Exploring the high field potential of Nb$_3$Sn Strand

* Charlie Sanabria is now at LBNL
Conventional Nb$_3$Sn $J_c$ has plateaued below FCC specifications

In this talk we examine the limitations and potential of PIT, RRP and APC Nb$_3$Sn strands to reaching FCC targets.

Notes:

- Nb$_3$Sn data from Parrell et al. 2004 http://dx.doi.org/10.1063/1.1774590
- Bi-2212 strand manufactured by OST

For Nb$_3$Sn recent improvements have been in $D_{eff}$, RRR and yield = Better magnet conductor
Recent $\text{Nb}_3\text{Sn}$ progress . . .

1. **PIT** - the **bundled barrier** has enabled **higher Sn** that allows **better A15**. $J_c > 2600 \text{ A/mm}^2$ at 12 T for $D_{\text{eff}}$ 36 $\mu$m wires. However, tubes have integrity issues and there is significant core centroid drift, suggesting that $J_c$ could be much higher.

2. **RRP** (internal Sn) - **Nausite control HT**: significant improvement for small sub-elements, **enables high $J_c$ and reduction in $D_{\text{eff}}$**.

3. Analysis of **dopant sites** suggest potential for high field gains in new alloys.

4. **APC** – multiple groups are working on strands that exploit the refined grain size by ZrO$_2$ precipitates demonstrated at OSU. The challenge now is to make sure expands beyond the degraded literature values
   - Long term, all the problems of PIT and RRP must be addressed with APC
   - Can enough A15 be made?
   - Will it have high enough $H_{c2}$ across the A15 layer?
   - Will properties be achievable at appropriate cost and yield?
PIT is held back in three major ways:

1. About 1/4 of the A15 formed is LG A15 (13% of 53% area)
2. Much tube remains unreacted (21-24%)
   - RRP is more effective with only ~10% un-reacted Nb
   - BEAS and CERN developed a new wire (bundle barrier) to drive up $J_c$ without loss of RRR – previous talk
   - Bundle barrier allows more tin, more reaction

What we found compared to non-bundle barrier wire
1. $J_c$ increases >5%, RRR was maintained
2. SG/LG A15 ratio decreased
3. Tin react-through and tube breakage
   - When the tube is compromised the A15 area loss can be catastrophic.
   - Sn react-through limits A15 area growth.
   - These are quite long, up to centimeters in length, occurring more in outer filaments

Only small grain A15 carries current
- Large and core grains of A15 do not carry any current

New wire designs have higher $J_c$, but lower SG/LG A15 ratio and higher layer $J_c$ – more Sn in the SG layer?

- Similar filament size to compare,
- $J_c$ over unreacted filament area
- Bundle barrier considered part of the Cu

*was previously reported over reacted area as $J_c = 2237$

<table>
<thead>
<tr>
<th>Sample description</th>
<th>Recommended HT (temp/dwell time)</th>
<th>$d_{eff}$</th>
<th>$J_c$ (A/mm²)</th>
<th>$J_c$ SG-layer (A/mm²)</th>
<th>Nb</th>
<th>Total A15</th>
<th>core A15</th>
<th>LG A15</th>
<th>SG A15</th>
<th>SG/LG ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Φ= 0.78 mm</td>
<td>620/100+640/90</td>
<td>38</td>
<td>2444*</td>
<td>6080</td>
<td>23.4%</td>
<td>56.0%</td>
<td>2.5%</td>
<td>13.3%</td>
<td>40.2%</td>
<td>3.0</td>
</tr>
<tr>
<td>Luca 0.70 mm</td>
<td>280/20 + 620/120 + 640/160</td>
<td>36</td>
<td>2658</td>
<td>6515</td>
<td>21.0%</td>
<td>59.2%</td>
<td>2.4%</td>
<td>16.0%</td>
<td>40.8%</td>
<td>2.5</td>
</tr>
</tbody>
</table>

% change with new barrier + more Sn - +8.8% +7.2% -10% -10% +20% -16%
Bundle Barrier wires have large, radial variation of filament area

Filament area

- Mean = 1290 μm²
- Median = 1298 μm²
- Std. Dev = 41 μm²

~12% reduction in area from inside to outside

• Important consideration during data analysis:
  Filament diameter increases by 2-3 μm from inside to outside
Bundle barrier allows exploration of higher Sn designs but architecture impacts filament integrity

- A15 in contact with Cu
- Kirkendall void: indicates a major Sn leak

<table>
<thead>
<tr>
<th></th>
<th>Med Sn</th>
<th>High Sn</th>
<th>Very High Sn</th>
</tr>
</thead>
<tbody>
<tr>
<td>“Luca”</td>
<td>“Bernardo”</td>
<td>“Bernardo 2”</td>
<td>“Lucio”</td>
</tr>
<tr>
<td>0.85 mm</td>
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<td></td>
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</tbody>
</table>

