

ADVANCES IN SUPERCONDUCTING STRANDS FOR ACCELERATOR MAGNET APPLICATION

Peter J. Lee and David C. Larbalestier, Applied Superconductivity Center, University of Wisconsin-Madison, Madison, WI 53706-1609, USA

Abstract

Considerable advances have recently been obtained in the critical current densities J_c of Nb₃Sn based superconductors - the prime candidates for the next generation of superconducting accelerator magnets. The non-Cu critical current densities now approach 3000 A/mm² at 12 T and 4.2 K in engineering quality strand. The design of new strands minimizes the amount of Cu in the package from which the Nb₃Sn is formed and increases the Sn level beyond that required to simply achieve A15 stoichiometry. The result is an A15 layer that is significantly more uniform than earlier generations of wire, both chemically and microstructurally, and wires that significantly surpasses previous Nb₃Sn strands in layer critical current density and in the specific grain boundary pinning force. Remarkably, these developments have been achieved in internal Sn based strands manufactured using both the modified jelly-roll technique with Nb-Ti alloy and the rod-in-tube approach with Nb-Ta alloy. The rod-in-tube approach is particularly exciting because it offers greater manufacturing flexibility. Advances have also been made in strand designs that offer the potential to reduce the large effective filament diameters, which are an issue with these new high- J_c strands. We review the latest developments in Nb₃Sn superconductors and compare their performance and potential with other round-wire high-field superconductors.

INTRODUCTION

The LHC marks the end of a series of colliders that have capitalized on increasing current densities available in Nb-Ti alloy based superconducting strand. Nb-Ti has proven to be a remarkably durable superconductor, dominating superconducting magnet design from the FNAL Tevatron (on-line in 1983) to the LHC (expected completion 2006). Multifilamentary superconducting strands based on Nb-Ti alloys are strong, ductile, and relatively inexpensive but are limited in operation to fields below ~11 T (2 K). In Figure 1 we compare the critical current density variation with applied magnetic field for superconductors of interest to accelerator magnet designers. With the exception of the Bi2223 tapes and the MgB₂-SiC [1] data, the critical current densities shown

are available in multifilamentary round-wire form suitable for magnet fabrication. Development of superconductors for accelerator magnets with fields greater than 11 T has focused on Nb₃Sn. Being brittle, the A15 structure of Nb₃Sn must be made from ductile components that can be drawn to wire, meaning that, unlike Nb-Ti, the materials package needed to make the superconductor contains more than just the A15 filament. Raising the J_c of Nb₃Sn is then both a question of maximizing the quantity of A15 within this package and of optimizing the A15 properties. The latest generation of Nb₃Sn strands can support non-stabilizer critical current densities in excess of 1000 A/mm² at fields up to 17 T at 4.2 K. Bi-2212, also a brittle material, can support ~1000 A/mm² out beyond 28 T. However, whereas Nb₃Sn conductors can be made with small Cu-stabilizer cross-sections, at present the engineering critical current density, J_e , of Bi-2212

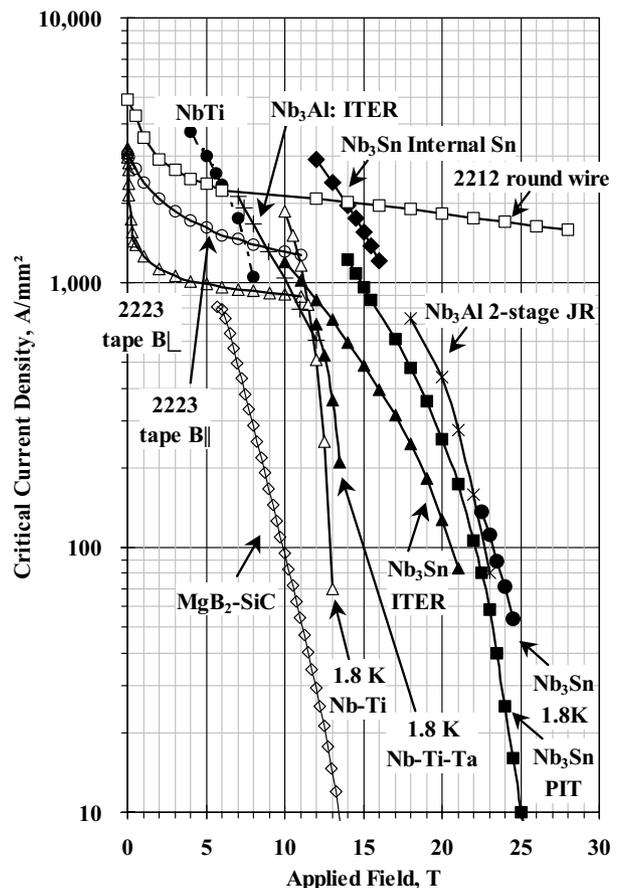


Figure 1: A comparison of critical current density with applied magnetic field for superconductors of actual or potential interest for accelerator magnets.

