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Chemical Solution Deposition Based Oxide Buffers and YBCO Coated Conductors

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1. Introduction

The main objective of this work is to conduct fundamental research in the broad areas of chemical solution based buffer and high temperature superconductor, namely Yttrium Barium Copper Oxide (YBCO) development. The results of this research provide new insights in buffer/superconductor areas and suggest methods to improve buffer/superconductor multi-layer thin film fabrication. The overall purpose is to develop a potentially lower-cost, high throughput, high yield, manufacturing processes for buffer/superconductor thin multi-layer film fabrication, and to gain fundamental understanding of the growth of solution buffer/superconductor layers for Rolling Assisted Biaxially Textured Substrate (RABiTS) templates. This understanding is critical to the development of a reliable, robust, long-length manufacturing process of second-generation (2G) wires for electric-power applications. In order to reduce the cost of superconductor wires, it is necessary to replace the existing physical vapor deposited three buffer layer RABiTS architecture of Yttrium Oxide, Y$_2$O$_3$ seed/Yttria Stabilized Zirconia, YSZ barrier/Cerium Oxide, CeO$_2$ cap with buffers deposited by industrially scalable methods, such as slot-die coating of chemical solution deposition (CSD) precursors [1-11]. Spin-coating is typically used to deposit short samples for optimizing the CSD film growth conditions. In a typical chemical solution process, metal organic precursors in suitable solvents are spin/dip/slot-die coated on either single crystal or biaxially textured substrates followed by heat-treating in a tube furnace under controlled conditions. Chemical Solution Deposition (CSD) process offers significant cost advantages compared to physical vapor deposition (PVD) processes [5-11]. Solution coating is amenable to complex oxides, and the materials utilization (yield) is almost 100%. The high-temperature superconductors (HTS) such as (Bi,Pb)$_2$Sr$_2$Ca$_2$Cu$_3$O$_{10}$ (BSCCO or 2223 with a critical temperature, $T_c$ of 110 K) and YBa$_2$Cu$_3$O$_{7-δ}$ (YBCO or 123 with a $T_c$ of 91 K) have emerged as the leading candidate materials for the first generation (1G) and second generation (2G) high temperature superconductor wires or tapes that will carry high critical current density in liquid nitrogen temperatures [1,2]. Here, we report the growth of buffer/YBCO superconductor film growth using a chemical solution method towards fabrication of second generation superconductor wires.

2. Chemical solution deposition of oxide buffers

The schematic of the standard RABiTS architecture developed by Oak Ridge National Laboratory and American Superconductor Corporation [3,4] is shown in Figure 1. The main...
goal is to replace the most commonly used RABiTS architectures with a starting template of biaxially textured Ni-5 at.% W substrate with a physical vapor deposited (PVD) seed layer

![Fig. 1](image-url)  
The schematic of the standard RABiTS architecture.

| Buffers | Structure type | % lattice mismatch | Ref. |
|---------|----------------|--------------------|------|
|         | Cubic lattice parameter a | vs. YBCO vs. Ni |      |
| BaCeO₃  | 4.377          | 13.55              | 21.59 | 12 |
| BaZrO₃  | 4.193          | 9.27               | 17.34 | 13 |
| Pb₀.₃₆La₀.₃TiO₃ | 3.916 | 2.37               | 10.34 | 15-17 |
| La₃TaO₇  | 11.054         | 2.16               | 10.26 | 18,19 |
| SrTiO₃   | 3.905          | 0.54               | 8.64  | 20,21 |
| Eu₂O₃    | 10.868         | 0.51               | 8.61  | 22 |
| NdGaO₃   | 5.431          | 0.12               | 8.22  | 23,39 |
| CeO₂     | 5.411          | 0.07               | 8.17  | 24 |
| Gd₂O₃    | 10.813         | -0.20              | 7.90  | 20,25-27 |
| La₂Zr₂O₇ | 10.786         | -0.75              | 7.35  | 28,29 |
| LaAlO₃   | 5.364          | -1.42              | 6.95  | 30 |
| Gd₃NbO₇  | 10.659         | -1.89              | 6.22  | 31 |
| Y₂O₃     | 10.604         | -2.64              | 5.47  | 32,33 |
| Gd₂Zr₂O₇ | 5.264          | -2.88              | 5.23  | 30 |
| Y₃NbO₇   | 5.250          | -3.50              | 4.61  | 34 |
| Yb₂O₃    | 10.436         | -3.87              | 4.61  | 34 |
| Ni       | 3.524          | -4.16              | 4.61  |      |

Table 1. Structure, lattice misfit data and chemical solution deposition (CSD) methods for various buffer layers. The lattice parameters were obtained from the International Center for Diffraction Data, Powder Diffraction Files. * Rhombohedral; ♦ Orthorhombic

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of $Y_2O_3$, a barrier layer of YSZ, and a CeO$_2$ cap layer by a chemical solution deposition method. To develop an all solution buffer/YBCO, it is necessary to either replace all three layers or reduce the number of buffer layers to one. The role of the $Y_2O_3$ seed layer is to improve the out-of-plane texture of buffer layer compared to the underlying Ni-5W substrate and $Y_2O_3$ is also an excellent W diffusion and good oxygen barrier [4]. The role of YSZ barrier layer is to contain the diffusion of Ni from the substrate into superconductor. In order to grow YBCO superconductor films with critical current densities, it is necessary to contain the poisoning of Ni into YBCO. Finally, the CeO$_2$ cap layer is compatible with CSD based REBCO films and has enabled high critical current density REBCO films. The optimized film thickness for each buffer layer is 75 nm and the typical YBCO layer thickness is ~ 1 µm carrying a critical current of 250-300 A/cm-width at 77 K and self-field. Researchers all over the world have developed several chemical solution deposited oxide buffer layers that are suitable for YBCO film growth. A partial list of several epitaxial oxide buffers grown using a CSD method have been reported in Table 1 [4]. It is possible for us to select a buffer layer to lattice match with either the substrate Ni/Ni-W or with YBCO. The list of chemical solution deposited buffer layers with YBCO superconductor films deposited on such buffers is reported in Table 2.

