Solution deposition of ultrasmooth alumina on long-length metallic substrate for 2G superconducting tapes

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Chemical solution deposition of smooth amorphous Al\textsubscript{2}O\textsubscript{3} thin films has been successfully used for planarization of long-length metallic substrates for 2G HTS superconducting wires production. A series of metal-organic precursors based on Al\textsubscript{3}(\textsuperscript{3+}PO\textsubscript{4}), Al(\textsuperscript{3+}acac\textsubscript{3}), acetic acid, monoethanolamine (MEA) and diethylenetriamine (DETA) was developed. Correlations between the precursor composition and the features of its two-step thermal-induced transformation to amorphous Al\textsubscript{2}O\textsubscript{3} were studied by \textsuperscript{1}H NMR, IR spectroscopy and thermal analysis. Utilizing the solutions of [Al\textsubscript{3}(\textsuperscript{3+}PO\textsubscript{4}) + 2.5MEA + 6HAcet] in \textsuperscript{3+}PO\textsubscript{4}OH and [Al(\textsuperscript{3+}acac\textsubscript{3})\textsubscript{n} + nDETA] in HAcet (n = 1 – 3) as precursors for dip-coating, we obtained the long-length Hastelloy C276 substrate covered by uniform amorphous 50–550 nm thick alumina films with surface roughness of about 0.5–1.5 nm. A 20 m long sample of 2G HTS wire with a commercial level of superconducting critical current of up to 300 A was prepared based on Hastelloy substrate tape planarized with Al\textsubscript{2}O\textsubscript{3}.

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1. Introduction

The rapid progress in the technology of second-generation high temperature superconducting (2G HTS) wires has driven the demand for low-cost and simple approaches to smoothing the surface of long-length metal substrates. Ion beam assisted deposition (IBAD) of magnesium oxide is one of the key techniques in 2G HTS wire fabrication; it provides biaxial texture of the MgO buffer layer, which serves as a template for further texture translation to the superconducting layer\textsuperscript{[1,3,4]}. This approach gives freedom in choosing the substrate material, but, at the same time, it requires that the substrate have a very smooth surface, in the range of 1 nm, in order for MgO crystals to be aligned within a few angle degrees\textsuperscript{[2]}. The conventional technique for smoothing metal substrates for 2G HTS wires has been electrochemical polishing (EP)\textsuperscript{[5]}; however, it produces toxic waste and requires vacuum deposition of diffusion barrier (Al\textsubscript{2}O\textsubscript{3}) and seed (Y\textsubscript{2}O\textsubscript{3}) layers before applying the IBAD-MgO layer. In addition, the parameters of the electropolishing process are sensitive to the chemical composition of the substrate, representing a considerable limitation of the technique.

Recently, a simple and low-cost technique for smoothing the surface of metal tapes by depositing yttria layer from a solution, called Solution deposition planarization (SDP), was proposed\textsuperscript{[6,7]}. Unlike subtractive polishing techniques that remove rough features from the surface, the SDP method is additive and it involves filling surface irregularities with amorphous or nano-crystalline yttria particles by repeating several cycles of chemical deposition from a solution of a metal-organic precursor (MOP) followed by thermal decomposition to form yttrium oxide. The key properties required from yttria films used for surface smoothing include continuity, good adhesion to the substrate surface, and low values of the average and root mean square roughness parameters, Sa and Sp, of below 1 nm. Several groups have reported successful SDP-Y\textsubscript{2}O\textsubscript{3} smoothing of nickel alloy tapes, reducing the Sa and Sp values on a 5 × 5 \textmu m\textsuperscript{2} surface area from Sa = 2.5 nm to 0.5–0.6 nm\textsuperscript{[6]} and from Sp = 21 nm to 0.5–0.6 nm\textsuperscript{[7]}, both at approximately 1 \mu m planarizing yttria layer thickness, and from Sa = 9.0 to 0.8 nm at a film thickness of about 300 nm\textsuperscript{[8,9]}. Superconducting critical current densities at liquid nitrogen temperatures exceeding 2 MA/cm\textsuperscript{2} were reported for 2G HTS wire samples on SDP-Y\textsubscript{2}O\textsubscript{3} substrates\textsuperscript{[5,6]}. Recently, Kim and co-workers achieved superconducting critical currents in the range of 190–420 A/cm in GdBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{7} 2G HTS wires on SDP-Y\textsubscript{2}O\textsubscript{3} planarizing layers\textsuperscript{[10]}.
The SDP method has demonstrated a clear potential for low cost 2G HTS wire manufacturing and its continued development is highly desirable. Further progress in the SDP technique can be achieved, in particular, by developing new planarizing layer compositions, a promising candidate being aluminium oxide. In 2G HTS wire architecture, vacuum deposited amorphous alumina layers are routinely used as a diffusion barrier stopping metal substrate oxidation on the one hand, and contamination of the superconducting layer with metal ions from the substrate, on the other hand [4,11]. Amorphous alumina layers on Hastelloy C276 substrate are crack-free [11] despite the moderate difference in thermal expansion coefficients between amorphous alumina (6–7 ppm K⁻¹) [12] and Hastelloy C276 (11–13 ppm K⁻¹ at 20–500 °C) [13]. Thanks to its high melting point of 2044 °C, alumina may remain in the amorphous and/or nanocrystalline state without recrystallization in a wide temperature range, approximately up to 800 °C, which is favourable for the high temperature deposition of subsequent oxide layers in the 2G HTS wire heterostructure.