This work was supported in part by the U.S. Department of Energy under Grants DE-FG02-91ER40643 (High Energy Physics) and DE-FG02-86ER52131 (Office of Fusion Energy Sciences).

The authors may be contacted at the Applied Superconductivity Center at the University of Wisconsin-Madison, Madison WI 53706, USA (phone: 608-263-1760; fax: 608-263-1087; e-mail: peterlee@wisc.edu). D. C. Larbalestier is also with the Department of Materials Science and Engineering and Department of Physics.

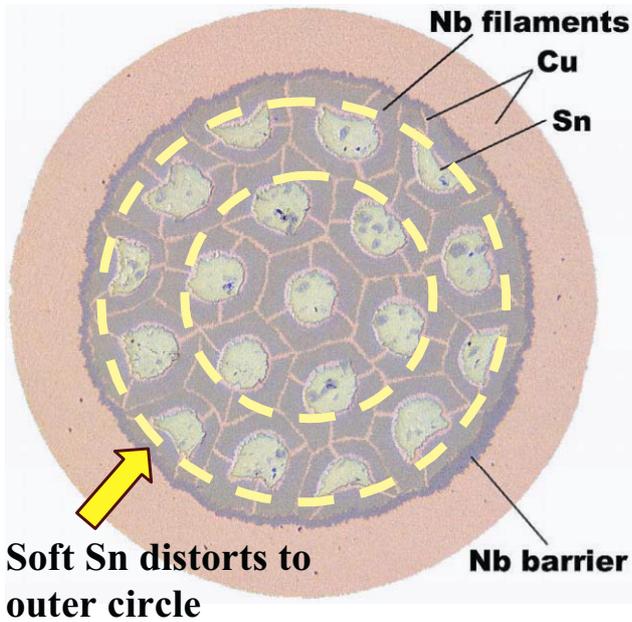


Figure 2: Cu-split sub-element high J_c strand produced by IGC-AS (now Outokumpu Advanced Superconductors).

conductors is still much reduced by the large Ag area in the strand. The latest generation of strand from OI-ST achieves J_c of 600 A/mm² at 12 T, 4.2 K with a 28 % Bi-2212 area [2]. Nb₃Al offers a high strength alternative but is much more expensive to manufacture than Nb₃Sn, which has not, so far, shown strength limitation in well designed high field accelerator magnets. MgB₂ has future long term potential for low cost and exhibits critical current densities that, although presently lagging behind Nb₃Sn, continue to show progress [e.g. 1]. A remarkable development in the past year has been the enhancement of the upper critical field by manipulation of the resistivity, with the upper critical field, H_{c2} , vs. temperature surface exceeding Nb₃Sn. [3]. In previous reviews we have examined the potential for High Temperature Superconductors [4] and MgB₂ [5] to high energy physics; in this paper we will focus on developments in Nb₃Sn.

PROGRESS IN Nb₃SN

Of the available superconductors, Nb₃Sn is the closest to targets set for the next generation of accelerator magnets [6]. The critical current density, J_c , (non-Cu, 12 T, 4.2 K) is very close to the 3000 A/mm² target, although the residual resistance ratio, RRR, is very low (2-13) in recent billets, due to diffusion barrier through-reaction in most cases. The effective filament size is still 2-3 times the 40 μm target, except for strands fabricated by the powder-in-tube (PIT) process. Piece length is reasonable good, considering the (250-1500 m) small billet sizes and the developmental nature of the strand for which two billets are rarely identical in design. New, more scaleable designs are showing promising results, for instance the Rod Restack Process at OI-ST that has achieved 2900 A/mm² (12 T, 4.2 K). There has been little

