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# Nb<sub>3</sub>Sn THIN FILM FOR THE PRODUCTION OF HIGHER GRADIENT SRF CAVITIES AT REDUCED COSTS\*

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## Abstract

High gradient superconducting cavities (SRF) will be needed for future accelerators. The higher gradient can achieve the high energy with fewer cavities. However, the accelerating field of niobium cavities is limited by the peak magnetic field on the cavity surface. Cavities coated with Nb<sub>3</sub>Sn have a significantly larger H<sub>c2</sub>, allowing the cavity to achieve a larger gradient. Measurements of Nb<sub>3</sub>Sn coated cavities have achieved about half the theoretical predicted gradient. It is possible to improve Nb<sub>3</sub>Sn plated cavity performance.

## INTRODUCTION

Niobium has been used as a fabrication material for superconducting cavities. Much R&D on niobium SRF cavities has been done over the years [1], but there are fundamental limitations to the performance of these cavities. 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<sub>sh</sub> [2]. Based on the limiting value of the peak magnetic field for niobium cavities the maximum accelerating gradient is ~40 MV/m. Nb<sub>3</sub>Sn is an attractive alternative because it has a high T<sub>c0</sub> = 18 K and H<sub>c20</sub> ≈ 30 T. SRF cavities with a layer of Nb<sub>3</sub>Sn coated on the cavity surface have been demonstrated to improve the cavity quality factor, but have not significantly improved the gradient. Most experiments to fabricate SRF cavities with a Nb<sub>3</sub>Sn coating have used a vapor diffusion process followed by a thermal reaction at high temperature to form the Nb<sub>3</sub>Sn compound to coat the cavity [3, 4]. A review of the SRF cavities coated with Nb<sub>3</sub>Sn using the vapor diffusion process is given in Ref. [5]. In this paper we will examine an alternate process, where an electrodeposition of copper and tin layers onto niobium, followed by reaction in inert atmosphere, forms a Nb<sub>3</sub>Sn film on the niobium substrate. Electrodeposition is likely to be less expensive than the vapor deposition technique previously mentioned.

## Nb<sub>3</sub>Sn SRF CAVITIES

SRF cavities with a thin layer of Nb<sub>3</sub>Sn coated onto the inner surface of the cavity should produce larger accelerating gradients as well as larger cavity quality factor, Q<sub>0</sub>, than niobium cavities. Figure 1 shows a theoretical comparison of Q<sub>0</sub> for 1.3 GHz Nb<sub>3</sub>Sn and Nb cavities which illustrates the effect of larger T<sub>c</sub> of Nb<sub>3</sub>Sn on Q<sub>0</sub>. Also the larger T<sub>c</sub> of Nb<sub>3</sub>Sn has the advantage of allowing the cavities to operate at 4.5 K rather than ~2 K that is used

for niobium cavities to obtain a higher gradient. This means less expensive refrigeration and more cryogenic reliability.

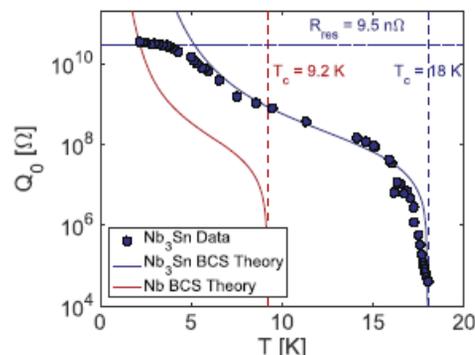


Figure 1: Calculation of Q<sub>0</sub> for 1.3 GHz SRF cavities with Nb<sub>3</sub>Sn and Nb inner surfaces. From Ref. [5].