Breached:
- A15 contacts Cu, RRR decreases

Breached with Kirkendall void:
- Severe Sn leak, A15 vol. suppressed

Unbreached:
- Fully continuous barrier

Best $J_c$ 2658*

Worst $J_c$ 2310

$J_c (12 T, 4.2 K)$ A/mm²
Centroid drift: Causes premature breach

Two main issues:

- Outer filaments become more elliptical as the wire is processed to final size [1][2][3].
- The powder core deforms at a different rate than the NbTa tube housing it, causing the centers to drift from one another.
  - This creates an uneven reaction front with a thick and thin side of the diffusion barrier
  - We measure the centroid drift as the distance between the centroid of the entire filament (orange) and the centroid of the A15 layer (green).
  - The centroid drift is normalized to the filament $d_{\text{eff}}$ derived from its area

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Filaments above 3% drift have a 25% failure rate, showing a Kirkendall void and reduced A15 volume.

```
Worst $J_c (12T,4.2K)=2310$
```

```
24 filaments > 3%
```

```
“Very High Sn” 0.7 mm, 2 worst leaks
```

```
5.3% 4.2% 3.9% 3.9% 3.7% 3.7%
```

```
Distance from wire center (μm)
```

```
Normalized centroid drift (% diameter)
```

```
Centroid drift of select filaments
```

```
Leaked filaments with Kirkendall void
```

```
Normalized centroid drift (% diameter)
```

```
“Very High Sn” A15%
```

```
(LG+SG) A15 area%
```

```
Normalized centroid drift (% diameter)
```

```
Centroid drift of select filaments
```

```
0.1% 5.3% 4.2% 3.9% 3.9% 3.7% 3.7%
```
Potential for RRP Nb₃Sn

- Nausite-control HT (NCT) provides breakthrough in $J_c$ for small $D_{\text{eff}}$
- Quaternary doping may help achieve FCC targets

Presented at FCC Week ’17
NCT heat treatments longer but much higher $J_c$

**Standard**

- Temperature: 215°C, 400°C, 665°C
- Time: 0, 100, 200 hours

**New**

- Temperature: 350°C, 665°C
- Time: 0, 100, 200, 300, 400, 500 hours

**Alternative**

- Temperature: 350°C, 620°C
- Time: 0, 200, 400, 600, 800, 1000 hours

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**Graph:**

- $J_c$ (A/mm²)
- B (T)
- Temperature (°C)
- Time (h)

**Legend:**

- 16315-5 (Ds = 40 μm) Alternative Heat Treatment
- 16315-5 (Ds = 40 μm) New Heat Treatment
- 16315-5 (Ds = 40 μm) Standard HT

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Work by Charlie Sanabria (CEC-ICMC’17) Accepted for publication in SuST

Charlie is continuing his RRP work at LBNL
In recent years $J_c$ in Nb$_3$Sn wires has been optimized at 12 T for the HiLumi LHC upgrade that requires balancing $H_{c2}$ and GB density for highest vortex pinning.

But FCC requires optimization of $J_c$ (16 T) that shifts the balance toward higher $H_{c2}$ and requires maximum uniformity across the whole Nb$_3$Sn layer.

- Can Ta and Ti be combined to improve disorder scattering?
  - Where do the Ta and Ti atoms sit in the Nb$_3$Sn structure?

Nb$_3$Sn is usually doped by Ti or Ta to enhance $H_{c2}$ by disorder scattering.
Where are the Ta and Ti going in the A15?

✓ An early experiment seemed to indicate that both dopants substitute on the Nb site


✓ More recently the differing effectiveness of Ti and Ta in maximizing $H_{c2}$ was interpreted as due to different doping sites (Ta on Nb, Ti on Sn)

Sn is more uniform and the *in-field* $T_c$-distributions are higher and more homogeneous with Ti

Slightly over-reacted (662-665°C/48h)

Doping with Ti produces a more homogeneous A15 composition and a tighter distribution of $T_c$

But double-doped has better $T_c$ distributions at 15 T

- Three 54/61 standard Sn, slightly under-reacted (640°C/40h) strands with different doping:
  - Only Ta (Ta#1),
  - Ta+Ti,
  - Only Ti (Ti#1)

Double doping $\Rightarrow$ high $H_{c2}$, $H_k$  
$\Rightarrow$ potential for FCC

**In-field Ta+Ti sharper than either Ti or Ta**

Double doping $\Rightarrow$ high $H_{c2}$, $H_k$  
$\Rightarrow$ potential for FCC

S.M. Heald, C. Tarantini _et al._,  
Scientific Reports 8, 4798 (2018)
Dopant sites identified by Extended X-ray Absorption Fine Structure

- It is sensitive to the local environment of a specific element:
- It works at x-ray energies above the absorption edge energy of the element under study
- Photoelectrons emitted by the element under study are scattered by the neighboring atoms revealing the local structure

**Binary phase example**

- **Nb site** has three closely spaced coordination shells ⇒ **3-peak structure**
- **Sn site** has a single nearest neighbor shell ⇒ **single peak**

EXAFS performed by Steve Heald, Advanced Photon Source, Argonne National Lab.

