Multiple characterization approach to enhance the performance of PIT Nb_3Sn conductors

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Abstract*— Optimization of critical current density, J_c, in modern Powder In Tube (PIT) Nb_3Sn superconducting wires is both a question of maximizing the amount of Nb_3Sn and its quality, while avoiding any diffusion barrier (DB) breakdown that will poison the stabilizing and protecting Cu. Multiple superconducting and microstructural characterizations have been used to follow the progression of the A15 reaction across the Nb(Ta) tubes and the residual annulus of unreacted tube that acts as the DB. We found that the non-Cu J_c (12T, 4.2K) reached 2400-2500 A/mm^2 for heat treatments (HT) between 620°C and 670°C without degradation of RRR below 100. This J_c was developed by conversion of 52-55% of the non-Cu package to A15, with DB and core residues of about 25% and 22% respectively. However about a quarter of the A15 in the high J_c samples was A15 formed at the interface between the core and the DB that has grain size well over 1 µm. This large grain A15 fraction is rather independent of HT condition, representing 14%-16% of the total non-Cu after short reaction time when very little fine grain A15 has formed and J_c is low. By comparison with RRP conductors, we found that this conductor produced only about 2/3 as much current carrying A15 as in recent RRP conductors. However the J_c referred to the fine-grain layer is about 20% higher than Rod Restack Process (RRP) conductors. Our detailed evaluation of the A15 layer by magnetization, specific heat and microstructure leads us to conclude that the large grain A15 layer plays little or no role in the transport J_c.

I. Introduction

Diffusion barrier integrity is crucial to maintaining good RRR for magnet stability needed in high field magnet accelerators like the LHC upgrade [1], however, the DB in PIT wire is the remnant unreacted tube [2] and thus the degree to which the filament can be reacted is limited by the degree to which the filaments can be safely reacted while retaining a continuous protective ring of Nb(Ta) that prevents Sn poisoning of the Cu. As the reaction proceeds radially from the roughly circular cores, any geometric distortions of the filaments leads to variations in the thickness of the DB and can ultimately lead to RRR degradation. Our previous work [3] has shown that modern PIT (and RRP) conductors show shape degradation that increases with radial position (Fig 1). With only a handful of diffusion barriers in outer filament rings breaking, the RRR degradation can rapidly drop to below 150. These Bruker PIT samples from CERN contain 192 Nb7.5wt.%Ta tube filaments of remarkable uniformity, but there is some non-uniform deformation, particularly in the outer rings. In addition we report the % volume fraction through digital analysis of transverse cross sections, analyzing the different A15 grain structures that form, and how they relate to other measured values such as J_c. Here we quantify this behavior.

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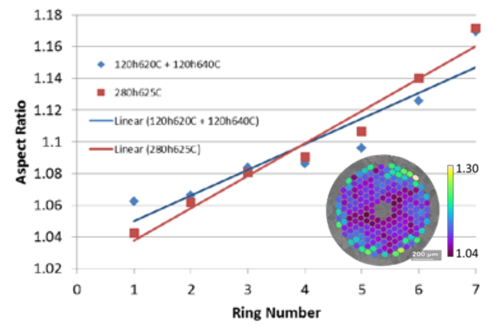
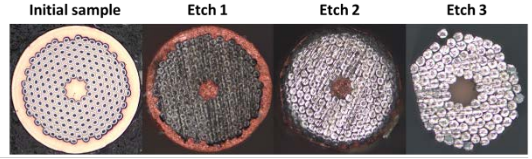


Fig. 1. Average aspect ratio as a function of radial position in the wire via filament ring number. All 192 filaments of both samples are included in the average of these 1mm diameter wires



	Initial sample	Etch 1	Etch 2	Etch 3
120h620C+ 120h640C	175	157	127	172
280h625C	166	138	93	173

Fig. 2. Below each cross section is the RRR measured at that stage it is clear that RRR is not monotonic with amount of Cu removed. Etch 3 is presented at slightly higher magnification.

II. Procedure

Digital imaging using a Zeiss EsB FESEM coupled with digital analysis software allows rapid and accurate evaluation of the positional variation and geometric distortion, ring by ring. In addition, grain size is measured for each layer as the grain boundaries are the dominant pinning mechanism for Nb_3Sn [4]. To determine the local impact on RRR by barrier breakthrough, we employ an acid etching procedure and subsequent resistivity measurements (Fig. 2).

First, 4 point contacts are made with silver epoxy and the initial resistivity measurement is taken. Then the silver epoxy is removed and the sample is immersed in a dilute nitric acid bath for 5-10 minutes. After the etch, 4 point contacts with silver epoxy are made again and the measurement is repeated, this time with an annulus of Cu removed. After another acid treatment the entire outer annulus of Cu is removed and we begin to see the filament pack. Lastly the copper surrounding the external 3 rings is removed, and filaments are carefully extracted during the process, keeping note of which ring they come from. These filaments are retained and analyzed further to obtain transport current values as a function of radial position. A similar digital imaging and analysis technique is used in determining the % volume fraction of the different phases present. By using algorithms to accurately reproduce a threshold between the different layers of Nb and A15 the % volume fraction can be calculated to high precision for easy comparison between samples.

