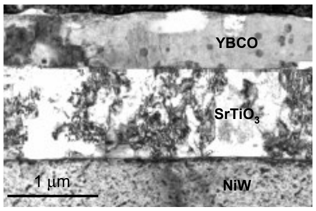
SAND2006-6630C

All Solution Deposited YBCO/$SrTiO_3$/NiW Coated Conductors

P.G. Clem, M.P. Siegal, D.L. Overmyer, C.D. Edney
Sandia National Laboratories, Albuquerque, NM 87185-1411



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Outline

All Solution Deposited 2nd Generation Coated Conductors

Motivation: robust materials and reproducible processing of YBCO tapes

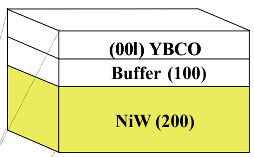
Growth of solution-deposited buffer layers
- material selection
- growth atmosphere
- control of film texture/avoiding random nucleation
  - use of seeded nucleation & liquid phases
- effect of buffer layer thickness (80 nm – 1 µm)

Solution-deposited YBCO for continuous coating
- limitations of single layer thickness: control of stresses
- control of Cu volatility
- single layer optimization: 0.3µm or 0.7 µm
- multilayer deposition

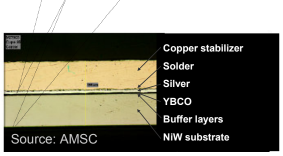
Summary

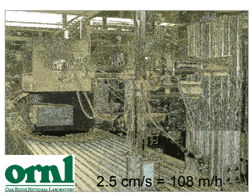
Schematic

Goal: all-MOD 2nd Generation Coated Conductors



Ideal situation:
Simplified structure
Behavior approaching PLD
RABiTS or IBAD MgO
High linear production speeds



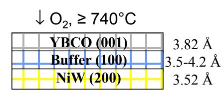


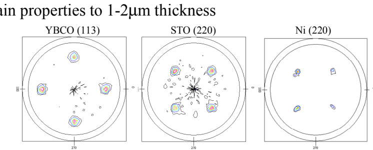
Functional roles of composite layers

Substrate
- (100) substrate cube texture
- Must be handleable

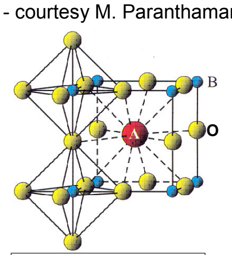
Buffer Layer
- Lattice match substrate (currently Ni-5%W)
- Provide smooth, (100) epitaxial template for YBCO
- Suppress oxidation of substrate metal

YBCO
- High structural perfection
- Maintain properties to 1-2µm thickness





Candidate buffer layers
- courtesy M. Paranthaman



Ideal ABO_3 Perovskite Structure showing the Octahedral and 12-fold co-ordination of B and A-site cations

Material	Lattice parameter (pseudo cubic) Å	% lattice mismatch vs. YBCO	% lattice mismatch vs. Ni	Property
$BaCeO_3$	4.377	13.55	21.59	Insulator
$BaZrO_3$	4.193	9.27	17.34	Insulator
$SrRuO_3$	3.941	3.08	11.17	Metal
$SrTiO_3$	3.905	2.16	10.26	Insulator
$SrTi(Nb)O_3$	3.905	2.16	10.26	SC
$LaMnO_3$	3.880	1.60	9.70	Insulator
$(LaSr)MnO_3$	3.860	0.98	9.07	Metal
$LaNiO_3$	3.859	0.98	9.07	Metal
$NdGaO_3$	3.841	0.51	8.61	Insulator
$LaAlO_3$	3.793	-0.75	7.35	Insulator
$RE(Ti,Zr)O_{3.5}$	3.6-3.9			

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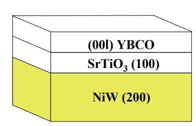
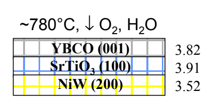
Proposed all-solution structure: YBCO//$SrTiO_3$//NiW

Why $SrTiO_3$?