Paranthaman et al. [14] demonstrated the successful formation of 2G HTS wire architecture on spin-coated SDP-Al₂O₃ layers on short samples. That result encourages detailed studies of amorphous alumina formation from various Metal Organic Precursors (MOP) leading ultimately to the development of Al₂O₃ planarizing layers on long-length metal substrate tapes suitable for transferring to 2G HTS wire production. In this paper, we present the synthesis and characterization of new MOP compositions based on mixed ligand complexes of aluminium acetylacetonate or aluminium isoproxide with amines and the results of continuous reel-to-reel deposition of SDP-Al₂O₃ layers on long metal substrate tapes.

2. Experimental

2.1. Metal-organic precursor solutions

2.1.1. Materials

The starting materials: aluminium tris-isopropoxide (Al (PrO)₃), aluminium tris-acetylacetone (Al(acac)₃), acetic acid (HAcet), monoethanolamine (MEA), diethylenetriamine (DETA), and non-absolute propanol-2 (PrOH), all of analytically pure grade, were purchased from Aldrich. All operations were carried out under atmospheric conditions.

2.1.2. Preparation of MOP solutions and gel samples

Aluminium metal-organic salts, Al(PrO)₃ or Al(acac)₃, were dissolved in a mixture of corresponding reagents (HAcet, H₂O, MEA or DETA, PrOH) under stirring at room temperature, producing stable clear solutions MOP1–MOP8 (Table 1). Aluminium concentration in all MOP solutions was 0.2 M.

The MOP solutions were subjected to thermal treatment at 70 °C for 120 min in air to form gel-like samples. Gel notation numbers in Table 1 correspond to MOP solution numbers from which they were produced.

2.1.3. Characterization of MOP solutions and gel samples

The viscosity of MOP solutions was measured using a SV-10 visbro-viscometer (AnD, Japan) at 23 ± 2 °C. Wetting angles on the surface of Hastelloy C276 tape, which was used as substrate for film deposition, was measured by sessile drop method.

¹H NMR spectra of gel samples were recorded with an Agilent 400-MR spectrometer (analytical frequency 400 MHz) in CDCl₃/ TMS. We recorded ¹H NMR spectra of MEA and DETA subjected to heating at 70 °C in air (MEA-70, DETA-70), Al(acac)₃, [Hacac-MEA], and [Hacac-DETA] mixtures (Supplementary, S1–S3) and used them for gel spectra interpretation.

IR spectra of gel samples were recorded with a PerkinElmer Spectrum One FTIR spectrometer using frustrated total internal reflection technique in the 4000–650 cm⁻¹ range (Supplementary, S4).

Thermal analysis of gel samples was performed using a Derivatograph Q-1500 D thermogravimetric analyser in the 20–1000 °C temperature range in air at a 10 °C/min heating rate (45 mg batch, alundum crucible).

