

STOICHIOMETRIC Nb₃Sn IN FIRST SAMPLES COATED AT CORNELL*

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Abstract

A cavity coated with the superconductor Nb₃Sn theoretically will be able to reach more than twice the maximum accelerating field of Nb in a cavity under the same operating conditions and will have a much lower BCS surface resistance at a given temperature. The SRF group at Cornell has recently developed facilities to fabricate Nb₃Sn on Nb. The first samples have been coated, and several tests have been performed to characterize them. Results presented include SEM images of the surface, anodization tests, a critical temperature measurement, a test for RRR degradation, and stoichiometry measurements using EDS and XPS.

INTRODUCTION

After years of development, niobium SRF cavities are now being fabricated that are limited in gradient by the RF critical field, not multipacting, thermal instability, field emission, or high field Q slope. To reach even higher gradients, new materials must be used that will allow the RF surface to tolerate higher magnetic without quench. Theory [1] predicts that coating a cavity with Nb₃Sn will allow it to reach twice the accelerating gradient of a niobium cavity. Nb₃Sn also benefits from a critical temperature that is nearly twice as high as that of niobium, which allows cavities coated with Nb₃Sn to operate with quality factors far exceeding those of Nb cavities at the same temperature.

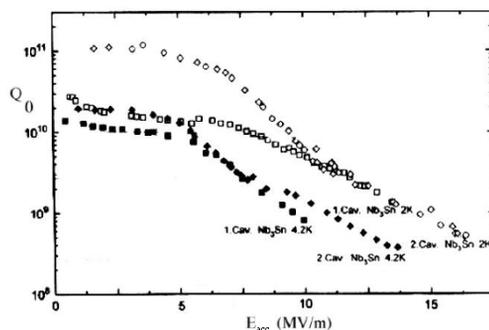


Figure 1: The best results obtained at University of Wuppertal for 1.5 GHz cavities vapor-diffusion coated with Nb₃Sn. From [4].

The great potential of Nb₃Sn-coated cavities has already been shown in the results obtained by researchers at University of Wuppertal in the 1980s-90s [2] [3]. They produced cavities with quality factors of 10¹¹ at 2 K and ac-

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celerating gradients competitive with the gradients of niobium cavities produced at the time. Some vertical test results are shown in Figure 1. Guided by their work, facilities at Cornell have been developed to fabricate Nb₃Sn films on niobium. This paper discusses those facilities and the first results obtained.

COATING PROCESS

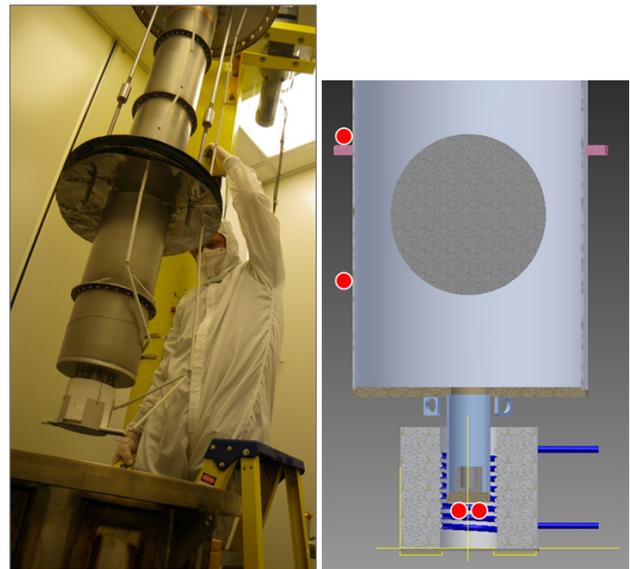


Figure 2: The Nb₃Sn coating chamber being inserted into the UHV furnace (left) and a cross section (right) showing the tin crucible surrounded by the heater and the substrate suspended above. The red circles represent thermocouples for temperature monitoring.

The vapor diffusion coating process was based on [2]. A UHV furnace containing a niobium substrate (e.g. a cavity), a tungsten crucible containing 99.999% Sn pellets, and a tungsten crucible containing a small amount of SnCl₂ is evacuated and degassed at 200°C. The coating chamber, shown in Figure 2, is self-contained to avoid contaminating the furnace with tin. The valves to the pumps are then closed and the temperature is raised to 500°C, at which point the vapor pressure of SnCl₂ is high enough for it to create tin nucleation sites on the Nb surface. After 5 hours, the furnace temperature is raised to 1100°C, and a heater raises the temperature of the Sn slightly higher, to 1200°C. The elevated temperature from the heater increases the vapor pressure of tin in the coating chamber. The furnace temperature and the tin temperature control respectively the rate of interdiffusion of niobium and tin in the sample and

the rate at which tin arrives at the sample surface, and they can be varied to affect the composition of the alloy produced. After 3 hours the heater is turned off, and the remaining tin vapor in the chamber is given 30 minutes to reach a surface and diffuse in, so as to avoid unreacted tin on the RF surface. Finally, the furnace is turned off and backfilled with nitrogen gas when it is cool.