- Single non-vacuum buffer layer
  - alternative: seed+difusion barrier+cap
  - others: $Gd_2Ce_2O_7$, $Gd_2Zr_2O_7$, $BaZrO_3$
  - can be grown thick (0.2µm/layer)



- Direct compatibility with BaF_2 YBCO
  - well-known compatibility of TFA YBCO with single crystal STO substrates
  - not reactive with water vapor, BaF_2
  - low STO oxygen diffusion rate
  - 0.3 µm STO → no NiO at 780°C, 20 min



$SrTiO_3$ // NiW film deposition schematic

Sr acetate → Dissolve in Acetic acid (HOAc)

Ti-alkoxide / Nb-alkoxide → Chelating Agent (Acetylacetone)

STO precursor

Diluting solvent (Methanol)

0.1-0.6M stock solution

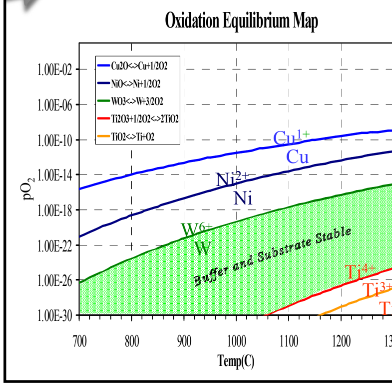
Thin layer deposition

Anneal

Process details:

- Films deposited by dip coating on Ni(5%W) substrates
- $SrTiO_3$ crystallizes at > 600°C
- Density, grain size increase with annealing temperature to 1100°C
- Microstructure is a strong function of annealing conditions

low $P(O_2)$ crystallization → $SrTiO_3$ + NiW only



$SrTiO_3$//Ni-W properties

- **Substrate oxidation → Misorientation**
- **Highly oriented**
- **Poor cystallinity and or orientation**

$SrTiO_3$ // NiW film deposition schematic

Sr acetate → Dissolve in Acetic acid (HOAc)

Ti-alkoxide / Nb-alkoxide → Chelating Agent (Acetylacetone)

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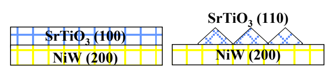
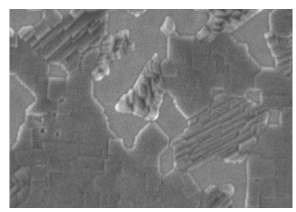
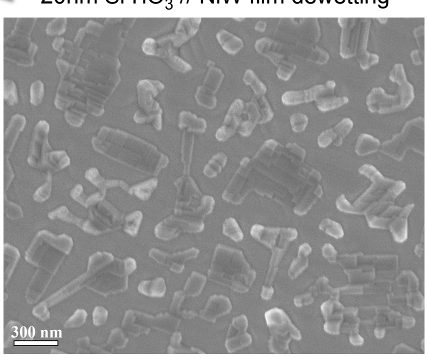
Epitaxial vs. random $SrTiO_3$ // NiW film growth





30nm $SrTiO_3$ from 0.1 M solutions on $Ni_{0.95}W_{0.05}$, annealed at 1000°C to cause dewetting

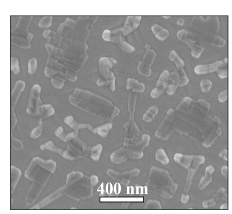
20nm $SrTiO_3$ // NiW film dewetting

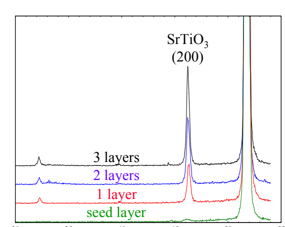


Seeded templates for $SrTiO_3$//NiW

Seeded growth of $SrTiO_3$ and $BaTiO_3$ has been shown to improve film orientation at lower temperatures [Schwartz, Clem, et al. J. Am. Ceram. Soc. **82**(9) 2359 ('99)].