2.2. Film deposition and characterization

2.2.1. Film deposition

As substrates for planarization, we used cold-rolled Hastelloy C276 tapes, 110 μm thick, 12 mm wide, 1.5 m long in preliminary experiments and 20 m long in experiments performed under optimized conditions, with initial surface roughness parameters of Sₙ = 4.1 ± 0.6 nm and S₁₀ = 5.5 ± 0.9 nm (5 × 5 μm² area AFM scan). Film depositions were performed using a specially designed reel-to-reel SDP system enabling up to 10 sequential passes through a dip-coating bath, solvent evaporation furnace (80–100 °C) and decomposition furnace (500–600 °C) (Fig. 1). The length of the tape substrate that can be processed in that system is limited only by the reel size. Theoretically, up to 10 different solution compositions and/or concentrations may be used in case divided dip-coating baths are used. In this work with used one common bath containing one solution for all deposition passes/cycles, and varied the solution composition in different experiments, but kept it the same within different passes of one experiment. Ozonized airflow (5–12 l/min, ozone concentration 0.0006 g/l) was supplied into the decomposition furnace. Tape motion speed in different experiments was varied in the 3–18 m/h range. The number of dipping

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Composition and properties of precursor solutions and corresponding gels.</th>
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<tbody>
<tr>
<td>MOP Solution</td>
<td>MOP composition</td>
</tr>
<tr>
<td>MOP1</td>
<td>Al(PrO)₃ + 2.5MEA + 6HAcet</td>
</tr>
<tr>
<td>MOP2</td>
<td>Al(PrO)₃ + DETA + 2HAcet</td>
</tr>
<tr>
<td>MOP3</td>
<td>Al(acac)₃</td>
</tr>
<tr>
<td>MOP4</td>
<td>Al(acac)₃ + 3H₂O</td>
</tr>
<tr>
<td>MOP5</td>
<td>Al(acac)₃ + MEA</td>
</tr>
<tr>
<td>MOP6</td>
<td>Al(acac)₃ + 3 MEA</td>
</tr>
<tr>
<td>MOP7</td>
<td>Al(acac)₃ + DETA</td>
</tr>
<tr>
<td>MOP8</td>
<td>Al(acac)₃ + 3 DETA</td>
</tr>
</tbody>
</table>

a Aluminium concentration in all MOP solutions was 0.2 M.
b MOP solution after solvent evaporation at 70 °C for 2 h.
c Temperature of gel transformation to Al₂O₃, according to TG in air (°C).
3. Results and discussion

In the SDP method, the formation of planarizing layers occurs according to Scheme 1 and the continuity and smoothness of planarizing layers depend on the nature of MOP.

Suitable aluminium-containing MOPs must form clear solutions, be stable against sediment formation in a wide range of concentrations (up to 0.4 M), wet the substrate surface well and form continuous gel-like precursor films (MOP films) upon evaporation of the solvent. If formed properly, MOP films readily transform to continuous and smooth alumina films at temperatures below the crystallization temperature of Al₂O₃ (no higher than 600 °C). In this work, following Scheme 1, we synthesized and characterized new MOP solutions, transformed them into bulk gel samples modelling MOP gel-like films, and studied the composition and thermal stability of the gels. Based on the data obtained, we chose the most suitable MOPs and used them for the deposition of planarizing alumina layers.

MOP solution → MOP film → Planarizing oxide film

Scheme 1. Stages in the solution deposition process.

3.1. Synthesis and characterization of precursors

3.1.1. MOP solutions

Two well-studied aluminium compounds, tris-isopropoxide, Al(‘OPr)₃, and tris-acetylacetonate, Al(acac)₃, often used for sol-gel syntheses of alumina bulk ceramics and coatings [14,16,17], were chosen as starting compounds for MOP syntheses.

The objective of the MOP syntheses was to modify the starting compounds in such a way that the resulting MOP solutions easily transform into suitable MOP films that ultimately produce good planarizing Al₂O₃ films (Scheme 1). We used acetic acid (HAcet) and amines as modifying agents. The selected amines, monoo- ethanolamine (MEA) and diethylenetriamine (DETA), can act as both hydrolyzing agents and chelating or bridging ligands in MOP [18–24]. Earlier we have demonstrated the efficiency of these amines as MOP modifying agents for the deposition of MOD-CeO₂ [25] and SDP-Y₂O₃ layers for 2G HTS wires [8,9].

