the deposition conditions and results for three sets of two samples each. The first series shows the influence of the used target, in the second series a sample with post-oxidation procedure was compared to a sample without post-oxidation procedure and in the third series the sputter power was increased from 1.2 kW to 2 kW. The main conclusion from XRD $\theta/2\theta$ spectra of these layers is that these parameters don't seem to have any significant influence. From comparison of the results from the two types of targets it seems that the plasma sprayed target gives more green phase in the layer.

b. Rotatable YBCO magnetron with additional Cu-sputtering

Although the deposited YBCO layers all showed preferential c-axis orientation in XRD $\theta/2\theta$, the c-axis parameter was always higher than it should be (ranging from 1.181 nm to 1.191 nm, while the correct c-axis parameter is 1.168 nm). The authors believe that this could be due to a Cu- deficiency in the layers, which was also confirmed by EDX measurements. Therefore an additional Cu sputter magnetron was installed in the chamber to compensate for the Cu-deficiency in the layers. Similar

experiments examining the influence of co-sputtering of Cu for YBCO deposition on YSZ buffered Hastelloy tape have been reported earlier [1]. Fig. 2 shows the XRD $\theta/2\theta$ spectrum of an YBCO layer on MgO, deposited under the conditions summarized in table II. Under these conditions a volume deposition speed of about $0.66 \mu\text{m.cm}^2/\text{min}$ was achieved. It is clear that there are two c -axis oriented YBCO phases in the layer ($c = 1.1708 \text{ nm}$ and $c = 1.1917 \text{ nm}$) together with some (103) oriented YBCO, CuO and maybe $\text{Cu}_2\text{Y}_2\text{O}_5$. There are also some decomposition peaks visible, maybe indicating a slightly too high deposition temperature. The resistivity measurement (Fig. 3) shows a high $T_{c,\text{onset}}$ (88K), but the transition is very broad and zero resistance is only reached at 68K. This observation can be explained by the presence of two YBCO phases : the YBCO phase with the lowest c -axis determines $T_{c,\text{onset}}$ but a full transition can only be achieved when the second YBCO phase also becomes superconducting. If the amount of the YBCO phase with the correct c -axis could be increased above the percolation limit, a full transition would be achieved at higher temperatures. The higher c -axis value and the lower T_c of the second YBCO phase could be caused by the relatively low oxygen partial pressure during deposition [5]. Based on the antisited disorder model [5][6] this could be explained by the fact that at low oxygen partial pressure the Y site is apt to be substituted by a Ba atom. For layers deposited at low oxygen partial pressure Park et al. [5] also observe an XRD peak that could be attributed to $\text{Cu}_2\text{Y}_2\text{O}_5$, which could be related to the Y-Ba substitution leaving an excess Y and Cu if the Ba is consumed for YBCO formation. Since it is not

possible to achieve a scalable process at higher pressure, because of reduction of deposition speed, the main possibility for avoiding the Y-Ba substitution will be to optimise the composition without changing the pressure dramatically [5].

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References

- [1] J. Denul, G. De Winter, R. De Gryse, , IEEE Trans. Appl. Supercond. 11 (1) (2001) 2909
- [2] I. Van Driessche, R. Mouton, F. Persyn, S. Hoste, E. Georgiopoulos, A. Tsetsekou, Inst. Phys. Conf. Series n° 167 (1999) 235
- [3] E. Georgiopoulos, A. Tsetsekou, C. Andreouli, Supercond. Sci Technol. 13 (11) (2000) 1539
- [4] I. Van Driessche, E. Georgiopoulos, J. Denul, A. Tsetsekou, C. Andreouli, I. De Roeck, G. De Winter, R. De Gryse, E. Bruneel, S. Hoste, EUCAS 2001
- [5] Y.K. Park, K. Park, S.G. Lee, D.C. Shin and J.C. Park, Jpn. J. Appl. Phys. 33 (1994) L718
- [6] V. Matijasevic, P. Rosenthal, K. Shinohara, A.F. Marshall, R.H. Hammond and M.R. Beasley, J. Mater. Res. 6 (1991) 682

Table I. deposition conditions of three series of two samples each, showing the influence of the used target (plasma sprayed or flame sprayed), the sputter power and the post-oxidation phase.

T_s (°C)	Pressure (Pa)	Target	Power (kW)	Oxidation Phase	XRD $\theta/2\theta$
700	5	<i>Flame sprayed</i>	1.2	2 h/500°C/800 mbar O ₂	c-axis + small (103) peak
700	5	<i>Plasma sprayed</i>	1.2	2 h/500°C/800 mbar O ₂	c-axis + small (103) + green phase
700	5	Plasma sprayed	1.2	2 h/500°C/800 mbar O₂ 30min/450°C/800 mbar O₂	c-axis
700	5	Plasma sprayed	1.2	<i>None</i>	c-axis
680	5	Plasma sprayed	1.2	4 h/600°C/800 mbar O ₂ 18 h/500°C/800 mbar O ₂	c-axis
680	5	Plasma sprayed	2	4 h/600°C/800 mbar O ₂ 18 h/500°C/800 mbar O ₂	c-axis

Table II. Deposition conditions of best YBCO layer on MgO so far deposited with the rotatable magnetron, with an additional Cu magnetron

T_s (°C)	700
Sputter pressure (Pa)	2
Ar/O ₂	1/1
Sputter power rotatable (kW)	2
Sputter power Cu-magnetron (kW)	0.6
Oxidation phase	1 h/550°C/800 mbar O ₂
$T_{c, \text{onset}}$	88 K

Fig. 1. Four XRD $\theta/2\theta$ spectra for YBCO samples on MgO deposited respectively at 630°C, 655°C, 680°C and 720°C

Fig. 2. XRD $\theta/2\theta$ spectrum of the best YBCO layer on MgO deposited with the rotatable magnetron so far (deposition conditions in table II)

Fig. 3. T_c transition of the YBCO layer in fig. 2