The Cornell procedure will include preparation techniques that have been developed in the last two decades. Already, Nb substrates—currently only flat samples have been coated, not full cavities—are HPR'd prior to coating. Soon, Nb substrates will be EP'd to give a smooth RF surface prior to coating, and also baked at 120°C.

NON-RF ANALYSIS OF Nb₃Sn COATINGS

Several different techniques have been employed to study the composition of the first coated samples produced. Only RF tests will determine if the coating process is satisfactory; these tests are intended just to confirm that Nb₃Sn is indeed produced using the apparatus. In the future, these tests will hopefully also serve to correlate RF performance to surface qualities.

One of the samples was anodized to test the surface composition. In this procedure, the sample is placed in 10% NH₄OH and a 75 V potential is applied between it and an anode. The thickness of the oxide that grows gives it a color that is characteristic of the material on the surface. Any unreacted tin will turn yellow, niobium will turn blue, and Nb₃Sn will turn pink-purple [5]. The color of the sample, shown in Figure 3, indicates that the material on the surface is Nb₃Sn.

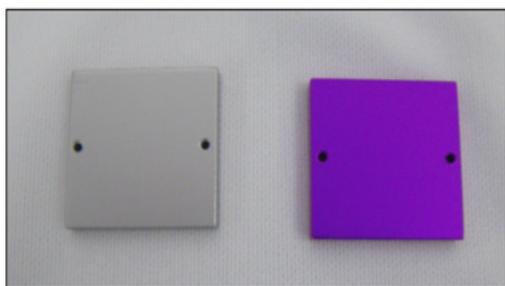


Figure 3: Nb₃Sn-coated sample (left) and an identical sample after anodization in NH₄OH at 75 V (right). The pink-purple color indicates Nb₃Sn phase on surface.

Images obtained using a LEO 1550 FESEM of the first samples coated are shown in Figure 4. Micron-sized grain growth was observed similar to that in images published by the group at Wuppertal.

To quantify the composition of the Nb₃Sn layer, an energy dispersive x-ray (EDX) analyzer connected to the FESEM was used. Firing 10 keV electrons at the sample and measuring the spectrum of x-rays emitted (shown in Figure 5), it probes the relative abundance of Nb and Sn up to approximately 0.5 microns into the surface [6]. An abun-

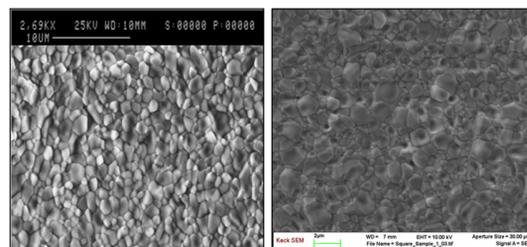


Figure 4: SEM images of Nb₃Sn produced at Wuppertal in 1996 (left) and at Cornell in 2011 (right).

dance of 24.2 ± 0.5 atomic percent tin (atm%Sn) was measured (uncertainty from [7]).

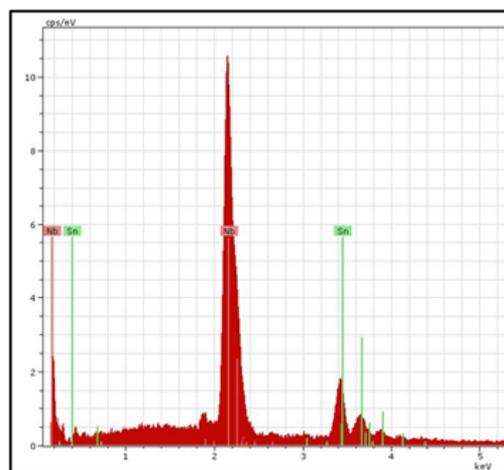


Figure 5: EDX spectrum of Nb₃Sn sample indicates composition ~ 24.2 atm%Sn.