A series of MOP solutions Al[PrOH]₃–HAcet–Q in iPrOH and Al(acac)₃–Q in HAcet (Q = MEA, DETA) was prepared (Table 1) and characterized against the suitability criteria for SDP MOP1–MOP8 solutions were, although stable clear solutions of Al[PrOH]₃ in non-absolute iPrOH (MOP1, MOP2) could be obtained only using a combination of HAcet and amines and after stirring for 24 h. For MOP3–MOP8 solutions, the viscosity values increased, the pH values increased with an increase of Q content. The values of wetting angles decreased with addition of MEA (MOPS, MOP6, Table 1) and increased with addition of DETA (MOP7, MOP8, Table 1) in comparison to MOP3, which did not contain any amine. Although wetting angle values for the solutions varied in a relatively wide range, even solutions with the higher wetting angle values appeared to be suitable for continuous alumina film deposition, as we show below in this paper.

3.1.2. Gel composition

An important step in the SDP process is the thermolysis of the gel-like MOP film in the course of its transformation into the smoothing amorphous oxide. We used bulk samples of Gel1–8 as models of MOP gel-like films. In fact, samples obtained after evaporating solvent from MOP3 and MOP4 were not gels but microcrystalline powders; therefore, in this case the notation ‘Gel’3 and ‘Gel’4 is arbitrary. The thermolysis result depends on the gel composition; therefore, we studied the composition of gel samples obtained after solvent removal from MOP solutions.

We anticipated that under the action of HAcet and amines both starting aluminium compounds, Al[PrOH]₃ and Al(acac)₃, would transform into mixed ligand complexes containing the acetate, amine and hydroxyl ligands in addition to or instead of the initial isopropoxide or acetylacetonate anions. In order to verify this suggestion, we studied the ¹H NMR spectra and IR spectra of the Gels (Supplementary, Table S1–S4), using ¹H NMR spectra of all starting reagents (¹H NMR, HAcet, H₂O, MEA, DETA) and Hacac-Q mixtures (Table S3) for interpretation.

In ¹H NMR spectra of Gel1 and Gel2 the proton resonance signals of ethylene, amino and hydroxy groups of MEA or DETA shifted relative to their positions in the spectra of MEA and DETA, and so did the signals of methyl groups in HAcet, Acet · iPrOH (Fig. 2, Table S2). Based on the ratios of the signal integral intensities, the compositions of Gel1 and Gel2 were described as mixed ligand hydroxocetate complexes Al[Acet](OH)₃(MEA)ₓ(‘PrOH)₀.₃₋ₓ(H₂O)₀.₅₋ₓ(HAcet)ₓ and Al[Acet]₀.₃₋ₓ(OH)₀.₅₋ₓ(DETA)ₓ(PrOH)ₓ, respectively. The IR spectrum data are in agreement with the proposed compositions (Table S4): the δ(NH₂), ν(COO) and ν(N – H + OH) bands were detected in the spectra of Gel1 and Gel2. The low intensity band in the ~1710 cm⁻¹ range was due to the small amount (x) of HAcet in the Gel1.
The data of $^1$H NMR (Table S3) and IR spectroscopy (Table S4) showed that the composition of Gel3 and Gel4 was identical to starting Al(acac)$_3$. In $^1$H NMR spectra only the signals of the acetylacetonate ion protons ($\delta$(CH$_3$) and $\delta$(CH)) occurred, and the IR spectra contained $\nu$(C–H), $\nu$(C–O) and $\nu$(C–C) bands and fully coincided with the starting Al(acac)$_3$ spectrum (Fig. 3). This means that the addition of HAcet or HAcet and H$_2$O did not result in the transformation of aluminium acetylacetonate into mixed ligand or hydroxo-complexes. This agrees with the very high stability constant of Al(acac)$_3$, $\log K = 23.7$ [26].