- On Ni(200), lack of seeding results in randomly oriented films
- Seeded film evolution:





New buffer: $Ba_{0.2}Ca_{0.8}TiO_3$//Ni

IIA-IVB ABO_3 solid solutions enable tailoring of lattice constant, D_{ox}

	Ca (0.99Å)		Sr (1.12Å)		Ba (1.34Å)
Ti (0.68Å)	**3.82**	⟷	3.91	⟷	4.01
Zr (0.79Å)	4.01		4.13		4.19
Hf (0.78Å)	3.99		4.07		4.17

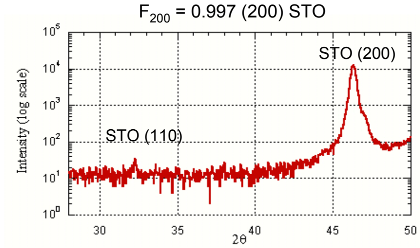
material	p-c unit cell	D_{ox}(800°C) Oxygen diffusion coefficient
MgO	4.20Å	2×10^{-19} cm²/s
$CaTiO_3$	3.82Å	1×10^{-15} cm²/s
$SrTiO_3$	3.91Å	2×10^{-12} cm²/s
YSZ	3.63Å	2×10^{-7} cm²/s
Ho_2O_3	3.75Å	7×10^{-3} cm²/s
Ni	3.52Å	

1µm $SrTiO_3$//$Ba_{0.2}Ca_{0.8}TiO_3$//Ni

$Ba_{0.2}Ca_{0.8}TiO_3$//Ni appears to be a reliable sol-gel buffer layer on Ni, NiW.

To date, this has been used as a seed/template, followed by 0.3-1µm $SrTiO_3$.

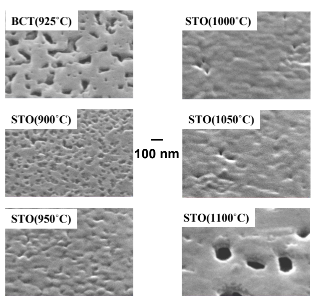
F_{200} = 0.997 (200) STO

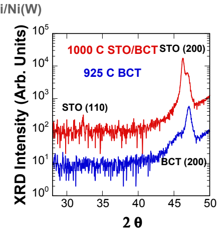


Optimizing STO/Ni(W) Growth Temperature

all anneals in wet 3% H_2/N_2, $p(O_2)$ ~ 10^{-18} atm

80 nm STO/80 nm BCT/Ni/Ni(W)





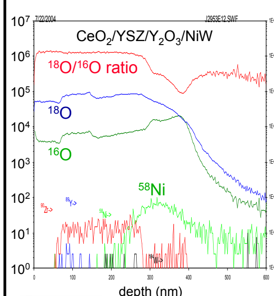
Lotgering Factor F

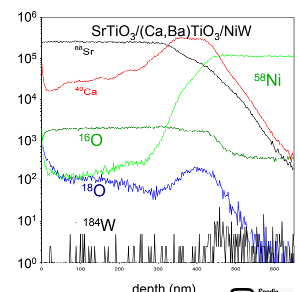
STO = 0.991

BCT = 0.995

Metal Diffusion in CeO_2/YSZ/Y_2O_3 and STO

SIMS depth profiles: both samples annealed in pure $^{18}O_2$ at 740 °C for 30 minutes