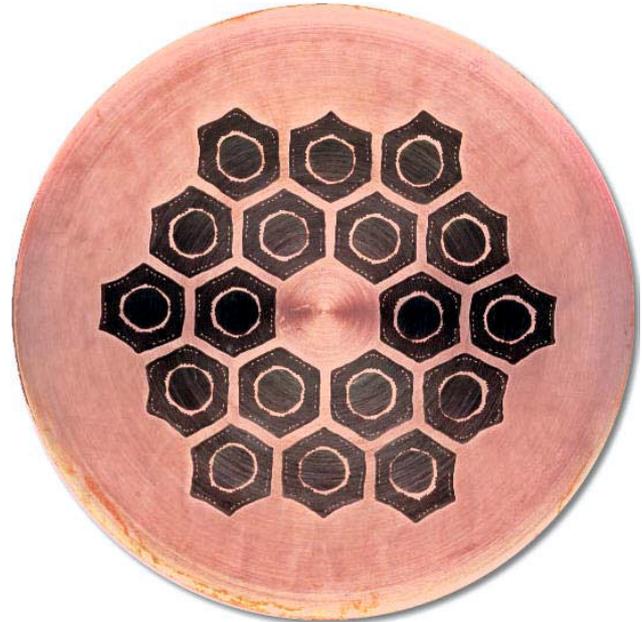


Figure 3: Hot Extruded Rod billet cross-section Image courtesy of Jeff Parrell, OI-ST.

progress in reducing heat treatment times for internal Sn to less than 150 hrs but PIT can be optimized at the desired 50 hr reaction. PIT wire costs are still limited by the small production scale and technical difficulties in scale-up.

Nb₃Sn has a big advantage over competing superconductors with respect to large-scale strand, cable and magnet production experience, such as record-setting

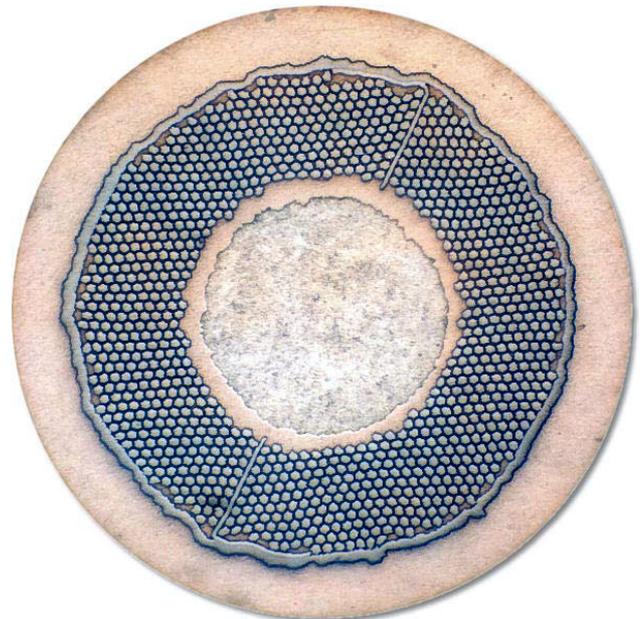


Figure 4: Single-stack, Internal Tin conductor "MEIT" manufactured by Supergenics LLC/Outokumpu Advanced Superconductors under a DOE-SBIR program. The single central Sn core reduces Sn distortion. It incorporates Nb-Ta fins to reduce the effective filament diameter. Image courtesy of Bruce Zeitlin of Supergenics, LLC.

dipole magnets at LBNL, the huge (150 ton, 13 T) ITER Central Solenoid Model Coil (ITER-CSMC)[7], It is also available from multiple vendors worldwide. Although the brittleness of Nb_3Sn dictates that it be used in the wind-and-react configuration for small-radius magnets, it can also be used in the react-and-wind approach for larger magnets [8].

Issues with Sn elements in Conductors

The use of elemental or weakly alloyed Sn has been crucial to the production of high critical current densities in internal Sn and even PIT strands. Higher Sn content pushes the Sn:Nb ratio in the A15, never always Nb_3Sn composition, closer to the stoichiometric ratio of highest H_{c2} and T_c [9]. Sn however is soft and not only becomes distorted itself during final wire drawing but also distorts the surrounding filament pack (see Figure 1), limiting the amount of post-billet-assembly processing that can be performed. Furthermore, in order to achieve useful piece length, the filament pack must be metallurgically bonded, which normally requires warm processing incompatible with the low melting point Sn (232 °C). One way around this problem is to assemble and warm-extrude the sub-element with Cu in the place of the Sn and then drill a hole in the Cu for the Sn after the other components are bonded. However, the smallest gun-drill diameter in a typical full-length 0.9 m billet is 4.7 mm. Taking account of the Sn distortion after assembly, this limits the number of sub-elements that can be stacked into a final billet without severe piece length problems.