The transformation of Al(acac)$_3$ into mixed ligand complexes did occur with amine addition (MOPS–8, Table 1) that was confirmed by the data of $^1$H NMR and IR spectroscopy of Gels 5–8 (Tables S3 and S4). The $^1$H NMR spectra of these Gels had a rather complicated character due to the reaction between Hacac and Q resulting in Schiff Bases (SB). According to $^1$H NMR spectra, the Hacac–MEA and Hacac–DETA solutions (Table S3) contained the mixtures of corresponding SB, enol- and keto-forms of Hacac, and Q; with the SB mole parts of approximately $\sim$ 0.5 in the Hacac–MEA mixture and $\sim$0.9 in the Hacac–DETA mixture. In the $^1$H NMR spectra of Gels 5–8 we found the proton resonance signals of SB, enol- and keto-forms of Hacac, Q, hydroxogroups, and HAcet/acet– (Tables S3). The integral intensities of these component signals depend on the nature and initial concentration of Q. The formation of SB is a result of Al(acac)$_3$ hydrolysis. The general composition of Gels 5–8 can be represented as $\{\text{Al}([\text{acet}]_k(\text{acac})_m(\text{OH})_n(\text{SB})_p(\text{Q})_q(-\text{Hacac})_x(\text{H}_2\text{O})_y(\text{HAcet})_z)\}$ where the coefficients $k$, $m$, $n$, $p$, $q$, $x$, $y$, $z$ vary depending on the starting precursor solution composition. The IR analysis data agreed with the $^1$H NMR results. The bands of stretching and deformation vibrations of acac–, Hacac, OH–, SB, and Q, and the low intensity (C–O) band of HAcet were found in the IR spectra (Fig. 3). The obtained data showed that the
3.1.3. Gel thermodysis

Thermal decomposition of the gels was studied by thermal analysis in air (Fig. 4). The TG curves of Gel3 and Gel4 were very similar and the gels transformed into Al2O3 at approximately 420 °C, like aluminium acetylacetonate. The temperature of Al2O3 formation for the other gels was in the 520–570 °C range (Table 1)—all below 600 °C, the allowable maximum temperature criterion for MOP for SDP-Al2O3—and increased in the following sequence: Gel1 (520 °C) < Gel2, Gel5, Gel6 (540 °C) < Gel8 (550 °C) < Gel7 (570 °C).

3.1.4. Selection of MOP suitable for planarizing Al2O3 layer deposition

Based on the analysis of gel composition and their thermodysis behaviour, we concluded that the addition of MEA or DETA produced MOP solutions suitable for the deposition of planarizing amorphous Al2O3 films by stabilizing Al(PrO)3 in non-absolute PrOH and promoting the hydrolysis of Al(acac)3 in HAcet and resulting in gels that decomposed to aluminium oxide at 520–570 °C.

We assessed the ability of solutions MOP 1–8 to form continuous precursor films on the surface of Hastelloy C276 substrates. Although MOP3 and MOP4 had the lowest decomposition temperature, they produced Al(acac)3 microcrystalline powder instead of gel-like films upon thermal treatment, therefore we did not use these MOP’s in Al2O3 film deposition experiments. The poor smoothing properties of a 1 M MOP3 solution were also shown in [14]: the surface roughness of Hastelloy C276 substrate in spin-coating deposition experiments was decreased from 9.5 to 2.4 nm only (on a 2 × 2 μm² area). Thus, we selected MOP 1, 2, 5–8 for further film deposition experiments.

3.2. Deposition and characterizati0n of Al2O3 planarizing films

The smoothing effect of solutions MOP 1, 2, 5–8 was tested in a series of experiments in the reel-to-reel dip-coating system (Fig. 1) with variable deposition parameters: substrate tape motion speed (3–18 m/h), the number of deposition cycles (1–10), decomposi-
tion temperature (500–600 °C), and ozonized airflow (5–12 l/min). The length of Hastelloy C276 substrate tapes was 1.5 m in preliminary experiments and 20 m in experiments performed under optimized conditions. In the preliminary set of experiments, we chose the more promising MOP solution compositions and deposition parameters based on the resulting Al2O3 film thickness, uniformity, roughness, and residual carbon content.

Films deposited using MOP 2, 5 and 6 were very thin (~20 nm after one deposition cycle) and not uniform (Fig. 5a), most likely due to the low viscosity values of these solutions (1.5–2.0 Pa s, Table 1).

Uniform alumina films (Fig. 5b) with excellent smoothing effect were obtained using compositions MOP 1, 7, 8 under the following optimized deposition conditions: tape motion speed of 18 m/h, decomposition temperature of 550 °C and airflow rate of 12 l/min (Table 2).

According to XRD data, all alumina films were amorphous, with only (111), (200) and (220) Ni-alloy reflections of Hastelloy C276 substrate present (Fig. 6).

