Much higher $J_c$ Nb$_3$Sn by Hf alloying of Nb-Ta: A route for achieving the FCC conductor targets applicable to all conductor types

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Key points of this talk: 1

Background:

- In 2018 we compared 1Hf and 1Zr alloyed Nb4at%Ta with and without an O source, finding that O-free Nb-4Ta-1Hf suffered no degradation of $H_{\text{irr}}$ (4.2 K)~23 T and the highest layer $J_c(16 \text{ T}, 4.2 \text{ K})$ of ~5000 A/mm$^2$ (RRP non-Cu $J_c$ of $0.6 J_{clayer}$) which exceeds FCC specification
  - Our study showed that internal oxidation is not the only route to fine grain A15, perhaps opening an avenue to simplify the manufacture of FCC magnet conductors

What have we done?

- Full range (30 T) VSM measurements on our own monofilaments and the first multi-filament Nb-Ta-Hf conductor (PIT made by HyperTech Research Inc (HTRI)).
- Compared drawability of Nb-Ta-Hf and the well known Nb-Ta.
- Performed a recrystallization study of Nb-Ta-Hf, and Nb-Ta composite
Key points of this talk: 2

What did we find?

– Verified shift in $H_{\text{max}}(4.2 \text{ K})$ above 5 T without suppression of $H_{\text{irr}}(4.2 \text{ K})$ $\sim$23.5 T.
– Found that Nb-Ta-Hf work hardens similarly to Nb-Ta alloy.
– Found that BOTH ASC/NHMFL and HTRI-sourced Nb-4Ta-1Hf prevent recrystallization of worked Nb-Ta-Hf during Nb$_3$Sn formation, setting up conditions for fine grain A15 phase formation.

So what?

– Recrystallization during A15 formation is prevented in Nb-Ta-1Hf, greatly enhancing $J_c$.
– Although, internal oxidation may offer additional benefits, we believe 50-75 nm grains start with inhibiting recrystallization of the starting alloy before A15 formation.
– Avoiding extra oxygen additions could greatly simplify application to existing Nb$_3$Sn production wires.
2018 Background: Potential of Nb-4Ta-1Hf alloy for high $J_c$ (>1500 A/mm$^2$, 16 T, 4.2 K) Nb$_3$Sn conductor was established with monofilament wires.

- Ta doped RRP, Ta-Hf, and Ta-Zr all have comparable $H_{irr}$ (4.2 K) ~23 T.

- $F_{p\text{max}}$ is substantially higher in Nb-Ta-Hf than Nb-Ta-Zr or Nb-Ta.
- Position of $F_{p\text{max}}$ ($H_{\text{max}}$) shifted to 5.8 T in the case of Nb-Ta-Hf without SnO$_2$.
- Enhanced $F_{p\text{max}}$ and $H_{\text{max}}$ above Ta-doped RRP occurs in all cases.

**Background:** 16 T Layer $J_c$ estimates of Nb-4Ta-1Hf conductor without SnO$_2$ exceed 3500 A/mm$^2$

<table>
<thead>
<tr>
<th>Alloy</th>
<th>SnO$_2$</th>
<th>$J_{clayer}$ (A/mm$^2$) 12 T</th>
<th>$J_{clayer}$ (A/mm$^2$) 16 T</th>
<th>Eq. RRP non-Cu $J_c$ (A/mm$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nb-Ta</td>
<td>No</td>
<td>3209 ± 916</td>
<td>1245 ± 355</td>
<td>747 ± 213</td>
</tr>
<tr>
<td>Nb-Ta</td>
<td>Yes</td>
<td>2237 ± 639</td>
<td>1003 ± 286</td>
<td>602 ± 172</td>
</tr>
<tr>
<td>Nb-Ta-Zr</td>
<td>No</td>
<td>3545 ± 1012</td>
<td>1281 ± 366</td>
<td>768 ± 219</td>
</tr>
<tr>
<td>Nb-Ta-Zr</td>
<td>Yes</td>
<td>5017 ± 1433</td>
<td>1684 ± 481</td>
<td>1010 ± 289</td>
</tr>
<tr>
<td>Nb-Ta-Hf</td>
<td>No</td>
<td>9609 ± 2744</td>
<td>3714 ± 1061</td>
<td>2229 ± 636</td>
</tr>
<tr>
<td>Nb-Ta-Hf</td>
<td>Yes</td>
<td>8523 ± 2434</td>
<td>3093 ± 883</td>
<td>1856 ± 530</td>
</tr>
</tbody>
</table>

- Ta-Hf doping without SnO$_2$ opens up existing conductor architecture options (**RRP, PIT and Distributed Tin, bronze-route**) without need to incorporate O source.