One method to circumvent this problem being explored by OI-ST, uses salt in the place of the Sn, allowing hot extrusion of both the sub-element and final composite assemblies [10]. This process, termed HER for Hot Extruded Rod, is illustrated by the billet cross-section shown in Figure 3. After the first extrusion, the salt cores can be washed away and replaced with Sn at a late stage in the processing. With hot extrusion permitted for both sub-element and final composite assemblies, this process has excellent scalability.

The lack of Sn distortion in the center of the composite in Figure 2 suggests another alternative. The Mono Element Internal Tin conductor "MEIT" [11], uses a single central Sn core with a concentric extrusion-bonded

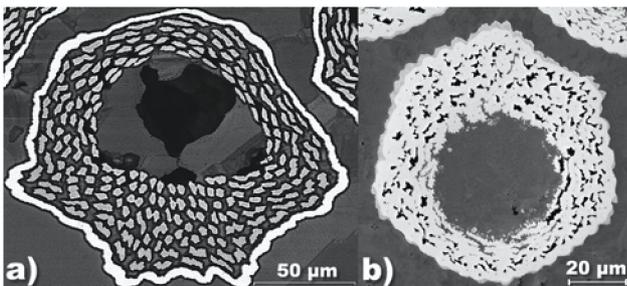


Figure 5: Comparison of Sub-element cross-sections in a) a high-Cu low hysteresis loss ITER-CSMC MJR strand fabricated by TWC, and b) a low-Cu, high J_c MJR strand fabricated by OI-ST.

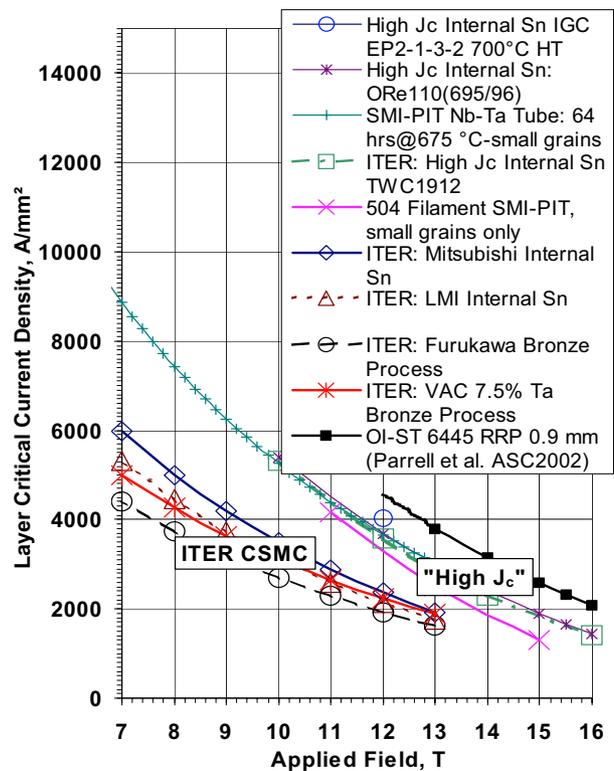


Figure 6: Comparison of the layer critical current densities for ITER-CSMC (low hysteresis loss) and recent "high J_c " Nb_3Sn composites. Note how different the intrinsic performance of the Nb_3Sn is.

Nb/Cu filament stack (gun-drilling of the solid Cu core is used to introduce the Sn after extrusion). Because there is now only one sub-element, the strand must be drawn to fine wire (< 0.2 mm diameter). Although such a fine wire may require a 2-level cable, the high symmetry means minimal Sn distortion (see Figure 4), high potential for good piece length, and low cost.