## VAPOR DIFFUSION TECHNIQUE

The vapor diffusion technique has been largely successful for coating Nb<sub>3</sub>Sn onto the niobium surface of a cavity [5] and has been the preferred technique for Nb<sub>3</sub>Sn SRF cavities. The method does require a higher temperature for the heat treatment reaction. For best performance pure Nb<sub>3</sub>Sn is formed at temperatures greater than 950° C in a Nb-Sn mixture with tin atomic fraction between 23% to 26%. At temperatures below 950° C there are contaminations from NbSn<sub>2</sub> and Nb<sub>6</sub>Sn<sub>5</sub> which are not superconducting at 4.5 K. The vapor diffusion process goes through five stages, although there are some variations in the process at different laboratories. These stages are summarized in Fig. 2 and include:

1. A degassing stage at 100° to 200°C where pumping on the chamber removes moisture.
2. A nucleation stage where sites are created on the chamber surface. This stage is performed at an intermediate temperature. A nucleation agent such as SnF<sub>2</sub> or SnCl<sub>2</sub> is used to provide uniformity of the nucleation sites.
3. A ramp up to the coating temperature.
4. The coating stage where the cavity is held at a constant temperature above 950° C where Nb<sub>6</sub>Sn<sub>5</sub> and NbSn<sub>2</sub> formation is disfavored. The temperature of the tin source must be high enough to provide a uniform coating, which can be several hundred degrees higher than the cavity surface.
5. The annealing stage to allow the excess tin at the

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surface to diffuse into the niobium layer and form Nb<sub>3</sub>Sn. This stage must be above 950°C.

The high temperature used in the vapor diffusion process can make the Nb<sub>3</sub>Sn coated layer brittle, which can limit the strain that can be tolerated. This can locally reduce its superconductivity.

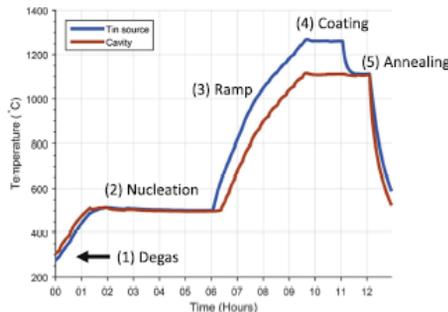


Figure 2: Temperature profile during the coating process used at Cornell. From Ref. [5].

### ELECTROCHEMICAL DEPOSITION

Electrochemical deposition techniques can provide an alternate approach to Nb<sub>3</sub>Sn coatings on SRF cavities. Electrochemical deposition techniques using a SnCl<sub>2</sub> molten salt eutectics to produce Nb<sub>3</sub>Sn coatings for cavities has previously been explored [6]. That approach required temperatures higher than 370° C during the plating process at very low pressures. We are proposing to use a novel electro-chemical technique that had been jointly developed by Politecnico di Milano and FNAL [7, 8], and further improved at FNAL to deposit tin and copper layers from aqueous solutions onto a niobium substrate. The most recent improved technique increased the T<sub>c0</sub> of the Nb<sub>3</sub>Sn films from 17.1 T to 17.7 K. Figures 3 and 4 show the T<sub>c0</sub> results obtained for one of the film samples using a resistance and a SQUID measurement respectively. The H<sub>c1</sub>(4.2K) for this samples was measured to be 550 Öe.

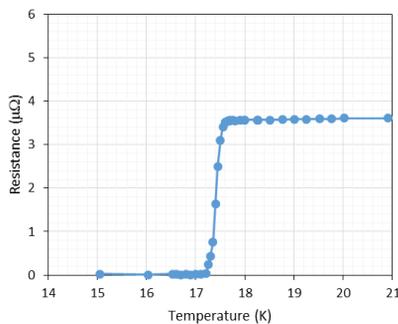


Figure 3: DC test at FNAL of T<sub>c0</sub> = 17.6 K of a Nb<sub>3</sub>Sn film sample obtained with the latest improved technique.

This approach allows the electroplating to be performed at near room temperature and at atmospheric pressure. The advantage of electrochemical deposition is its simplicity, accurate control, and low costs. Electrodeposition should be among the least expensive ways to

produce SRF cavities. Furthermore, electrochemical techniques can eliminate the problem of a low tin concentration at the niobium interface, which is a fundamental limit of the current vapor deposition techniques. Electrodeposition can be performed on any 3D surface such as the inner surface of SRF cavities.