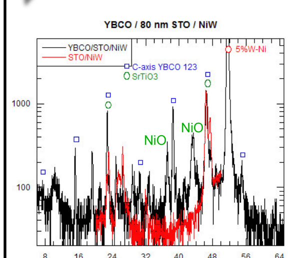


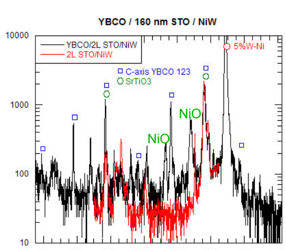


American Superconductor

Ni diffusion limited to ~100nm at buffer interface for both

TFA YBCO// 80-160nm $SrTiO_3$//NiW

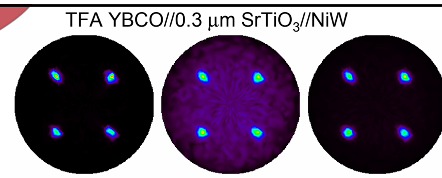




$SrTiO_3$//NiW substrate

STO Δφ = 5.8 degrees

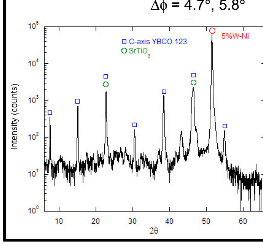
TFA YBCO//0.3 µm $SrTiO_3$//NiW



Ni-5%W(220) Δφ = 4.7°, 5.8°

STO(220) Δφ = 5.9°,7.4°

YBCO (013) Δφ = 5.7°,7.4°



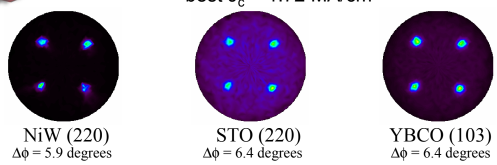
champion J_c = 1.7 MA/cm²

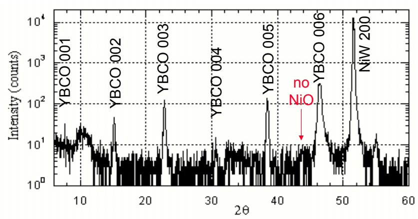
Process method:
TFA/DEA YBCO
RTA pyrolysis (30 seconds)
1 Torr conversion (4-8 minutes)

$SrTiO_3$//NiW substrate

STO Δφ < 6 degrees

Current 0.6 µm TFA YBCO//0.6µm $SrTiO_3$//NiW best J_c = 1.72 MA/cm²





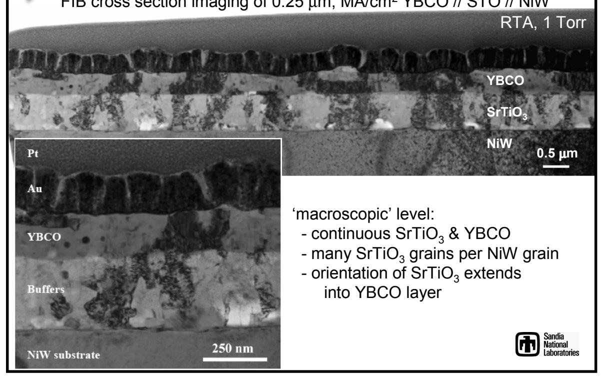
Process method:
TFA/DEA YBCO
RTA pyrolysis
1 Torr conversion

$SrTiO_3$//NiW substrate

TEM of all solution YBCO // $SrTiO_3$ // NiW:

images: T. Holesinger, LANL

FIB cross section imaging of 0.25 µm, MA/cm² YBCO // STO // NiW

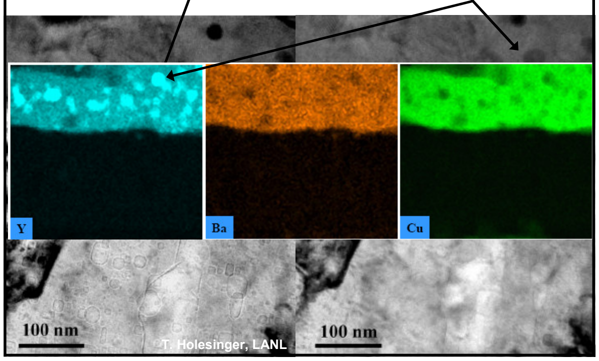


'macroscopic' level:
- continuous $SrTiO_3$ & YBCO
- many $SrTiO_3$ grains per NiW grain
- orientation of $SrTiO_3$ extends into YBCO layer

TEM reveals nanoscale YBCO//STO//NiW features:

a) nanoscale porosity within STO

b) Y_2O_3 precipitates in YBCO, little apparent porosity



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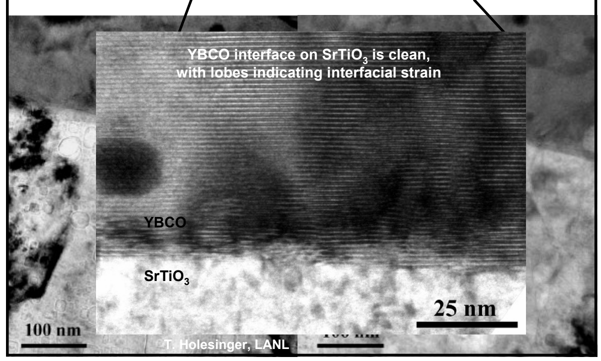
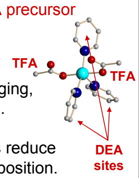
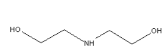


DEA-modified TFA method for YBCO

Dissolve Ba, Y, and Cu acetates in trifluoroacetic acid
↓
Add diethanolamine
↓
Dilute with solvent
↓
YBCO stock solution (M=0.2)

- Diethanolamine (DEA) prevents film buckling during low $P(O_2)$ processing.
- DEA stabilizes the Cu TFA precursor
  - *Increased solubility*
  - *Reduced volatility*
- DEA likely permits Cu bridging, and forms larger oligomers.
- Increased steric bulk helps reduce volatility and slows decomposition.