Issues with Sub-Element Size

To produce high overall J_c , the non-Cu sub-element must have a minimum of Cu filler and a high Sn concentration. This can enhance the A15 fraction to ~ 0.5 and dramatically enhances the layer critical current density. Figure 6 contrasts the high-Cu, low hysteresis loss ITER-CSMC strands with recent, low-Cu, "high- J_c " designs. Although some ITER-CSMC strands had high Sn:Nb ratios, the additional Cu required to isolate the individual filaments for low hysteresis loss (see Figure 5a) resulted in inferior Nb_3Sn -layer quality, even though the overall Nb_3Sn composition difference between the 2 designs was only ~ 1 at.% Sn [12]. Unfortunately the low-Cu requirement for highest J_c results in complete physical bonding of the individual Nb_3Sn filaments into a continuous ring of Nb_3Sn (see Figure 5b). Not only is the effective filament diameter, d_{eff} , increased by physical joining of the filaments but the core of the sub-element is

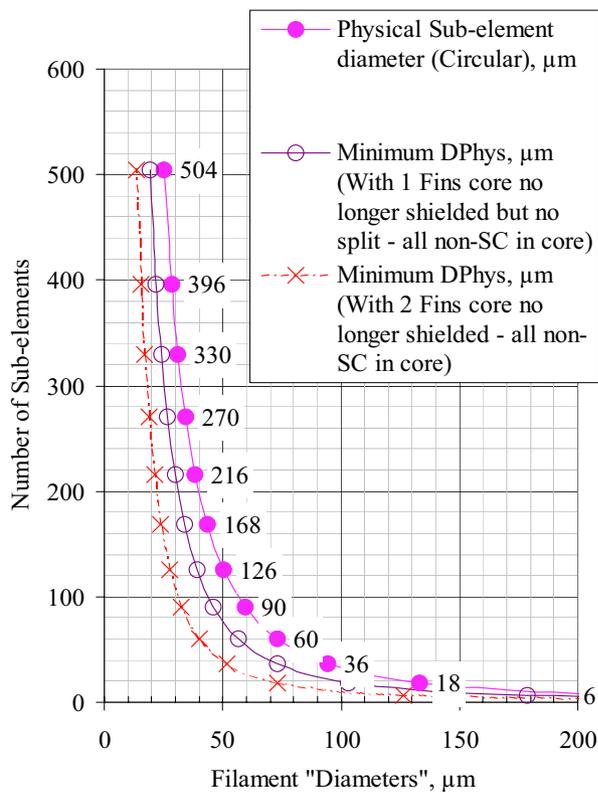


Figure 7: Calculated physical filament diameters based on simple circular geometries for a typical cable strand wire diameter (0.8 mm) and for a Cu:Non-Cu ratio of 1. The typical R&D high- J_c internal-Sn billets have 18-37 sub-elements in their restack, limiting them to sub-element diameters $>100 \mu\text{m}$. The Nb-Ta fins of Figure 4 can avoid shielding of the residual sub-element Sn-cores (open circles) by dividing the sub-element into two components, which do not shield their core (X). This plot was suggested by one made by R. Scanlan [13].

also shielded, making the entire sub-element cross-section behave as one and proportionately increasing the loss.

This small size of R&D billets and the problem of Sn distortion limits the restack numbers of sub-elements so far to 18-37. By using simple scaling arguments, we can see that this limits the sub-element diameters to $>100 \mu\text{m}$ (Figure 7), well above that desired for low hysteresis loss for both HEP and fusion magnet use.

Supergenics LLC has developed a sub-element splitting technology using Nb-Ta dividers (patent pending) that should “un-shield” the sub-element core by subdividing the sub-element filament pack. In Figure 7 we model the effect of “un-shielding” the core with one Nb-Ta divider, as well as the added benefit of un-shielding the core and splitting the A15 volume with two dividers. Figure 7 shows that there must be at least 60 sub-elements to approach the desired $40 \mu\text{m}$ effective filament size target. Although 504-filament PIT conductors which exceed the $40 \mu\text{m}$ target have been made, this goal seems much more feasible for internal Sn than experience with internal Sn is much more widespread than with PIT.

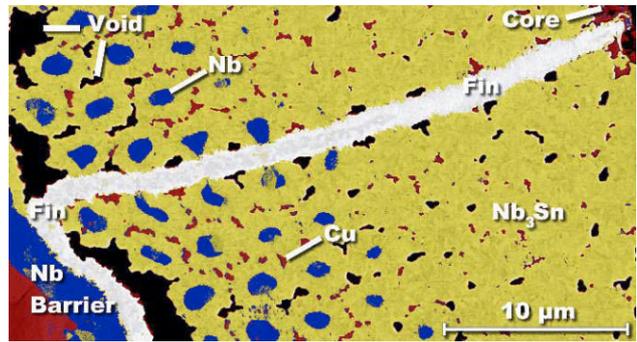


Figure 8: A false color atomic number sensitive electron backscatter image of a sub-element cross-section near a Nb-Ta fin after reaction. Composite fabricated by Outokumpu Advanced Superconductors/Supergenics and heat treated by E. Barzi at FNAL.

