

# NANO-ENGINEERING Nb<sub>3</sub>Sn THIN FILMS TO IMPROVE WIRE PERFORMANCE AND REDUCE COST

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## Abstract

State of the art Nb<sub>3</sub>Sn wires have plateaued in the performance of critical current density  $J_c$ . Chemical and geometric optimization of the wire layout has produced Nb<sub>3</sub>Sn wires with a maximum  $J_c(4.2\text{ K}, 16\text{ T}) \sim 1300\text{ A/mm}^2$ . A future high energy hadron collider that is being considered to follow the LHC would need larger  $J_c$  and be cost effective. The approach to improving the performance of Nb<sub>3</sub>Sn conductor would be to introduce enhanced flux pinning mechanisms with nano-engineering techniques.

## INTRODUCTION

Future high-energy proton colliders with center-of-mass energies up to 100 TeV are being discussed as a future step beyond the LHC. Such a machine would use magnets with higher fields than today's colliders to keep the machine circumference reasonable. Dipole magnets for this collider will require fields in the range from 16 to 20 T. To achieve this goal not only will the critical current of the superconductor need to be improved, the superconductor will need to be cost-effective. Although high-temperature superconductors (HTS) are being considered as possible conductor choices for magnets in this field range, these conductors have some unfavourable properties. YBCO conductor is only available in tape format and the  $J_c$  is sensitive to its orientation to the magnetic field.  $J_c$  is largest when the field is in the plane of the tape (the crystal ab-plane), however a dipole will have a field component perpendicular to the conductor plane in part of the magnet. Furthermore, because of the tape format, YBCO conductor will have large persistent magnetization currents that may affect unwanted harmonics which may be difficult to correct as the magnet is powered from injection to the operating field. Bi2212 requires Ag and/or Ag-Mg alloy to stabilize the conductor. As a large fraction of the Bi2212 wire is silver it is unlikely that the conductor cost will be practical to construct the magnets for the entire collider ring. This paper will concentrate on improving the performance of Nb<sub>3</sub>Sn wire.

## Nb<sub>3</sub>Sn PROPERTIES

Nb<sub>3</sub>Sn is a type II superconductor with a critical temperature  $T_{c0} \approx 18\text{ K}$  and an upper critical field  $H_{c20} \approx 30\text{ T}$ . This can be compared to NbTi conductor, which is the preferred superconductor for magnets with fields less than 9 T, with  $T_{c0} = 9.8\text{ K}$  and  $H_{c20} = 14.5\text{ T}$ . In the range from 10 to 16 T the engineering current density,  $J_E$ , is competitive with HTS conductors, however the  $J_E$  falls off fast

with increasing magnetic field. There are challenges with using Nb<sub>3</sub>Sn. As the material is brittle the critical current can be degraded by strain which complicates coil winding and limits its use at high field by Lorentz forces. This leads for magnets to the wind-and-react technique, where the coil is wound before heat treating the Nb<sub>3</sub>Sn Rutherford-type cable in inert atmosphere. The cable is made of dozens of wires. The wire itself is formed from billets that are extruded and drawn to the final strand size where the cross-section area of the wire is reduced by a factor of  $10^5$ .

## PINNING CENTERS

Magnetic fields below the critical magnetic field  $H_{c2}$  can enter into a type II superconductor as discrete quantized flux units (fluxons). For a superconductor without defects the fluxons which repel each other would be distributed uniformly. In the presence of an external magnetic field, Lorentz forces would cause the fluxons to move which would locally generate heat causing the superconductor to go normal. Defects in the superconductor crystal lattice can inhibit fluxon motion. These defects can be caused by lattice vacancies, grain boundaries, impurities, etc. The critical current density of a superconductor is determined by the condition at which the force that the pin exerts (pinning force) is equal to the Lorentz force on the fluxon. Increasing the density and strength of the pinning centers should improve  $J_c$ .