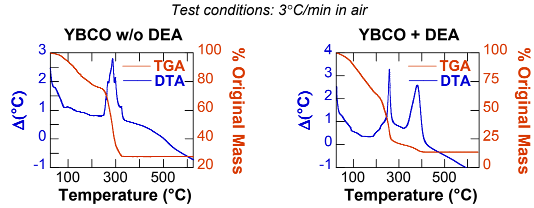


DEA

DEA-TFA YBCO Process

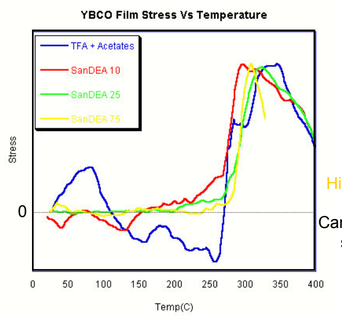
Thermal Decomposition of YBCO: Effect of DEA addition

Test conditions: 3°C/min in air



- DEA addition spreads out precursor decomposition range, compared to use of TFA alone.
- The two stage decomposition preserves film integrity. (no cracking, buckling, or porosity development)

in situ stress measurements of modified TFA-YBCO precursor chemistries



TFA route (0.1 µm)
↓
DEA added (0.1 µm)
↓
PVP added (0.25 µm)
↓
Higher viscosity (0.75 µm)

Can reduce organic removal step to 10-20 seconds

PVP – polyvinylpyrrolidone,
H. Kozuka et al., J.Am.Ceram. Soc. 86(1), 33 (2003)

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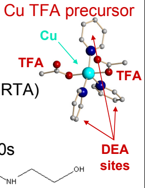
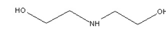
DEA-modified TFA route for YBCO

Dissolve Ba, Y, and Cu acetates in trifluoroacetic acid
↓
Add diethanolamine
↓
Dilute with solvent to 0.2M YBCO solution

- Diethanolamine stabilizes Cu TFA precursor
  - *- Increased solubility*
  - *- Reduced volatility*
- Rapid thermal annealing (RTA) pyrolysis (~ 100°C/s)
- Pyrolysis dwell time: 20-60s
- Method may enable RTA multilayering of 0.25-0.75µm films (4 x 0.6µm → 2.4µm)





coating → pyrolysis → crystallization → oxygenation
2 cm/s (72m/h) 60s 2 - 10 min 30 min

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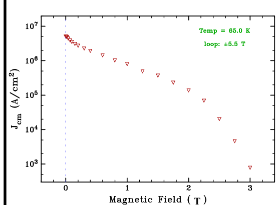
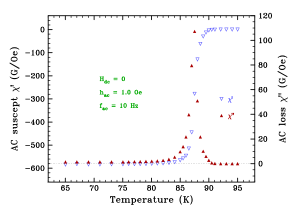
0.3 µm TFA-DEA YBCO//MOD $SrTiO_3$//$LaAlO_3$ J_c = 4.4 MA/cm²





$J_{c\text{-}m}$ (65K, 0T) = 5 MA/cm²
$J_{c\text{-}m}$ (65K, 1T) = 0.9 MA/cm²

T_c = 88.5 K

Three approaches to thicker films:
- thick single layers (0.7 µm)
- multipyrolysis
- multicrystallization

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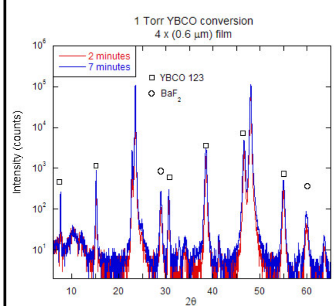
Thicker YBCO film deposition by RTA multipyrolysis: 4 x 0.6µm YBCO