Nb₃Sn is the name given to the A15 phase of the Nb-Sn system, which has a composition ranging from approximately 18 to 25 atm%Sn. A. Godeke has reviewed the strong variation in the properties of the alloy with composition [9]. For example, the variation of T_c and H_{c2} with composition is shown in Figure 6. The data indicate that higher Sn content will yield a higher T_c , which is desirable to reduce the BCS resistance. Godeke notes however that above 24.5 atm%Sn the alloy will undergo a spontaneous lattice distortion at low temperatures which can lead to strain and, Godeke suggests, reduced RF performance [10]. Godeke recommends alloying close to 24.5 atm%Sn but below it, which would mean that the coating produced by the Cornell furnace is right in the desired composition range. Unfortunately, Godeke points out, there is no information available on the variation of H_c or H_{c1} with composition.

X-ray photoelectron spectroscopy has a much shallower scan depth than EDX, on the order of 10 nm [8]. By alternatively performing an XPS scan and sputtering the sample with Ar⁺ ions, the variation in composition with depth could be studied. The results in Figure 7 show that the Nb₃Sn layer has approximately uniform composition up to

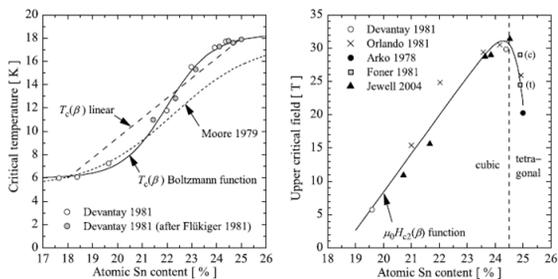


Figure 6: Literature data for T_c versus composition (left) and H_{c2} versus composition (right). From [10].

a depth of $\sim 1.5 \mu\text{m}$. The first surface scan is not shown as it showed foreign elements, likely from handling outside the clean room. The composition is normalized to the scan after the first etch, as differential sputtering rates give the absolute composition very large uncertainty.

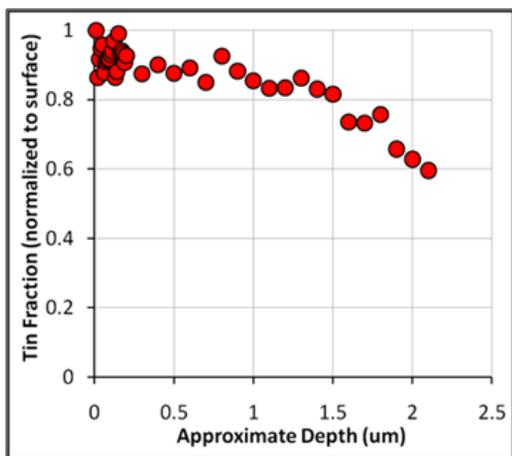


Figure 7: Alternately sputtering sample and performing XPS scans shows composition versus depth.

To better characterize the Nb_3Sn layer, its superconducting properties were measured. The critical temperature of superconductors is often measured by induction [5], using coupled coils on either side of the sample to determine the temperature at which the sample expels magnetic flux due to the Meissner effect. An apparatus is currently being manufactured at Cornell to perform this measurement. Fortunately, while it is being fabricated, an apparatus was available to perform a simple 4-wire measurement of the transition. It involves passing a low-frequency AC current through the sample and measuring the induced voltage with a lock-in amplifier while varying the temperature of the sample. The 4-wire measurement has the disadvantage of generally measuring only the highest T_c path between the leads, but it is a good early characterization to make. The measured transition is shown in Figure 8. A large discontinuity in resistance indicating a superconducting transition occurs at $18.1 \pm 0.2 \text{ K}$. This is very close to the highest recorded value for Nb_3Sn , 18.3 K [9]. No discontinuity is observed at any other temperature, indicating that a com-

plete path is available between the leads with this critical temperature. The non-zero resistance below the transition at 18.1 K may result from not having the correct reference phase on the lock-in amplifier to remove any reactive components of the impedance.