The orientation of the fluxon vortex with respect to the grain boundary geometry affects the force and consequently  $J_c$ . The transverse pinning case [1, 2] occurs when the fluxon is aligned with the defect as in the boundary region between grains. In this case the pinning force is approximated by the pinning energy divided by the vortex spacing. In the longitudinal pinning case the intergranular vortex slides along the grain boundary and is pinned by the inhomogeneities. The longitudinal pinning force is approximated by the gradient of the Josephson coupling energy of the vortex [1, 2].

The flux pinning centers in un-doped Nb<sub>3</sub>Sn are largely at the grain boundaries. The pinning force is inversely proportional to the grain size for sizes greater than 100 nm. Fig. 1 shows this relationship for Nb<sub>3</sub>Sn. The grain size is largely dependent on the reaction heat treatment temperature to form Nb<sub>3</sub>Sn [3] and increases rapidly with that temperature. To obtain small Nb<sub>3</sub>Sn grain size it is desirable that the heat treatment be at temperatures as low as possible, however at temperatures below 600° C the heat treatment does not produce Nb<sub>3</sub>Sn with the desired purity. At 615° C (the lowest temperature that is

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typically used) the grain size is 100 to 110 nm. Fig. 2 shows the dependence of grain size as a function of the heat treatment temperature. The most effective pinning occurs when the size of the defect is comparable to the superconductivity coherence length and the fluxon spacing matches the defect spacing (eg. grain size). The coherence length of Nb<sub>3</sub>Sn in a 12 T field at 4.2 K is ~5 nm. The grain size even at the lowest reaction temperature is much larger than the optimum.

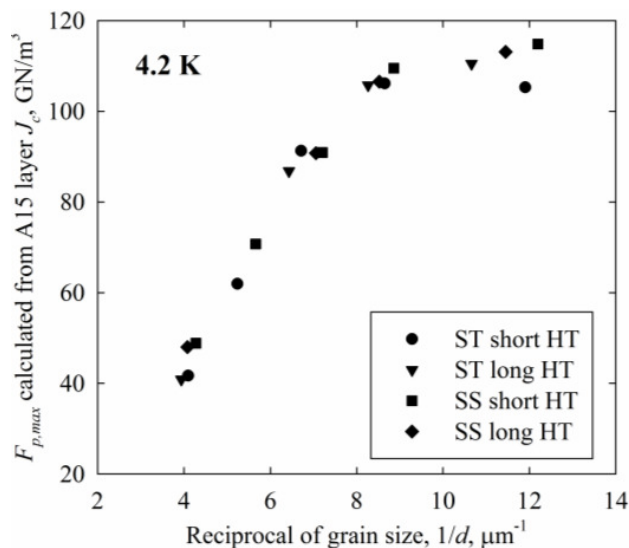


Figure 1: Pinning force as a function of the reciprocal grain size. From Ref [3].

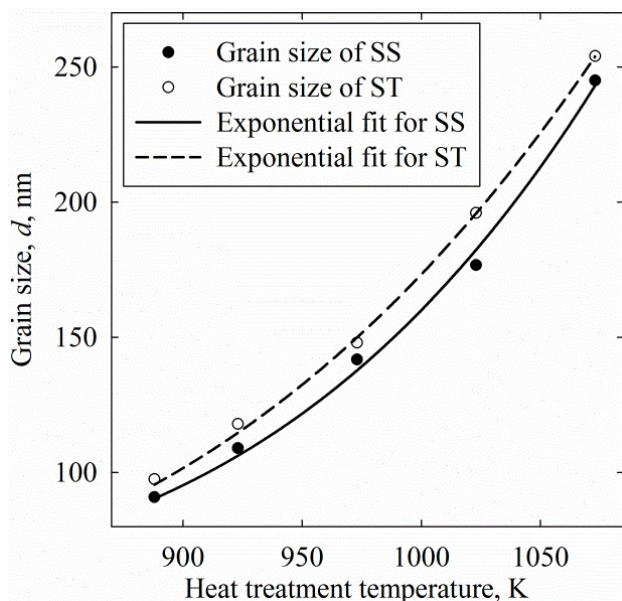


Figure 2: Dependence of the Nb<sub>3</sub>Sn grain size on the heat treatment temperature. From Ref [3].

