Advances in ultrafast transient liquid assisted growth (TLAG) of YBa$_2$Cu$_3$O$_7$ coated conductors

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**TRANSIENT LIQUID ASSISTED GROWTH: TLAG**

A high throughput non-equilibrium kinetically controlled growth process

**DEPOSITION METHOD**

- Nanocrystalline precursors
- Transient liquid + $Y_2O_3$
- YBCO nucleation
- YBCO growth

**PRECURSORS** → \((Ba – Cu^{III}-O)(l) + RE_2O_3(s) → REBa_2Cu_3O_{7-δ}\)

**BaCO_3(s) + CuO(s) + Y_2O_3(s)** → **TLAG-CSD**

- FF metal-organic solution
- L. Saltarelli et al., ACS Appl. Mat. & Interf. (2022)

**REBCO target deposited at low T and P_{O_2}**

**TLAG-PLD**

- A. Queralto et al, SUST (2023)

**Solid-solid reaction**

- Fast atomic diffusion
- Ultrafast growth rate, G, up to 2000 nm/s demonstrated
- Large area deposition
- Simple reactor
- High throughput
- Low cost/performance ratio
Pyrolyzed F-free CSD films

- Propionate precursors + additives
- Optimised solutions of various stoichiometries yield homogeneous nanocrystalline layers

- Reduced sizes of the nanocrystalline YBCO precursors favour greatly atomic mobility, enabling ultrafast growth rates

- BaCO₃ (orthorhombic): 10 - 30 nm
- CuO: 10 - 25 nm
- Y₂O₃: 5 - 6 nm

- Nanoscale homogeneous distribution of the phases throughout the layer

Multifunctional colloidal ink (Patent EP22382741)

L. Saltarelli et al., ACS Appl Mat Interfaces (2022)
CSD INKS AND PYROLYZED MULTIDEPOSITED FILMS

Multifunctional ink formulation
- Uniform pyrolyzed layers
- Low porosity
- Nanocrystalline and homogeneous
- Thickness beyond 3 µm tested
- BaCO$_3$ eliminated in TLAG

Multifunctional colloidal ink
- Hybrid Hydrolitic-Solvothermal Synthetic Process (H2S2)
- BaZrO$_3$, BaHfO$_3$ Np
- Similar performance
- Crystalline BZO NPs retain their shape and small size in the pyrolysis

TLAG in-situ growth evaluation

from 100 ms down to 2 ms acquisition time

In-situ growth XRD synchrotron experiments

TLAG Kinetic phase diagrams

\[ T \ (°C) \]

\[ 1000 \, P_0 \ (\text{bar}) \]

\[ T_P \]

\[ T_E \]

\[ T_{\text{ins}} \]

\[ \text{BaCu}_2\text{O}_2\text{O}_{2(\text{s})} + \text{YBCO} \]

\[ \text{BaCu}_2\text{O}_2\text{O}_{2(\text{s})} + \text{CuO}_{(\text{s})} \]

\[ \text{BaCu}_2\text{O}_2\text{O}_{2(\text{s})} + \text{CuO}_{(\text{s})} \]

\[ 1000/T (\text{K}^{-1}) \]

\[ G_{\text{max}} \ (\text{nm s}^{-1}) \]

\[ P_{\text{O}_2} \ (\text{x}10^{-3} \text{ bar}) \]

Epitaxial films and nanocomposites at 2000 nm/s


by in-situ resistivity and in-situ XRD
In-Situ ALBA Synchrotron installation

NCD-SWEET BEAMLINE

HIGH THROUGHPUT EXPERIMENTATION

Synchrotron XRD

Mass spectroscopy

Resistivity

Experiment conditions

PTI+ TRANS-ENER+

Transición Energética Sostenible
Fast optimization using Compositional Gradients

Combinatorial DoD Ink Jet Printing

YBCO  GdBCO

Y content ~88%
Y content ~65%

in-situ synchrotron XRD

Data is segmented by positions for analysis

Case position 1

A. Queralto et al., ACS Appl. Mater. Interfaces (2021)
**YBCO TLAG-CSD FILMS**

- 50 nm/s 1500 nm/s
- $\gamma_{\text{eff}} = 2.5-3.5$

**Nanocomposites**

- YBCO (103)
- BZO (110)
- YBCO (102)

- 6% BZO

$\gamma_{\text{eff}} = 2-2.5$

- $J_c(5 \text{ K})$ MA/cm$^2$
- $J_c(77 \text{ K})$ MA/cm$^2$

- $\text{Nanoparticles}$ (mol%)

- $\text{Final } P_{\text{O}_2} (\times 10^{-3} \text{ bar})$
YBCO TLAG-CSD NANOCOMPOSITE FILMS

Extended to technical substrates in collaboration with SUMITOMO ELECTRIC

Slot-die coating (6 layers)

2.8 ± 0.08 μm

Buffer layers

J_c(5K) = 24 MA/cm², I_c, 750 nm (77K) = 130 A/cm-w

High density of defects is present as well as embedded nanoparticles
VORTEX PINNING IN TLAG-CSD NANOCOMPOSITE FILMS

H<\*: single vortex pinning regime
(measure of the density of pinning centers)

T = 30K

T = 77K

TLAG films respond to vortex pinning determined by high density of defects
(SF, point vacancies) and nanoparticles
OPTIMIZING PINNING LANDSCAPES

Different microstructure requirements for different regions

F. Valles et al, Comm Mat. (2022)
T. Puig et al. (to be published)
OVERDOPING: A ROBUST METHOD FOR PINNING

\[ J_c^2 \propto n_H E_c(n_H) \]


Condensation energy and charge carrier density increases in the overdoped state

\[ J_c(p^*) \approx \frac{1}{5} J_d(p^*) \]

See also: M. Miura et al. NPG Asia Mat. (2022)
Contributions from CC Materials Research

First time line

Materials R&D

CC R&D

CC scalab.

CC Industry

CC devices PoC

Eng. R&D

CC devices

Eng. Industry

MARKET

Advanced CC R&D

Second wave of devices

Low-cost, High-throughput, High-performance

\[
\frac{\text{Cost}}{\text{Performance}} = \frac{\text{total cost per year}}{G \times L \times W \times J_c} = \frac{\€}{kA \times m}
\]

- Fast growth-rate methods
- Optimization of robust vortex pinning schemes
- Growth of wide tapes

High-throughput experimentation can be of great help
Conclusions

- TLAG is a non-equilibrium growth methodology with ultrafast growth rates achieving high performance Coated Conductors compatible with CSD

- In-situ synchrotron XRD experiments have been essential to understand the kinetic growth mechanisms and obtain epitaxial layers at ultrafast growth rates

- Pinning landscapes can be optimized at different regions depending on applications needs, overdoping being very appealing

- High Throughput Experimentation initiatives are foreseen to fasten optimization schemes

- Industrialization of TLAG-CSD should lead to a high throughput and low cost manufacturing process which uses simple and large area reactors for R2R CC production