Where is Ta? EXAFS says on both sites!

- **The best fit requires Ta to be on both sites:**
  - **32-30% Ta on the Sn site in the 54/61**
  - **21% Ta on the Sn site in the 108/127**

**Ti is on the Nb site only!**

EXAFS performed by Steve Heald, Argonne National Lab.

Chemistry(EDS)+EXAFS: “High-Jc” Ti is off-stoichiometry

Ti samples are particularly Sn deficient

Thus Nb:Sn stoichiometry is *not* the defining quality factor for doped Nb₃Sn!

The dopant sites do not explain the ratio.
Other defects must be involved: vacancies or antisites

Besson *et al.*, Phys.Rev. B 75, 054105 (2007): “The defect structure is found to be of antisite type, with small amounts of Nb vacancies, and Sn vacancies showing a trend towards instability.”

Vacancies are very rare in Nb₃Sn
⇒ Off-stoichiometry occurs by antisite disorder
Strong effect of disorder on $H_{c2}(0)$

- We estimated the long range order parameter, $\eta$, considering the chemical composition, the dopants sites and the antisite disorder.

Using $H_{c2} \propto \frac{T_c}{\tau}$ and $\frac{1}{\tau} \propto (1 - \eta^2)$


$\tau$, scattering time

- The Ta samples are the most ordered ($\eta$ highest values).
- The Ti doped samples are more disordered than Ta.
- The Ta+Ti sample is more disordered than both Ta and Ti 54/61.

Except for Ta#1 (most under-HT’ed), samples follow a linear trend despite the large number of approximations $\Rightarrow$

Both dopants and antisites are needed to increase $H_{c2}$

The promise of APC* conductors

* Here APC = “Added Pinning Centers”

• $H_{c2}$ in previous non-doped APC conductors was depressed w.r.t binary Nb$_3$Sn: $\sim 20$ T vs expected 24 T (Binary PIT) – $H_K(4.2$ K)

• We found this to be true for wires made by Supramagnetics (Motowidlo et al. SuST 2017: $H_K 18-20$ T.

• Can we increase $H_{c2}$, still maintaining the 50-100 nm grain size advantage?

Grain size of Nb$_3$Sn between 50-100 nm
Monofilament wire testbed with custom Nb-Ta-Zr alloys

Details:
- **In-house alloy rod:**
  - Arc melted, Nb-7.5wt%Ta (Nb-4Ta) with Zr (>1 at %).
  - For comparison Nb-4Ta composite also fabricated.
- **Cu-Sn (SnO$_2$)**
  - Cu-Sn mixed in the weight ratio to form Cu$_6$Sn$_5$.
  - SnO$_2$ addition < 0.01 moles/volume.
  - Composites made with and without SnO$_2$
- **Ta barrier**
  - Commercial seam welded Ta tube, no initial heat treatment
Fine grain Nb₃Sn observed in monofilament with Zr

High temperature grain growth restricted even without O₂
Ternary additions boost pinning force

1. With or without SnO$_2$: **Maximum pinning force** is obtained in $+1X$ and $+3Zr$.
2. SnO$_2$ suppresses maximum, except for $+3Zr$.
$H_{\text{max}}$ vs. $H_{\text{irr}}$: $O_2$ depresses $H_{\text{irr}}$ (except 1x)

- The field peak is unchanged or shifted at higher field with Nb4Ta and Nb4Ta1Zr, but it is strongly shifted at lower field with Nb4Ta1X and Nb4Ta3Zr
- In most cases SnO$_2$ suppresses $H_{\text{irr}}$ (Nb-4Ta still has the best $H_{\text{irr}}$)
Key Points

- PIT and RRP are close to final optimization but do not meet FCC targets
  - RRP: Improvements will have to come from greater uniformity of $H_{c2}$ across the layer
    - Ti is better than Ta but mixed Ti and Ta may be even more uniform judged by $C_p(15 \, T)$ measurements
  - PIT: Problem of too much LG A15 has not been resolved but higher Sn in the core can raise the $J_c$ of the SG A15 towards RRP levels
    - To get these gains in practice requires new approaches to Nb tube uniformity
- APC is promising but published results all show strong reduction of $H_{c2}$, even if finer grains show positive distortion of the pinning force curve
  - Exploration of variable Zr with Nb-4Ta base show less reduction of $H_{c2}$ and clear complexities about having added O in the system.
Where next?

• Where next?
  – RRP HT focus for now is with Sanabria at LBL
  – PIT needs feedback from Bruker and its users
  – Site disorder and $H_{c2}$ – is quaternary doping better than ternary for optimizing $H_{c2}$ throughout the WHOLE A15 layer?
  – APC: need to understand competing effects of reduced grain size and ZrO$_2$ insulating point pinning in raising the field, $h_{\text{max}}$, at which $F_p$ peaks
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