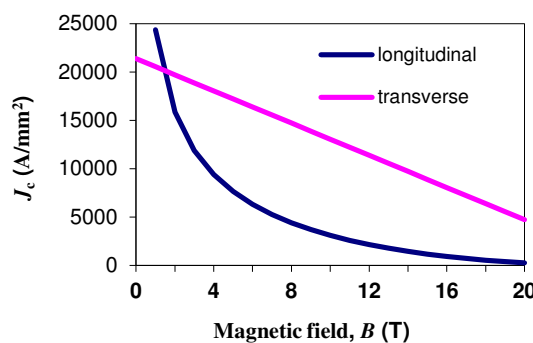


Figure 3: Critical current density versus magnetic field for transverse and longitudinal pinning. This comparison is made for specific parameters indicated in Ref [1].

An approach that appears effective in reducing grain size is the internal oxidation technique [4]. It was shown using this technique that a Nb<sub>3</sub>Sn wire could achieve a layer  $J_c = 9600 \text{ A/mm}^2$  at 4.2 K and in 12 T field. This technique used a Nb-Zr alloy surrounding a Sn core with a SnO<sub>2</sub> powder layer between the alloy shell and the Sn core. During the heat treatment reaction where the Nb<sub>3</sub>Sn is formed, ZrO<sub>2</sub> is also formed and precipitates out with a relatively uniform distribution. The ZrO<sub>2</sub> particles effectively limit the average Nb<sub>3</sub>Sn grain size to 36 nm, which is a third of the grain size of the non-doped strands. Internal oxidation had the effect of nearly doubling the layer critical current of the Nb<sub>3</sub>Sn over the  $J_c$  of the commercially available Nb<sub>3</sub>Sn.

We are proposing a complementary approach to that of Xu et al. [4] with the goal of enhancing the transverse pinning component. It is expected that both intragranular and intergranular vortices exist in Nb<sub>3</sub>Sn so both transverse and longitudinal pinning should be present. The equiaxed grain structure of Nb<sub>3</sub>Sn favors longitudinal pinning for most of the field domain. Fig. 3 shows  $J_c$  calculated as a function of external field for both transverse and longitudinal pinning orientations for a set of parameters (given in ref. [1]) typical for Nb<sub>3</sub>Sn. As the  $J_c$  for longitudinal pinning varies as  $\sim 1/B^{1/2}$ , the transverse pinning should be more important at higher fields.

Nb-Ti conductor was optimized using a network of thin  $\alpha$ -Ti ribbons as a coherent defect structure for pinning. Such an approach couples the pinning structure to the Nb-Ti lattice. This coupling can result in very large  $J_c$ 's as the localized closed current grain loops are more coherent. This approach may be applicable for Nb<sub>3</sub>Sn using a dense distribution of appropriate strips or ribbons in the pre-reacted Nb<sub>3</sub>Sn wires.

## PROPOSED STUDY

We are proposing to study how to improve  $J_c$  of Nb<sub>3</sub>Sn multifilamentary superconducting wires by using flux pinning techniques. In a manner similar to the introduction of  $\alpha$ -Ti ribbons into Nb-Ti superconductor we would introduce a high density distribution of strips or ribbons into the pre-reacted Nb<sub>3</sub>Sn wire. Potential material candidates for the strips are tantalum, molybdenum and va-

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nadium. These material candidates are sufficiently ductile for wire drawing and they do not interact with the other materials during the heat treatment (Tantalum is likely to be the best choice.). To obtain the ribboning structure for the transverse component of pinning, lithographic methods will be first attempted. 3D architectures of Nb<sub>3</sub>Sn are created by using optical and electron beam lithography. Resist patterns will be exposed on the substrate using optical and electron beam lithography. After that, multi-layered nanowires will be grown using electrodeposition. The electron beam will be used for gratings whose widths are smaller than 0.5 μm.