High residual carbon content in SDP films is very undesirable, since during subsequent high temperature treatment steps in oxygenating environment, when the other layers of 2G HTS wire are being processed, carbon may form carbon monoxide gas that will disrupt and damage continuous layers. In addition, high residual carbon content may lead to poor texture in IBAD-MgO films [27]. According to our EDX data the approximate content of residual carbon in all films is about 2%. This value is as low as for bare Hastelloy, 2.5 ± 1.1%, thus it satisfies the condition of IBAD-MgO technique.

The film thickness increment per each deposition cycle was 50–120 nm (Table 2). For MOP 1, 7, the film thickness increased linearly

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**Fig. 4.** TG curves of Gels 1, 3 and 7. All samples decomposed to form aluminium oxide at temperatures below 600 °C.

**Fig. 5.** The SEM plain view of amorphous Al2O3 films after single SDP cycle from MOP5 (Film 5–1, a) and MOP1 (Film 1–1, b). The Film 5–1 has non-uniform surface with many conglomerates. The Film 1–1 demonstrates smooth uniform surface.
Table 2
Properties of Al₂O₃ films deposited from solutions MOP 1, 7, 8 under optimized conditions.a

<table>
<thead>
<tr>
<th>MOP</th>
<th>Number of deposition cycles</th>
<th>Roughness, 5 × 5 μm²</th>
<th>Film thickness (nm)</th>
<th>Residual carbon content (at.%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>S₁(nm)</td>
<td>S₂(nm)</td>
<td></td>
</tr>
<tr>
<td>Film 1-1</td>
<td>1</td>
<td>1.0 ± 0.6</td>
<td>1.2 ± 0.7</td>
<td>~95</td>
</tr>
<tr>
<td>Film 1-5</td>
<td>1</td>
<td>0.9 ± 0.2</td>
<td>1.1 ± 0.3</td>
<td>~560</td>
</tr>
<tr>
<td>Film 7-1</td>
<td>7</td>
<td>0.5 ± 0.1</td>
<td>0.8 ± 0.4</td>
<td>~55</td>
</tr>
<tr>
<td>Film 7-2</td>
<td>7</td>
<td>0.4 ± 0.1</td>
<td>0.7 ± 0.4</td>
<td>~100</td>
</tr>
<tr>
<td>Film 7-5</td>
<td>7</td>
<td>0.3 ± 0.1</td>
<td>0.5 ± 0.1</td>
<td>~330</td>
</tr>
<tr>
<td>Film 8-1</td>
<td>8</td>
<td>2.7 ± 0.6</td>
<td>3.7 ± 1.0</td>
<td>~120</td>
</tr>
<tr>
<td>Film 8-5</td>
<td>8</td>
<td>1.0 ± 0.1</td>
<td>1.3 ± 0.2</td>
<td>~360</td>
</tr>
</tbody>
</table>

For bare Hastelloy C276 substrate Sₐ = 4.1 ± 0.6 nm, Sₕ = 5.5 ± 0.9 nm (5 μm scale).

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**Fig. 6.** XRD patterns of bare Hastelloy C276 substrate and amorphous SDP-Al₂O₃ Film 1-5. XRD patterns of all Al₂O₃ films were identical containing the substrate reflections only.

**Fig. 7.** Dependence of surface roughness (S₂) on the number of deposition cycles for alumina films on Hastelloy C276 substrates deposited using MOP 1, 7, 8. Surface smoothing to 1 nm level occurs after 5 deposition cycles. With MOP 1 and 7 this level of smoothing is achieved already in 1–2 deposition cycles.

**Fig. 8.** AFM data on surface morphology (5 × 5 μm² scan area) for Film 1-1, Sₐ = 1.0 ± 0.6, Sₕ = 1.2 ± 0.7 (a) and Film 7-1, Sₐ = 0.5 ± 0.1, Sₕ = 0.8 ± 0.4 (b). Blanc lines show cross section profile. Both films were obtained in a single deposition cycle.
obtained layer. Commercial roughness comparing developed MOP with roughness (Fig. 3.3.70 with Fig. 3.4.1). Critical currents are at the present commercial level, in the 250–300 A range, with a uniform distribution along the taper length.

