# AN INNOVATIVE Nb<sub>3</sub>Sn FILM APPROACH AND ITS POTENTIAL FOR SRF APPLICATIONS \*

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### Abstract

A novel electro-chemical technique to produce Nb<sub>3</sub>Sn films on Nb substrates was developed and optimized at Fermilab. The Nb<sub>3</sub>Sn phase is obtained in a two-electrode cell, by electrodeposition from aqueous solutions of Sn layers and Cu intermediate layers onto Nb substrates. Subsequent thermal treatments in inert atmosphere are realized at a maximum temperature of 700°C to obtain the Nb<sub>3</sub>Sn superconducting phase. Several superconducting Nb<sub>3</sub>Sn films were obtained on Nb substrates by studying and optimizing most parameters of the electro-plating process. Samples were characterized at Fermilab, NIMS, KEK and JLAB, including EPMA analyses, DC and inductive tests of critical temperature  $T_{c0}$ , and lower critical field  $H_{cl}(4.2)$ K) by SQUID. In parallel to sample development and fabrication at FNAL, at JLAB and KEK effort was put into etching and electro-polishing techniques adequate to remove the Cu and bronze phases from the samples' outer surface. This is necessary prior to measurements at JLAB of the surface impedance of flat samples in a setup that make use of an RF host cavity.

# **INTRODUCTION**

The two most important aspects of SRF performance are the accelerating field  $E_{acc}$  and the cavity quality factor,  $Q_{0.}$ The accelerating gradient in SRF cavities is proportional to the peak magnetic field on the cavity wall. At RF frequencies the peak magnetic field is limited by the metastable superheating field,  $H_{sh}$  [1]. The maximum accelerating gradient expected for Nb cavities in the TESLA shape, for instance, is ~40 MV/m. Nb<sub>3</sub>Sn is an attractive alternative because it has a higher  $T_{c0}$  of up to 18 K. With a theoretical  $H_{sh}$  of 0.42 T, as compared to a  $T_{c0}$  of 9 K and  $H_{sh}$  of 0.25 T for Nb [2], SRF cavities with a thin layer of Nb<sub>3</sub>Sn coated onto their inner surface should produce larger accelerating gradients than Nb. The larger  $T_{c0}$  of Nb<sub>3</sub>Sn allows for SRF cavities with a  $Q_0$  at 4.2 K about 30 times larger than for Nb. It also has the advantage of allowing the cavities to operate at 4.5 K rather than at ~2 K that is used for Nb cavities to obtain a higher gradient. This means less expensive refrigeration and more cryogenic economy and reliability.

Nb<sub>3</sub>Sn coated SRF cavities have been demonstrated to improve the cavity quality factor, but have not produced the theoretically expected gradient. Perhaps the longest

\* Work supported by U.S. DOE contract No. DE-AC02-07CH11359 † barzi@fnal.gov standing Nb<sub>3</sub>Sn technique for SRF is that of a vapor diffusion process followed by thermal reaction at high temperature to form the Nb<sub>3</sub>Sn [3, 4]. It consists of five stages, including degassing at 100° to 200°C to remove moisture; nucleation at intermediate temperature; ramp up to and holding at a coating temperature between 1100°C and 1300°C for Nb<sub>3</sub>Sn formation; and finally annealing to diffuse excess tin. This process has not produced so far full cavity assemblies because the scale-up from single cell is challenging. Nb<sub>3</sub>Sn coating on 5-cell CEBAF-type cavities was achieved at JLAB [5].

In this paper an alternate process, that of chemical electrodeposition, followed by reaction in inert atmosphere, will be described. The electroplating is performed at near room temperature and at atmospheric pressure. The advantage of electrodeposition is its simplicity, accurate control, and low costs. Also, it can be performed on any 3D surface such as the inner surface of SRF cavities. Electrodeposition should be among the least expensive ways to produce SRF cavities. It should also provide a more uniform Nb<sub>3</sub>Sn coating. Improving the quality of the Nb<sub>3</sub>Sn could reduce the large gap between the theoretical and measured  $H_{sh}$ .

# **EXPERIMENTAL METHOD**

An electro-chemical deposition technique, which had been collaboratively developed with Politecnico di Milano [6, 7] and is under patenting process, to produce superconducting Nb<sub>3</sub>Sn films on Nb substrates, was reproduced at Fermilab. In the past year, the equipment and consumables were procured, and the complete electro-chemical system was commissioned.

In electroplating the metallic coating is deposited on another metal surface through an electrolyte solution. The metal to be plated acts as the cathode and when the appropriate current is applied, positively charged ions traveling from the anode into the solution will discharge and get deposited on the cathode until a film of desired thickness is formed. Fig. 1, top shows a schematic with two symmetric anodes for bilateral coating of flat samples. Fig. 1, bottom shows the concept for scale-up to 3D surfaces, such as internal coating of cylinders, as first step to the internal coating of SRF cavities.

Figure 2 shows the sequence of deposited layers. First, using the Nb as cathode and Cu anode, a thin seed layer of

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Cu is deposited in an acid solution. The Cu lowers the formation temperature for the A15 compound and suppresses the unwanted NbSn<sub>2</sub> and Nb<sub>6</sub>Sn<sub>5</sub> phases. In a second electroplating step, the resulting Nb/Cu sample is used as cathode, and a thick layer of Sn is deposited with a Sn anode within a commercial Sn-rich solution. And finally, on the resulting Nb/Cu/Sn sample, a Cu layer is again deposited using a Cu anode. Each electrodeposition step is carried out at near room temperature and at atmospheric pressure. Details are illustrated in [7].

