

DEVELOPMENT OF A Nb₃Sn CAVITY VAPOR DIFFUSION DEPOSITION SYSTEM*

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Abstract

Nb₃Sn is a BCS superconductors with a superconducting critical temperature higher than that of niobium. The feasibility of technology has been demonstrated at 1.5 GHz with Nb₃Sn vapor deposition technique at Wuppertal University [1]. The benefit at these frequencies is more pronounced at 4.2 K, where Nb₃Sn coated cavities offer RF resistances an order of magnitude lower than that of niobium. At Jefferson Lab we started the development of Nb₃Sn vapor diffusion deposition system within an R&D development program towards compact light sources. Here we present the recent progress of the coating system development.

INTRODUCTION

The focus of the Basic Energy of Science's Inverse Compton Source (BES-ICS) project R&D is on development of novel superconducting structures that would operate at 4.5 K. The quadratic frequency dependence of niobium surface resistance in superconducting state drives the useful frequency for ICS cavities into mid- to lower-hundreds MHz range. The drawback is the increase in size of such cavities and consequently in the size of a cryomodule. As an alternative to niobium, we looked into the possibility of using other superconductors with higher critical temperatures. Nb₃Sn has been identified as the most promising candidate among other materials for a number of reasons: it is most widely researched other than niobium superconductor for accelerating cavity application, quality factors above those of niobium at 4 K has already been demonstrated on accelerating structures, and there is ongoing work on this superconductor at other institutions.

Here we will present the current status and future plans for Nb₃Sn development at Jefferson Lab.

BACKGROUND

Superconductivity in Nb₃Sn was first discovered by Bernd Matthias [2], who used liquid Sn over Nb powder in a closed-off quartz tube to form the compound. Nb₃Sn has thermal conductivity about 1000 times lower than that of niobium and also poor formability, which precludes its use

as a sheet material. Fortunately, Nb₃Sn has thermal expansion coefficient ($9.8 \cdot 10^{-6}$ 1/K at 20 °C) close to that of niobium ($7.6 \cdot 10^{-6}$ 1/K at 20 °C), which allows to use Nb₃Sn as a layer on top of niobium. A number of techniques have become available to deposit Nb₃Sn layers: chemical vapor deposition, co-evaporation, vapor diffusion, ion plating, tin bath submergence. The early work on the compound included Sn chemical vapor deposition at RCA, co-evaporation at GE [3], and vapor deposition and annealing at BNL [4]. However, the first successful results with superconducting RF cavities [5–10] usually credit Nb₃Sn deposition method presented by Saur and Wurm [11]. Initially, a sealed-off quartz tubes with Nb and Sn inside were used to produce parts of Nb₃Sn resonator. It was found that the quartz tubes cause contamination of the coated surfaces, and so the research moved on to all-niobium chambers. There were two persistent problems that had to be dealt with. One problem was Sn 'droplets' that sometimes formed on the surface and had poor superconducting properties, the other problem was the absence of complete coating on the Nb surface.

The Sn 'droplets' turned out to be hard to deal with, because they could not be dissolved in HNO₃ or HCl acid. Stimmell [10] noted a blue tinge to those droplets when they were anodized to 30 Volts, which suggests dissolved Nb in the 'droplet'. A number of methods were used to remove the droplets: FeCl₃ + CuSO₄ + acetic acid at 50 °C with ultrasonic agitation, alternate immersion HNO₃ and HF at room temperature with ultrasonic, annealing the material at high temperatures, and boiling in 37 % HCl to remove the droplets, but all treatments resulted in inferior surfaces [10, 12]. It appears from these efforts that the best practice is to avoid these features altogether. This is related to the second problem, which is achieving a whole surface coating.