The first attempts by Supergenics LLC/Outokumpu Advanced Superconductors under a DOE-SBIR program at using such Nb-Ta dividers has been successful in producing composites of 18 and 36 sub-elements, although significant reaction of the Nb-Ta alloy fin with Sn has reduced its effectiveness. In Figure 8, we show a false-color, atomic-number-sensitive, electron-backscatter image near a Nb-Ta fin after reaction. The inside filaments (right, yellow) are evidently fully reacted while the outer filaments (left) furthest away from the original Sn core have unreacted Nb cores (blue). Near the original Sn core, the fin is full reacted by Sn, apparently robbing the outer filaments of needed Sn. The result is a significantly reduced J_c of the composite. The “fin”, however, has been successful in stopping the reaction with the outside Nb diffusion barrier (blue, left), so that a continuous shielding external Nb₃Sn layer is not produced.

Closer examination of the Nb-Ta reaction region in Figure 9 reveals a complex reaction producing changes in composition and thickness of the grain boundaries in the

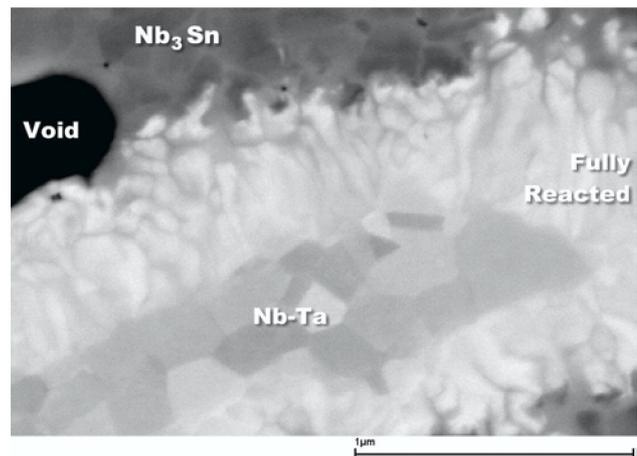


Figure 9: Atomic-number sensitive, electron-backscatter image of the Nb-Ta divider in Figure 8 in the fully reacted region. The grain boundaries in the reacted divider and the adjacent Nb₃Sn are clearly of variable composition.

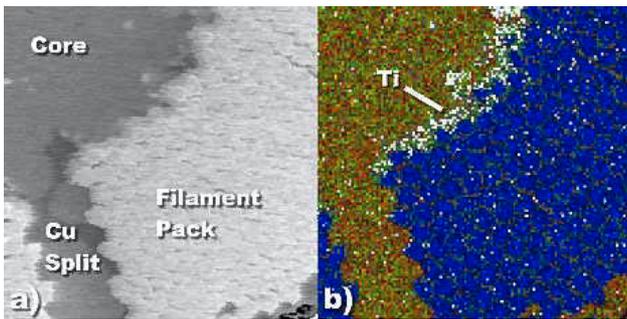


Figure 10: Detail of Core/Filament Pack region of OAS 0.4 mm dia. monocore strand after 150h/530°C heat treatment in the investigation of Suenaga [15] and Uhlrich [14]. In a) an atomic-number sensitive electron backscatter image and b) a spectral image (energy dispersive x-ray) show that Ti (white) segregates to the inner side of the Nb (blue) filament pack (Cu in red, Sn in green).

Ta(Nb)₃Sn and the Nb₃Sn. This suggests that an alternative barrier, perhaps Ta, might be more effective in implementing this promising concept for reduced d_{eff} .

Issues with Ti

Alloying the Nb with Ti (or Ta) is required to raise H_{c2} and maximize high field J_c but increases the cost of the Nb alloy and can reduce piece length. Thus incorporating Ti into the Sn core has been investigated too.. With ever decreasing Cu content in the filament pack, however, this route has become less effective because Ti reduces the mobility of Sn through the filament pack [15]. Energy dispersive x-ray analysis has indicated that the Ti forms a Nb-Sn-Ti ternary phase at the core/filament-pack interface. The segregation of the Ti can be clearly seen in the EDS spectral image in Figure 10b.