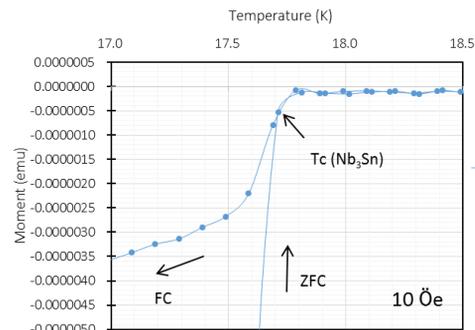


Figure 4: M-T curve obtained with SQUID magnetometer at NIMS. The T<sub>c0</sub> = 17.7 K.

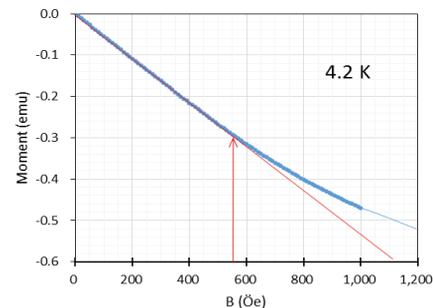


Figure 5: M-H curve obtained with SQUID magnetometer at NIMS. The H<sub>c1</sub>(4.2 K) = 550 Öe.

We are proposing to use the electrochemical technique that has been developed for niobium substrates, along with thermal solid diffusion, to put a Nb<sub>3</sub>Sn coating onto a niobium surface. The electroplating procedure plates a layer of copper onto the niobium substrate followed by a tin layer onto the copper. Tin does adhere well to the copper layer. A second copper layer is applied as a barrier layer during formation of the liquid phases. The electrodeposition procedure is carried out at near room temperature and at atmospheric pressure. The procedure is described in detail in Ref. [8]. The most efficient method of forming Nb<sub>3</sub>Sn is through solid diffusion. The temperature profile during solid diffusion which is shown in Fig. 6 is used to control the reaction process. Initially the temperature is set just below the tin melting point to relax stresses in the metal layers and start diffusion between copper and tin. The second step raises the temperature to 450° C to allow the formation of a liquid tin phase and start the inter-diffusion with niobium and copper. This avoids the movement of the material boundaries. The final temperature step to 700° C for 24 hours forms the Nb<sub>3</sub>Sn superconducting phase. With the ternary Nb-Cu-Sn system only Nb<sub>3</sub>Sn remains after the heat treatment [8]. The diffusion from the Cu-Sn solid solution to the

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Nb-Sn solution passes through compounds with the A15 stoichiometric ratio such as the  $Nb_3Sn$  compound [9]. The copper lowers the formation temperature for the A15 compound and suppresses the unwanted  $NbSn_2$  and  $Nb_6Sn_5$  contaminants. This temperature reduction is one of the advantages of the electrochemical deposition/solid diffusion method.

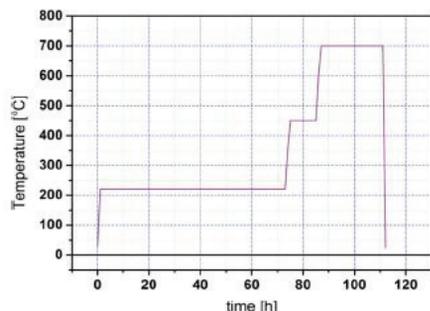


Figure 6: Temperature profile during Nb/Cu/Sn/Cu solid diffusion.