We would plan to produce thin film samples for the study. The Nb<sub>3</sub>Sn samples would be produced using a novel electro-chemical technique [5] which was subsequently improved. The procedure uses a niobium substrate upon which layers of copper, tin and copper, respectively are deposited. The first copper layer is necessary to provide to provide copper to the Nb-Sn system which reduces the temperature needed for the formulation of the superconducting A15 crystal structure of Nb<sub>3</sub>Sn. The second copper layer is applied as a barrier layer against the tin and bronze liquid phases during the heat treatment. The electrodeposition process is carried out at near room temperature and atmospheric pressure. The electrodeposition procedure is described in detail in Ref. [6]. The Nb<sub>3</sub>Sn layer is formed through solid diffusion at a temperature around 650° C in the presence of copper. In the ternary Nb-Cu-Sn system only Nb<sub>3</sub>Sn is formed [7]. Without the copper, NbSn<sub>2</sub> and Nb<sub>6</sub>Sn<sub>5</sub> would not be converted to Nb<sub>3</sub>Sn at temperatures less than 900° C. The thin film approach provides a quick turnaround of samples allowing a cost effective way to optimize the flux pinning properties and to test against theoretical models.

To study the characteristics of the samples we will employ GDOES spectroscopy to determine the layer composition. Fig. 4b shows the composition profile of the Nb/Cu/Sn/Cu sample studied in Ref. [6]. X ray diffraction (XRD) shown in Fig. 4a is used to reveal the cubic crystalline structure of the Nb<sub>3</sub>Sn phase that is expected for an A15 compound. XRD is also sensitive to the presence of NbSn<sub>2</sub> contamination. SEM can be used to produce cross section images of the plated layers with micron accuracy of the layer thicknesses. Fig. 5 shows SEM images of two samples from Ref. [6]. The images show Nb<sub>3</sub>Sn layers with thicknesses of 8.0±0.5 μm (left) and 5.7 μm (right). Outside the Nb<sub>3</sub>Sn layer is a bronze layer formed from the excess tin alloyed with the copper protective layer. The measurement of the Nb<sub>3</sub>Sn layer thickness and the grain size are important data for the sample characterization.

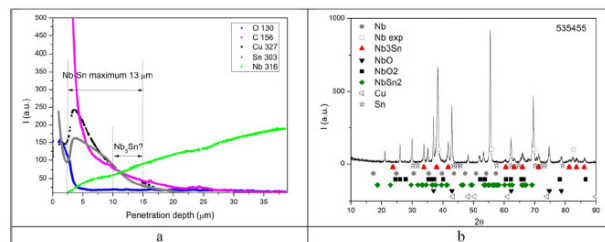


Figure 4: (a) XRD pattern and (b) GDOES analysis of Nb/Sn/Cu after heat treatment [6].

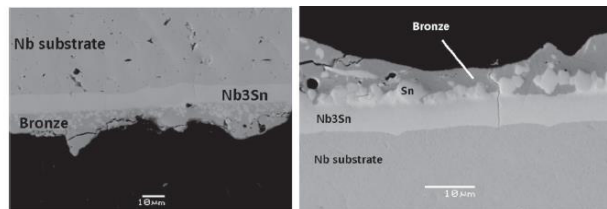


Figure 5: SEM cross-section of two samples analysed in [6].

## CONCLUSIONS

We have presented a model? to improve the critical current density of Nb<sub>3</sub>Sn using flux pinning techniques. Introducing a high-density distribution of strips or ribbons into pre-reacted Nb<sub>3</sub>Sn in a manner similar to the α-Ti ribbons used with Nb-Ti superconductors could improve the J<sub>c</sub>.

Films would be used as a faster turn-around method for research versus using actual billets. If a successful mechanism is identified, the goal will be to transfer the successful mechanism to conventional billet manufacturing techniques to produce Nb<sub>3</sub>Sn wires with larger J<sub>c</sub> for magnet applications.

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