J_c (77K) = 2 MA/cm² obtained for 0.6µm RTA layers on STO//LAO

2.4 µm YBCO deposited by multilayering four 0.6 µm films with intermittent 60 s RTA

Max YBCO signal at 7 minutes; at 10, 20 minutes (20Å/s), YBCO intensity ↓, 2nd phase intensity ↑

Optimizing film composition:
- multipyrolysis interface is Cu-rich
- effort continuing on $SrTiO_3$//NiW
- no J_c > 300 kA/cm so far

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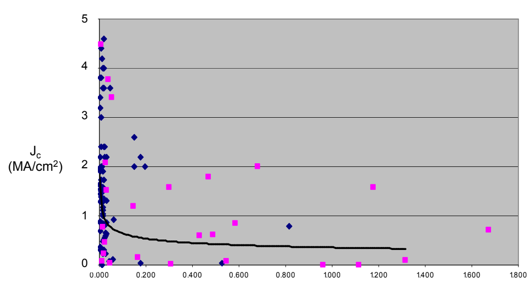
Vacuum converted of MOD YBCO/MOD STO/LAO J_c vs. normalized BaF_2 content



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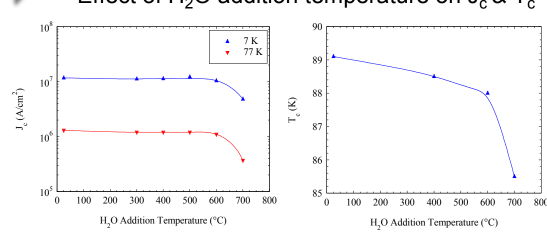
Multi-crystallization feasibility: Effect of H_2O addition temperature on J_c & T_c



- H_2O vapor introduction temperature up to 600°C has little effect on YBCO film J_c at 7 K and 77 K.
- T_c gradually decreases as H_2O addition temperature 600 °C.

∴ Low water, low TFA-content solutions enable YBCO multicrystallization.

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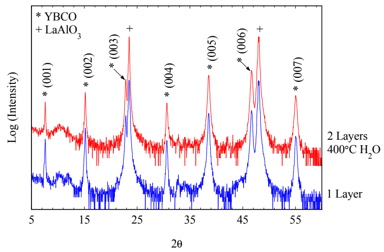
Ex-situ crystallization of a second YBCO layer by delayed water introduction



	1 Layer	2 Layers
Jc (77 K):	1.3 MA/cm^2	1.1 MA/cm^2
Jc (7 K):	12 MA/cm^2	10 MA/cm^2

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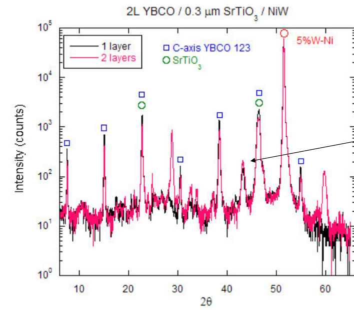
Multicrystallization of TFA-DEA YBCO on STO/NiW



BaCuO phase 'cuprite'?

J_c ~ 400 kA/cm^2

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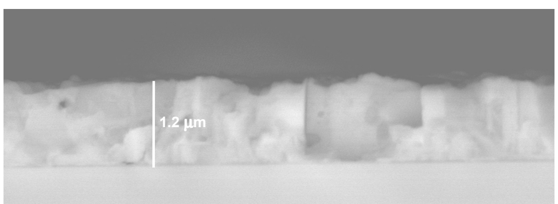
Thick (1.2 µm) TFA-DEA YBCO from multicrystallized layers (not optimized)



t = 1.2 µm, J_c < 1 MA/cm^2

[Cu] being optimized and extended to 4 x 0.6 µm

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Summary

All-solution deposited coated conductors are feasible:

1) Dense $SrTiO_3$ may be deposited on NiW at 1000°C at low PO_2
2) Metal and oxygen diffusion are limited by STO or CeO_2/YSZ/Y_2O_3
3) TFA/DEA YBCO may be RTA annealed and converted at 20 Å/s
4) All-solution YBCO//STO//NiW : 0.6µm, J_c up to 1.72 MA/cm^2
5) Multilayering+vacuum conversion an approach to 2µm+ TFA films

Key work: optimizing multilayered YBCO on thick (0.25µm) STO

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