Nb<sub>3</sub>Sn is formed through solid diffusion by reacting the multi-layered samples in inert atmosphere (argon) using the temperature profile of Fig. 3, at a maximum temperature of 700°C.



Figure 1: Schematic of electro-chemical cell for bilateral coating of Nb flat samples (top), and internal coating of Nb cylinders (bottom).

 $\operatorname{Sn}$ 

Nh

#0

#2

#3

#1

Figure 2: Sequence of deposited layers (left), and pictures of sample at each deposition step (right).



Figure 3: Temperature profile during Nb/Cu/Sn/Cu solid diffusion.

Reacted Nb<sub>3</sub>Sn samples were characterized using an Electron Probe Microanalysis (EPMA) for elemental quantitative analysis with Wavelength-Dispersive Spectroscopy (WDS), transport test of critical current  $I_c$  (B) up to 14 T to determine the upper critical field  $\mu_0 H_{c20}$  as a free parameter in the  $I_c(B)$  data fitting, resistive and inductive critical temperature  $T_{c0}$ , as well as SQUID measurements of  $H_{cl}(4.2K).$ 

Reacted Nb<sub>3</sub>Sn film samples were also sent to JLab and to KEK for developing the appropriate etching and/or electropolishing techniques to remove the few micrometres of Cu and bronze phases at the samples' outer surface.

#### RESULTS

Several superconducting Nb<sub>3</sub>Sn films of thickness of up to 12 µm were obtained on Nb substrates of 0.3 mm to 0.5 mm thickness by studying and optimizing most parameters of the electro-plating process, including:

Bath composition and anode materials for each of the three deposition steps;

Current densities, deposition times, stirring rates, and cathode and anode relative orientation in DC mode;

Current densities, deposition times, stirring rates, cathode and anode relative orientation, pulse frequencies, and duty cycles in pulsed mode.

The pulsed mode was eventually chosen for all three deposition steps.



Figure 4: Cross-section of a sample with EPMA results (NIMS).

#### Figure

4 shows a picture of the longitudinal cross section of a sample (sample No. 23). EPMA was performed and allowed identification of bronze phases as well as Nb oxides on the outer surface of the Nb<sub>3</sub>Sn layer. Clearly, for SRF operation these residues must be removed by either etching, electropolishing or mechanically. Fig. 5 shows the  $T_{c0}$ result of 17.5 K (defined as in [7]) obtained for the same sample as measured resistively. The same sample was also measured by SQUID (Fig. 6) and the resulting  $T_{c0}$  was 17.6 K. The two results are remarkably consistent. Fig. 7 shows

> **Technology** Superconducting RF

514

the *M-H* curve obtained with the SQUID magnetometer. The  $H_{cl}(4.2K)$  for this sample was 600 Oe.



Figure 5: DC test at FNAL of  $T_{c0} = 17.5$  K of a Nb<sub>3</sub>Sn film sample.



Figure 6: *M-T* curve obtained with SQUID magnetometer at NIMS. The  $T_{c0} = 17.6$  K.



Figure 7: *M*-*H* curve obtained with SQUID magnetometer at NIMS. The  $H_{c1}(4.2 \text{ K}) = 600 \text{ Oe}$ .

For this sample, an upper critical magnetic field,  $\mu_0 H_{c20}$ , of 23.2 T was obtained as a free parameter in the  $I_c$  (*B*) data fitting. The measured  $T_{c0}$  value of 17.5 K was used as a fixed parameter in the fit.

It is important to know how well these thin film samples would perform in a cavity. A TE011 cavity with a demountable endplate, where a disc sample can be placed, can be used for the measurement of the surface impedance. A cavity test facility of this type exists at JLab [8].

### **CONCLUSIONS**

High gradient, inexpensive superconducting cavities (SRF) will be needed for future accelerators. However, the accelerating field of Nb cavities is limited by the peak magnetic field on the cavity surface. Nb<sub>3</sub>Sn SRF cavities should produce larger accelerating gradients and a larger quality factor,  $Q_0$ , than Nb cavities. Also, the higher  $T_{c0}$  of Nb<sub>3</sub>Sn allows cavities to operate at 4.5 K rather than ~2 K that is used for Nb cavities to obtain a higher gradient. This means less expensive refrigeration and more cryogenic reliability.

Nb<sub>3</sub>Sn coated SRF cavities, with Nb<sub>3</sub>Sn produced by vapor diffusion followed by a thermal reaction at high temperature, have achieved only a fraction of the theoretical predicted gradient. Nb<sub>3</sub>Sn plated cavity performance may be improved by using electrochemical deposition. The advantages are its simplicity, accurate control, and low costs. Electrodeposition should be among the least expensive ways to produce SRF cavities, and it should provide a more uniform Nb<sub>3</sub>Sn coating. Improving the quality of the Nb<sub>3</sub>Sn could reduce the large gap between the theoretical and measured  $H_{sh}$ . Electrochemical techniques may also eliminate the problem of a low Sn concentration at the Nb interface, which is a fundamental limit of the current vapor deposition techniques. Lastly, electrodeposition can be performed on any 3D surface such as the inner surface of SRF cavities. Nb<sub>3</sub>Sn plating experiments of Nb cylinders are under way.

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