## THEORETICAL PREDICTION

The two most important aspects of SRF performance are the accelerating field  $E_{acc}$  and the cavity quality factor,  $Q_0$ .  $E_{acc}$  is limited by the peak magnetic field reaching or exceeding the superheated critical magnetic field,  $H_{sh}$ . The superheated metastable critical limit is possible because the RF period is much shorter than the relaxation time of the Meissner stage. A theoretical estimate of  $H_{sh}$  can be made using the one-dimensional Ginzburg-Landau equation [10]. This estimate is however most valid for the range where the temperature is near the superconducting critical temperature. Figure 7 shows a comparison of niobium and  $Nb_3Sn$  measurements with the G-L model predictions. The figure shows reasonable agreement for the niobium measurements with the theory for  $T > 0.5 T_c$ . However, the  $Nb_3Sn$  measurements are much lower than the predictions. The difference between niobium and  $Nb_3Sn$  is not well understood. The coherence lengths of niobium and  $Nb_3Sn$  are quite different (which is also reflected in  $H_{c2}$ ).  $Nb_3Sn$  is a more granular material than the defect free niobium that is used for SRF cavities. The electrochemical deposition should provide a more uniform  $Nb_3Sn$  coating. Improving the quality of the  $Nb_3Sn$  could reduce the large gap between the theoretical and measured  $H_{sh}$ .

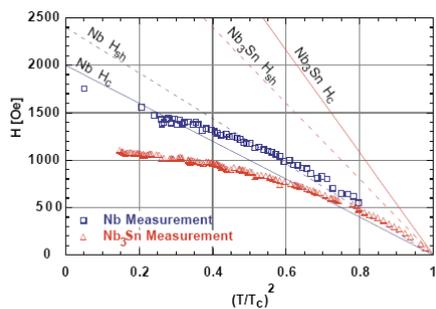


Figure 7: Comparing Nb and  $Nb_3Sn$  measurements with  $H_{sh}$  predictions [10].

## RESEARCH PROPOSAL

As an R&D effort we would like to use thin film  $Nb_3Sn$  samples to study processing properties. These samples can be fabricated, processed and tested with rapid turnaround as a cost-effective approach to optimizing the electrodeposition process to uniform plated layers with a minimum number of defects. The microstructure of the plated layers can be examined. Glow-discharge optical emission spectroscopy (GDOES) can be used to study tin, copper and niobium composition of the layers. X ray diffraction (XRD) can reveal the cubic crystalline structure of the  $Nb_3Sn$  phase that is expected for an A15 compound. XRD is sensitive to the  $NbSn_2$  contamination phase. A scanning electron microscope (SEM) can be used to produce cross section images of the plated layers with micron accuracy for layer thickness. Figure 8 shows an SEM image of a  $Nb_3Sn$  sample from Ref. [8]. The  $Nb_3Sn$  layer has a thickness of  $5.7 \pm 0.5 \mu m$  on the niobium substrate. Outside the  $Nb_3Sn$  layer is a bronze layer formed from the excess tin alloyed with the copper protective layer which should be removed through etching and electropolishing. The measurement of the  $Nb_3Sn$  layer thickness and grain size are important data for the sample characterization.

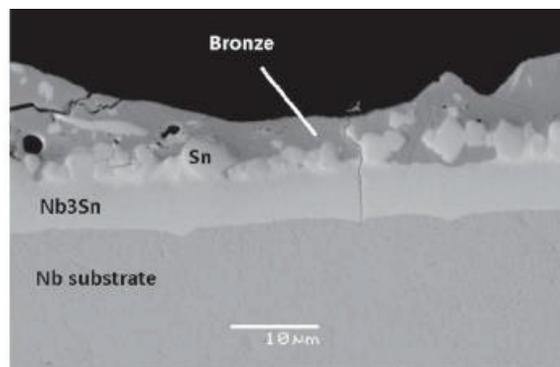


Figure 8: SEM cross-section of a sample analysed in [8].

It is important to know how well these thin film samples would perform in a cavity. A test facility that can include the  $Nb_3Sn$  thin film sample in the cavity resonant circuit would be needed. This would allow the measurement of the surface impedance which can be obtained from the measurement of  $Q_0$ . A TE011 cavity with a demountable endplate where the sample can be placed can be used for the measurement of the surface impedance. A cavity test facility of this type exists at Jlab [11].

## CONCLUSIONS

We have presented a prescription for a more cost-effective approach to coating niobium RF cavities with a  $Nb_3Sn$  layer. In this process, electrodeposition of copper and tin layers onto a niobium substrate is followed by a solid diffusion heat treatment at  $700^\circ C$ . This procedure should be less expensive than the vapor diffusion process that is typically used.

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