We have developed several metal-organic precursor solution compositions for SDP-Al2O3 containing aluminium isopropoxide, aluminium acetylacetonate, acetic acid and amines dissolved in isopropanol or acetic acid. Suitable metal-organic precursor solutions were stable and clear; upon solvent removal they produced gel-like films containing mixed ligand complexes of aluminium with the general formulas Al(Alc)n(mOH)n(MEA)p(PrOM)q(HAcet)z(H2O)z and [Al(acac)2n(OH)2n(SB)2n(Q)n(HAcet)z(H2O)z], where SB is Schiff Base and m, n, p, q, x, y, and z values depend on the starting precursor solution composition and thermal treatment regime.

Thermal decomposition of the gel-like precursor films at temperatures below 600°C resulted in the formation of amorphous Al2O3 films with a smooth surface. Optimized precursor compositions and deposition conditions produced planarizing Al2O3 films on Hastelloy C276 substrate tapes that reduced the initial bare substrate roughness values from about 5 nm to below 1 nm (Sa and Sz at 5 × 5 µm² AFM scan area) in as few as 5 SDP cycles, which is a considerable advancement from the previously reported 10–40 cycles of SDP-Y2O3 required for similar smoothing results. Moreover, with MOP solution containing Al(acac)3 and DETT dissolved in HAcet, we succeeded in planarization of the initial Hastelloy substrate to roughness values below 1 nm with a 100 nm thick alumina film deposited in a single SDP cycle. This result is very important for the development of cost-effective technology of 2G HTS wires.

The superconducting properties of a 20 m long 2G HTS wire sample deposited on the SDP-Al2O3 substrate developed in this work were at the level of present commercial quality 2G HTS wires, with superconducting critical currents at 77 K in self-field in the range from 250 to 300 A and a rather uniform critical current distribution along the entire wire length.

The results of this work encourage further development of SDP-Al2O3 technology.

**Acknowledgments**

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3.3. Deposition of 2G HTS wire architecture onto Al2O3-planarized Hastelloy substrate

We used a 20 m long Al2O3-planarized Hastelloy tape (Fig. 1-1) as a substrate in the deposition of a complete 2G HTS wire architecture, which was kindly performed for us by our colleagues at SuperOx Japan LLC. A detailed description of the SuperOx 2G HTS wire architecture and fabrication processes can be found in ref. [11]. One SDP cycle producing an about 100 nm thick Al2O3 film with Sz = 1.0 nm replaced two processing steps: electropolishing of Hastelloy tape and vacuum deposition of Al2O3 diffusion barrier layer. The superconducting properties of the resulting 2G HTS wire based on the SDP-Al2O3 substrate were at the level of present commercial quality 2G HTS wires: the superconducting critical currents at 77 K in self-field were in the range from 250 to 300 A with a rather uniform distribution along the entire wire length (Fig. 9). This result verifies the suitability of MOP compositions developed in this work for SDP-Al2O3 intended for subsequent 2G HTS wire fabrication.

4. Conclusions

Solution deposition planarization is a very promising technique for metal tape substrate preparation in low-cost 2G HTS wire fabrication. Amorphous alumina films are a viable alternative to the more conventional Y2O3 planarizing layers.

with the number of deposition cycles, which was not the case with MOP 8.

For practical reasons, it is desirable to achieve good planarization effect in as few as possible deposition cycles. Alumina films obtained using MOP 1, 7 and 8 demonstrated planarization to roughness values in the range of 1 nm and below after 5 cycles (Table 2, Figs. 7 and 8), which is a considerable advancement comparing to previously reported results for Y2O3 planarization where similar roughness values were reached only after 10 [8,9] or 30–40 [6] deposition cycles. Even more importantly, compositions MOP 1 and, especially, MOP 7 produced Al2O3 films with very low roughness already after a single deposition cycle (Fig. 7). This result is very important for the development of cost-effective technology of 2G HTS wires, since an about 10-fold reduction of surface roughness to sufficiently low values is achieved with a simple one-stage procedure with a 50–100 nm thick film instead of multi-stage depositions of 300 to 1000 nm thick films [6,8,9].

**Fig. 9. Architecture of the SuperOx 2G HTS wire on planarized substrate (a) and positional critical current measurement data (THEVA TapeStar) along the length of 2G HTS wire based on Film 1-1 (b). Critical currents are at the present commercial level, in the 250–300 A range, with a uniform distribution along the taper length.**
Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.materesbull.2016.02.014.

References