Materials opportunities to boost HTS-CC
Example of Transient liquid Assisted Growth

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Materials Science opportunities for CC

Decrease the cost/performance ratio

1. Decrease the cost/performance ratio by developing high throughput growth methods:
   Transient Liquid assisted growth, TLAG-CSD process (100-1000 nm/s)

2. Use of High Throughput Experimentation strategies for fast optimization of processes:
   Combinatorial approaches and Machine Learning algorithms

3. Foster higher critical currents by engineering the vortex pinning landscape for each application:
   Understanding the (H,T,θ) pinning landscape for engineered nanocomposites

4. Increase pinning force by increasing charge carrier density:
   Tune charge carrier density by oxygen overdoping
1. TLAG-CSD process

An opportunity for high-throughput, scalable and low cost production of Coated Conductors at ultrafast growth rate

- Non-equilibrium process kinetically controlled by supersaturation reaching ultrafast growth rates 100 nm/s – 1000 nm/s from a transient liquid-solid reaction

- Supersaturation is the driving force for nucleation, controls liquid properties, substrate wettability and c-axis window. It can be modified by: Ba-Cu-O liquid composition, RE solubility, T and PO2 ramps and (T, PO2) values reached

- Environmentally friendly fluorine free precursors based on BaCO3- CuO reaction

- Well-matched with large area deposition, simple and large area reactor of low cost investment

Pyrolyzed films and intermediate phases

BaCO₃ decomposition verified in 2.5 µm thick pyrolyzed layers

In-situ XRD synchrotron exp.
Ultrafast (non-equilibrium) growth by TLAG-CSD

Kinetic phase diagrams

**TLAG-CSD films properties**

\[ J_c (77K, sf) = 2-5 \text{ MA/cm}^2 \text{ and higher in-field performance} \]

in TLAG nanocomposite

**TLAG Coated Conductors**

**Slot-die coating deposition**
15 cm, 1 µm pyrolyzed film (2 layers)

**IJP deposition**
1 µm pyrolyzed film (1 layer)

T deposit 50ºC

- Homogenous depositions and pyrolysis

**TLAG CC growth on provider 1**
(5 cm long tapes tested) - P-route

- Smooth interfaces
- \( J_c \sim 1 \text{ MA/cm}^2 \) at 77 K for 500 nm

**TLAG CC growth of nanocomposites**
provider 2 – T-route

- Small NPs (8-10 nm)
- Epitaxial and dispersed BZO NPs
2. High Throughput Experimentation by combinatorial approaches and Machine Learning algorithms

- Solution synthesis
- Y-Ba-Cu, Gd-Ba-Cu
- Machine learning
- Combinatorial inkjet printing
- Thermal annealing
- Ex-situ / In-situ characterization
- Composition, Temperature, Epitaxial Fraction, Time (s), peak intensity, Composition
Fast optimization using compositional gradient samples

Solution synthesis

YBCO GdBCO

Combinatorial IJP

Pyrolyzed film

YBCO GdBCO

Y content ~88%

Y content ~65%

Morphological changes

Y content ~88%

Y content ~65%

13 combinatorial samples were used instead of 78 regular samples to reach the same mapping resolution

A. Queralto et al., ACS Appl. Mater. Interfaces (2021)
Machine learning for predicting process magnitudes

Case: Predicting the Total Volume Deposited

IJP deposition data of 231 samples

Correlation matrix

Tree-based ensemble algorithms

A Queralto et al, to be published
3. Understanding the \((H,T,\theta)\) pinning landscape for engineered nanocomposites

\[ J_c(T) = \frac{J_{c,iso-wk}(T)}{J_c} + \frac{J_{c,iso-str}(T)}{J_c} + \frac{J_{c,aniso-str}(T)}{J_c} \]

- defect dimensionality
- orientation
- size


Lot’s of opportunities for TLAG Nanocomposites

J. Banchewski et al, to be published
Vortex pinning defects in TFA/TLAG - CSD films

- **Stacking Faults**
  - Point defects: Cu-O vacancies in the SF

- **Twin boundaries**
  - YBa$_2$Cu$_3$O$_{7-x}$ [010]

- **Nanoparticles**
  - BaZrO$_3$

- **Nanostrain**
  - YBa$_2$Cu$_3$O$_{7-x}$

- **Broken twin boundaries**
  - Intergrowth
  - Dislocation
  - NP

- **High density of SF**
  - Small epitaxial NP

- **Small a-axis crystals**

Synergistic effect of nanostrain and small NP in TLAG nanocomposites

References:
- J. Gutierrez et al., Nat Mat (2007)
- A. Llordés et al., Nat Mat (2012)
- L. Soler et al., Nat Comm (2020)
- R. Guzman et al, APLMat (2017)
**Optimized pinning landscapes**

Low and intermediate $T$ - Intermediate and high $H$

14 years of measurements

High density of isotropic defects:
- Cu-O vacancies, short SF, small NP (better Np than nanorods)

Large density of strong isotropic and anisotropic defects with long vertical coherence:
- nanoparticles, nanostrain, long nanorods, long twin boundaries
  (Nanorods and nanoparticles will add effects)

High density of anisotropic strong defects with very long vertical coherence:
- long twin boundaries, elongated nanorods, thick CSD nanocomposites
  combined with other auxiliary strong or weak isotropic defects to lessen vortex creep excitations
  (1D-2D mandatory but all defects will help to diminish creep)

2D or 1D vertical defect
- e.g., twin boundaries, nanorods, dislocations

2D or 1D horizontal defect
- e.g., stacking faults, platelets

3D defects
- e.g., nanoparticles, nanostrain

0D defects
- e.g., vacancies, atomic inclusions

F. Valles et al, *to be published*
4. Tune charge carrier density by oxygen overdoping

Pinning force $F_p = \sum_i^N f_{p,i} (B, T) \propto J_c$

$F_p \propto E_c$ condensation energy

$J_d^2 \propto n_s E_c$ (three independent experimental parameters)

$n_H$ and $E_c$ increases in the overdoped state, and consequently $J_c$ should increase

$p_{opt} = 0.16$ holes/CuO$_2$-plane: optimal doping for maximum $T_c$

$p^* = 0.19$ holes/CuO$_2$-plane: Critical doping (QCP)

Strong increase of $J_c$ in the overdoped state

$J_c(p^*) \approx \frac{1}{5} J_d(p^*) = 90 \text{ MA/cm}^2$

$J_d(p^*) \approx 500 \text{ MA/cm}^2$

$J_c(p^{\text{opt}}) \approx \frac{1}{10} J_d(p^{\text{opt}})$

$J_d(p^{\text{opt}}) \approx 330 \text{ MA/cm}^2$

Strong increase of $J_c$ with $n_H$
(x4 from $p_{\text{opt}}$ to $p^*$)

Overdoped YBCO film

Overdoping is a robust method to reach ultrahigh $J_c(H)$

A. Stangl et al, Scientific Reports (2021)
CONCLUSIONS

- Material science should help Coated Conductors to reach the market

- Cost / Performance ratio needs to decrease and high-throughput low cost growth processes with high critical current strategies can be employed

- TLAG-CSD is a non-equilibrium growth method able to grow high performance YBCO nanocomposites using low cost CSD methods at ultrafast growth rates

- Nanoengineering combinations of different defects should be used to optimize pinning landscape for each application

- Overdoping can be a robust and feasible way to further increase $J_c$ in nanoengineered Coated Conductors