| CSD Buffer Layers | Stacking for YBCO | $J_c$ (MA/cm$^2$) | Reference |
|-------------------|-------------------|------------------|-----------|
| CeO$_2$          | YBCO (CSD)/CeO$_2$ (Sputtered)/YSZ (Sputtered)/CeO$_2$ (CSD)/Ni-W | 3.3 | 39 |
| YSZ              | YBCO (CSD)/CeO$_2$ (CSD)/YSZ (CSD)/CeO$_2$ (CSD)/Ni | 0.5 | 35 |
| $Y_2O_3$         | YBCO (PLD)/CeO$_2$ (Sputtered)/YSZ (Sputtered)/$Y_2O_3$ (CSD)/Ni-W | 1.2 | 31 |
| Eu$_2$O$_3$      | YBCO (ex-situ BaF$_2$)/CeO$_2$ (Sputtered)/YSZ (Sputtered)/Eu$_2$O$_3$ (CSD)/Ni | 1.1 | 20 |
| Gd$_2$O$_3$      | YBCO (PLD)/CeO$_2$ (Sputtered)/YSZ (Sputtered)/Gd$_2$O$_3$ (CSD)/Ni-W-Fe | 1 | 36 |
| Ce-Gd-O         | YBCO (CSD)/CeO$_2$ (CSD)/CGO (CSD)/Gd$_2$O$_3$ (CSD)/Ni | 0.1 | 37 |
| SrTiO$_3$        | YBCO (CSD)/STO (CSD)/Ni | 1.3 | 38 |
| La$_2$Zr$_2$O$_7$| YBCO (e-beam)/CeO$_2$ (Sputtered)/YSZ (Sputtered)/LZO (CSD)/Ni | 0.48 | 26 |
| La$_{1/4}$Zr$_{3/4}$O$_y$ | YBCO (PLD)/La$_{1/4}$Zr$_{3/4}$O$_y$ (CSD)/Ni-W | 0.55 | 42 |
| Gd$_2$Zr$_2$O$_7$| YBCO (MOCVD)/GZO (CSD)/Ni | 1.3 | 33 |
| Gd$_3$NbO$_7$    | YBCO (PLD)/GNO (CSD)/Ni-W | 1.1 | 30 |

Table 2. List of chemical solution deposited oxide buffer layers with $J_c$ of the high temperature superconducting YBCO films deposited on such buffers.
3. Chemical solution deposition of REBCO

Currently, chemical solution based synthesis of YBCO uses a trifluoroacetate (TFA) based precursor approach [5]. In this approach, the precursor solution is prepared by dissolving Yttrium, Barium and Copper trifluoroacetates in methanol. Then the precursor solution is spin/slot-die coated on RABiTS templates followed a two-stage heat-treatment to convert the precursor films to high quality YBCO. In the first stage (pyrolysis), there is a significant bottle neck to processing rates for these films because the shrinkage stresses developed in the films during pyrolysis need to be accommodated using very slow heating rates. The reactions taking place during the synthesis are illustrated below:

\[
Y(\text{OOCCF}_3)_3 + 2 \text{Ba(OOCCF}_3)_2 + 3 \text{Cu(OOCCF}_3)_3 \rightarrow 0.5 \text{Y}_2\text{O}_3 + 2 \text{BaF}_2 + 3 \text{CuO} + (n\text{CO}_2 + m\text{C}_x\text{O}_y\text{F}_z)\] \hspace{1cm} (1)

\[
0.5 \text{Y}_2\text{O}_3 + 2 \text{BaF}_2 + 3 \text{CuO} + 2 \text{H}_2\text{O} \rightarrow \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} + 4\text{HF}\hspace{1cm} (2)
\]

Significant efforts were made to increase the growth rate by replacing part of the metal TFA precursors with non-fluorine based precursors and also adjust the water and oxygen pressure during the growth of YBCO films. Another advantage of the TFA process is to introduce mixed rare earths and Zirconium oxides into the starting precursors to enhance the flux-pinning properties of REBCO films [5,40,41]. Chemical solution deposition method may prove to be a promising route for producing a low-cost all-CSD buffer/YBCO based coated conductors. The main challenge is to fabricate high-temperature superconductor tapes in kilometer lengths in carrying 1000 A/cm-width. Industries from US and Japan are leading in this area while industries from Europe, Korea, and China are only few years away.

4. Summary

In summary, RABiTS template with several possible architectures based on chemical solution deposition methods have been developed and superconductivity industries around the world are in the process of taking the technology to the pilot scale to produce commercially acceptable 500 meter lengths. The research in the area of second generation high temperature superconductor wire technology to increase the flux pinning properties of YBCO superconductor and to reduce the ac loss in these wires for various electric-power applications such as transmission cables, fault-current limiters and high-field magnets is continuing ahead.

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