Nb-4Ta-1Hf rod does not recrystallize during A15 formation, unlike Nb-4Ta (or pure Nb used in RRP wires)

- Fine grain alloy rod leads to halved Nb₃Sn grain size.
- Hf appears to increase the recrystallization temperature of Nb-4Ta
- Sn penetrates Nb alloy by preferential grain boundary diffusion
Multi-filament conductor made by HTRI (without SnO$_2$) confirms the monofilament result of $H_{\text{max}}$ shift beyond 5 T, $H_{\text{irr}}$ (4.2 K) of 23.5 T.

The Hyper Tech Nb-4Ta-1Hf tubes were independently sourced.
Hyper Tech Hf conductor with SnO$_2$ - $H_{\text{max}}$ shift to 9 T, suggesting additional oxide pinning centers add to fine A15 grain pinning.

Non-standard shape suggests inhomogeneity. But $H_{\text{irr}}$ (12 K) is high.

Thanks to Xingchen Xu (FNAL), and Xuan Peng (Hypertech)
Development of multi-filament conductors with Nb-Ta-Hf alloys

• Can Nb-4Ta-1Hf alloy be drawn to large strains?

• Does the worked multi-filament microstructure survive the Nb$_3$Sn reaction heat treatment temperature?
Cu/Nb-4Ta and Nb-4Ta-1Hf multifilament wires were drawn to large strains.

- 30 g ingot arc-melted ingots.
- Ingots swaged to 3 mm diameter rod.
- Cu-sheathed, restacked and drawn to larger strains.

The total true strain \(\ln \left(\frac{A_0}{A}\right)\) is ~7

Note that these 30 g ingots could not be properly recrystallized and they did not deform uniformly at large strains.
Worked microstructure persists in Nb-Ta-Hf after 3 h at 650°C-700°C. Nb-Ta shows new grain growth already at 600°C.
Recrystallization of Nb4Ta is evident in hardness
HyperTech tube confirms delayed recrystallization in Nb-Ta-Hf alloy during Nb$_3$Sn reaction heat treatment.

$625^\circ C/740h$

$675^\circ C/385h$

Hf appears to raise Nb-4Ta recrystallization temperature significantly.

Thanks to Xuan Peng (Hyper Tech) and Xingchen Xu (FNAL) for wire sample.
Workability of Nb-4Ta, Nb-4Ta-1Hf IS limited by initial cast microstructure.

- Nb-4Ta (as cast)
  - Fracture at strain of ~9

- Nb-4Ta-1Hf (as cast)
  - Fracture at strain of ~10.5

- Large mm size starting grains lead to non-uniform deformation of the alloy rod in a soft Cu sheath.

- Large scale industrial Nb-4Ta-1Hf alloy from HC Starck Inc expected in July with fine recrystallized grain structure

Nb-4Ta ATI rod is in progress.
Hardening behavior of Nb-4Ta-1Hf is similar to Nb-4Ta.

- Nb-Ta-Hf has a higher hardness compared to Nb-Ta.
- This does not limit the deformability of the conductor.
Is 1at% Hf addition to a Nb-4Ta alloy optimum?
Monofilament studies show benefit to pinning force by doubling the Hf content to 2 at%. $H_{\text{max}}$ (4.2 K) is higher without suppression of $H_{\text{ irr}}$ (4.2 K).

- Consistent peak shift above 5 T in Nb-4Ta-2Hf conductor.
- Considering industrial alloy of Nb-4Ta-2Hf now.
- Increasing Hf content could provide a control knob for delaying recrystallization.
Conclusions

• **Hf additions to Nb-4Ta provide \(<100\text{ nm}\) Nb\(_3\)Sn grain size due to additional GB diffusion paths provided by enhanced recrystallization temperature.**
  – Demonstrated in both ASC monofilaments and Hypertech multifilament conductors for Nb\(_3\)Sn reaction heat treatments at 625°C - 675°C.
  – Enhanced \(H_{\text{max}}\) (4.2 K) and unsuppressed \(H_{\text{irr}}\) (4.2 K) is verified by Hyper Tech multifilament conductor
  – Hyper Tech wires with Sn-oxide may provides interesting opportunities also.

• **Multifilament Nb-4Ta-1Hf failed at a higher strain of 10.5 than the strain of 9 in Nb-4Ta. Cast grain structure was the cause in both cases.**
  – Larger batches of alloy with controlled grain size from commercial vendors are expected imminently in progress.
  – Rod microstructure evolution during A15 heat treatment and its effect on fine grain Nb\(_3\)Sn formation in Hf based alloys needs exploration.

• **Nb-Ta-Hf conductors provide avenues in various architecture types.**
  – FG Nb\(_3\)Sn by optimization of Hf doping provides a direct avenue to implement the new alloy in RRP, bronze route, and PIT configurations.
  – Additions of oxygen as advanced by Ohio State-Hypertech-Fermilab seems to enhance \(H_{\text{max}}\) and are being evaluated in PIT conductor form.