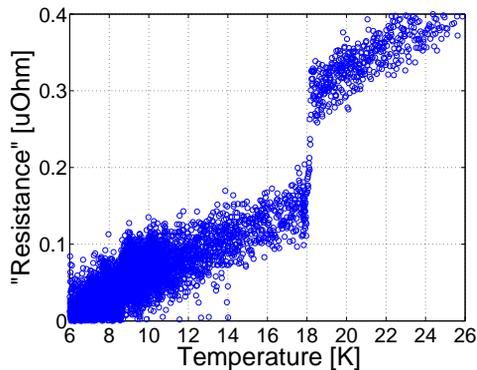


Figure 8: 4-wire measurement of the Nb_3Sn sample indicates a critical temperature of $\sim 18.1 \text{ K}$

In the same 4-wire measurement, an estimate was made of the RRR degradation in the Nb substrate resulting from the coating process. During the coating process, the valves to the pumps are closed when the furnace temperature is above 200°C , potentially allowing residual gases to reduce the RRR of the substrate, which was initially 280. To check for this, the resistance of the sample was monitored from the transition up to room temperature, as plotted in Figure 9. RRR is defined as the ratio of the 300 K resistance to the normal 4.2 K resistance. The normal resistance at 4.2 K could not be measured, but a lower bound for the RRR of 210 could be obtained using the resistance just above the transition. This shows that the RRR degradation during coating is minimal.

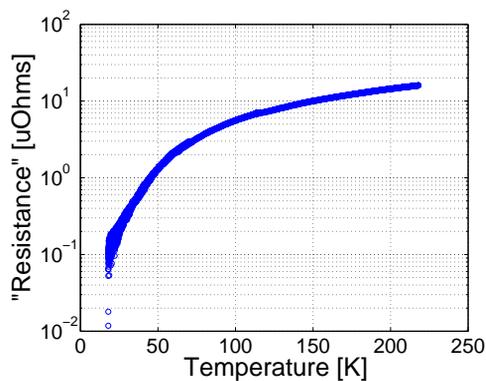


Figure 9: Resistance of sample versus temperature shows that minimal RRR degradation occurred during coating process.

RF TESTING OF Nb₃Sn COATINGS

A Nb bottom plate for the Cornell TE pillbox cavity was coated with Nb₃Sn so that RF tests could be performed. Images before and after coating are shown in Figure 10. It was also made from RRR 280 Nb and it received a 130 μm BCP and HPR before coating. A 1-day degas was performed before high temperature treatment to ensure minimal RRR degradation. A small witness sample was coated with the bottom plate, which showed a composition of 23.6 ± 0.5 atm%Sn under EDX analysis (though its tin content is likely somewhat smaller than that of the bottom plate, which was closer to the tin source), and turned pink-purple when anodized. These tests demonstrate the reliability of the coating process to produce Nb₃Sn of the desired composition.

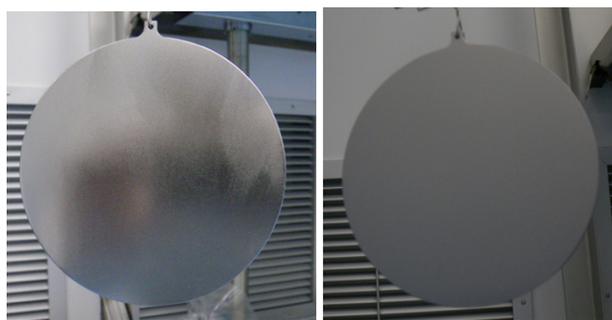


Figure 10: Pictures of pillbox cavity bottom plate before (left) and after (right) Nb₃Sn coating.

RF testing in the pillbox cavity, models of which can be seen in Figure 11, will begin as soon as commissioning of the cavity is complete. A small T-map has also been developed for precise measurement of the small surface resistances expected from Nb₃Sn. See [11] for details.

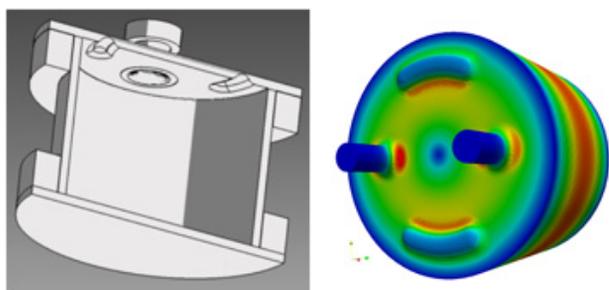


Figure 11: CAD (left) and CST (right) models of the Cornell TE Pillbox Cavity.

CONCLUSIONS

Very promising results have been obtained on the first samples produced by the Nb₃Sn program at Cornell. T_c measurements, EDX, and anodization all suggest that the desired composition has been achieved. The next step will be RF testing in a TE pillbox cavity. Fabrication of

a full single-cell cavity is planned for 2012. Full-cavity T-mapping will be employed during testing to identify spots with reduced RF performance, and any spots showing lower performance will be cut out of the cavity to identify coating defects and find a way to remedy them.

ACKNOWLEDGEMENTS

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