III. Results and Discussion

A. Macro-and Microstructural Measurements

Since the A15 reaction front occurs radially from the filament centers, the deformation of the external filament leads to breakthrough of the DB bringing the A15 into contact with the Cu stabilizer at the shortest distance from the center. This typically occurs in only 2-3 filaments in a given cross-section(Fig 3), however, we also observed evidence of external A15 reaction from barrier breakdown outside the filaments with apparently intact DBs in the current plane of view (polished cross section), evidenced by Cu/Sn interdiffusion.

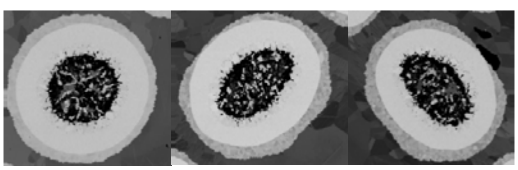


Fig. 3. Circular filament diameter is approximately 50um. 1) A typical inner ring filament has an aspect ratio close to unity, uniform barrier thickness, very isotropic. 2) A typical outer ring filament, aspect ratio up to 1.3, thinning of barrier on one side with thickening on the other. 3) An outer ring filament that has succumb to barrier breakthrough and subsequent Sn leak due to non uniform deformation

Filament aspect ratio increases almost linearly ring by ring from the center of the strand outwards (Fig. 1). Even with the small number of compromised DBs observed in these strands the RRR declines rapidly to less than 150. However, the amount of A15 produced shows that while enough Sn is leaking into the Cu to sufficiently degrade RRR, it does not have a measurable impact on A15 area. We measured 52-55% of the non-Cu package to be A15, with DB and core residues of about 25% and 22% respectively. However about a quarter of the A15 in the high J_c samples was A15 formed at the interface between the core and the DB that has grain size well over 1 μm. This large grain A15 fraction is rather independent of HT condition, representing 14%-16% of the total non-Cu after short reaction time when very little fine grain A15 has formed and J_c is low. In addition to having grain boundaries that have too great a separation for effective vortex pinning, we observe from longitudinal cross-sections that the large grain layer is discontinuous in the current carrying direction. Recent samples reacted at higher temperatures (650°C and 670°C) suggests that higher temperature HT further disconnects the large grain A15 annulus from the small grained region.

B. RRR

From our etching experiment, it is observed that most of the RRR degradation occurs in the regions of the outer two rings of filaments. It is expected for the worst quality Cu to be around the outer two rings where filament distortion is greatest and barrier breakthrough is most common. Inversely, the best Cu should be furthest from the Sn sources: namely the outermost Cu as well as Cu surrounding the core. Therefore, we expect to see a curve with some minimum RRR appearing near ring 6 or 7 and this is in fact the distribution we see in Fig. 4, an inset of the sample is included to guide the eye. While there remains a large volume of good stabilizing Cu in the core and the annulus outside the filament pack, it can become catastrophic to not have local interfilamentary Cu which can manage large current.

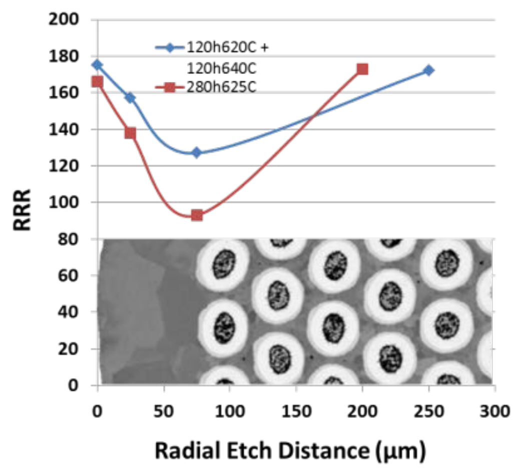


Fig. 4. By incrementally etching Cu away and subsequently measuring RRR, we can see a profile of where the worst Cu resides

IV. Conclusion

Studying the microstructure and resistance ratios of various heat treatments provides us with valuable information that can be used to help optimize PIT strands for high J_c while retaining enough DB to keep RRR >150. The amount of large grain A15 which forms is nearly independent of HT after only a short reaction time, limiting the amount of the filament that can be converted to the high-J_c fine grain A15. RRR measurements show that pushing to higher temperatures and time will form more small grain A15, but as the diffusion barrier begins to breach, the A15 comes at a high cost of conductor stability. A broader range of HT evaluations are underway with a view to finding the limits to reaction of this conductor which is very promising as a high J_c, fine filament conductor for high field magnet use.

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