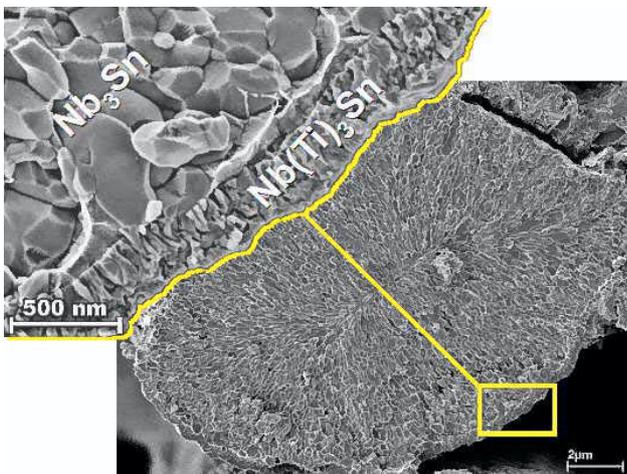


Figure 11: High resolution field emission scanning electron microscope image of an outer filament in a 1st generation OI-ST HER strand where the Ti has only partially penetrated the Nb filament. Fracture of the filaments reveals two sizes of A15 grains, The Nb(Ti)₃Sn grains are much smaller than the pure Nb₃Sn grains.

An extreme case of non-uniform Ti distribution can be seen in Figure 11, where the Ti only partially penetrates the Nb filament – leading in this case to 2 distinct A15 layers. New methods of Ti alloying are now being investigated under the DOE-SBIR program.

ACCELERATOR CONDUCTOR ISSUES

Remarkable progress has been made towards a new generation of conductors that meet the goals of the high energy physics community. As the preceding sections indicate, however, there is still plenty to do. Among the key issues that still need to be resolved are:

- 1 How low can the effective filament size for “High J_c ” Nb₃Sn strand be reduced?
- 2 Can the cost of PIT strand be reduced so that it competes with internal Sn?
- 3 Can the expected cost reduction in internal Sn conductors be achieved without sacrificing properties?
- 4 How close are we to the intrinsic performance limits for Nb₃Sn strand J_c ?

REFERENCES

- [1] S.X. Dou, et al., App. Phys. Lett. 81(18), pp. 3419-3421 (2002).
- [2] Private communication from S. Hong and K. Marken.
- [3] A. Gurevich, et al. “Anomalous enhancement of the upper critical field in the two-gap superconductor MgB₂ by selective tuning of impurity scattering” (submitted to Nature December 2002).
- [4] D. C. Larbalestier and P. J. Lee, IEEE 1999 Particle Accelerator Conference, vol. 1, pp. 177-181, 1999
- [5] L.D. Cooley, C.B. Eom, E.E. Hellstrom, and D.C. Larbalestier, 2001 Particle Accelerator Conference, Vol.1, pp: 203 –207, 2001.
- [6] R. M. Scanlan, IEEE Transactions on Applied Superconductivity, 11(1), pp. 2150 –2155 (2001).
- [7] N. Martovetsky et al., IEEE Trans. Applied Superconductivity, 12(1), pp. 600-5 (2002).
- [8] R. McClusky, K.E. Robins and W.B. Sampson, IEEE trans. on Magnetics, 27, pp. 1991-1995 (1991).
- [9] H. Devantay, et al., *J. Mat. Sci.*, 16, 2145 (1981).
- [10] US Patent Number 5534219
- [11] B. A. Zeitlin, et al. Adv. in Cryo. Eng., 48, pp. 978-985, 2002.
- [12] P. J. Lee et al., paper 2MK05 presented at ASC 2002, to be published in IEEE Trans. Applied Superconductivity.
- [13] R. Scanlan, “ J_c and $D_{effective}$: Where are we now, and what do we need to do?” presentation at the Low Temperature Superconductor Workshop, Napa, 2002.
- [14] J. Uhlrich, University of Wisconsin REU project in collaboration with M Suenaga, LBNL, 2002.
- [15] M. Suenaga, “Sn Diffusion Effects in High J_c Internal Tin Processed Wires,” presentation at the Low Temperature Superconductor Workshop, Napa, 2002.