The solutions devised early at Siemens to create a continuous Nb₃Sn coating on Nb surface was to anodize the Nb surface up to 100 Volts prior to deposition. In addition, Pfister et al. proposed to use SnCl₂ or SnF₂ in the deposition process. Siemens group speculated that the presence of an anodic oxide film on the Nb substrate prevents reaction of the substrate with condensing Sn vapor during the early stages of the firing cycle and allows a continuous Sn film to accumulate before diffusion into the Nb begins, but the mechanism of initial stages of Nb₃Sn formation has never been clearly understood. In the later works at Wuppertal University, the anodization voltage was reduced to 10 Volts

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to prevent RRR degradation due to dissolution of oxygen from the anodization layer.

SAMPLE COATING

We chose the Sn vapor diffusion process because of its simplicity and the former results with Sn vapor diffusion process at Wuppertal University. In Fig.1 one of the best results with superconducting Nb structures coated with Nb₃Sn at Wuppertal University are shown [1]. The experiments were done with 1.5 GHz CEBAF-shape cavities. At 2 K, the niobium cavity results, shown with crosses, are slightly worse than Nb₃Sn coated cavity at low fields, but the niobium cavity has a higher quality factor and reaches higher fields than the coated cavity. However, Nb₃Sn cavity quality factor at 4.2 K shown in the picture with the solid squares is a factor of 50 higher than that of niobium cavity shown in the lower right corner for comparison.

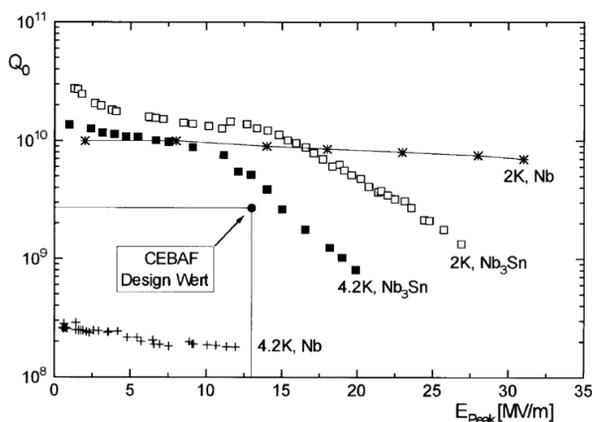


Figure 1: One of the best results with Nb₃Sn coating on accelerating structures [1]. The crosses present the data for 1.5 GHz niobium cavity at 2 and 4 K. The squares show the data for Nb₃Sn coated cavities at 2 and 4 K. Note that the quality factor of Nb₃Sn cavity is a factor 50 better than that of niobium one at 4 K.

As a first step of the project we decided to coat several samples using the setup provided by P. Kneisel. In Fig. 2 we show setup components. An all niobium chamber with pump out tube hosted two niobium flat 2" samples for material characterization and surface impedance measurements, and one coaxial sample for penetration depth measurements. Flat samples were supported by a niobium holder, while coaxial sample was mounted on a tungsten wire. In the bottom of the chamber a cut-out was made to hold high purity tin shots and SnCl₂ powder. To prevent direct contact between the tin and samples, tin and SnCl₂ were separated from the samples with high purity Nb foil.

After assembly the chamber was put into one of the JLab existing furnaces. Guided by Wuppertal experience with Nb₃Sn deposition we chose to hold the furnace at 500 °C for one hour so that SnCl₂ vapors (melting temperature 247 °C, boiling temperature 623 °C) deposit onto the samples' surface and create Nb₃Sn nucleation centers. After



Figure 2: Components for Nb₃Sn deposition on samples at Jefferson Lab.

that the temperature was ramped for two hours to 1200 °C and held at the temperature for 2 hours. After the cycle was finished, the heating was turned off and the chamber cooled down naturally.

After deposition, the chamber was removed from the furnace and cut open. All samples exhibited complete surface coating with Nb₃Sn, however two of the samples, the coaxial sample and the large grain 2" flat sample, had Sn 'droplets' on the surface. The other flat sample had complete coating and no visible features on the surface, see the top left corner in Fig. 3. The coaxial sample was placed into an existing JLab sample measurement system, and the transition temperature was measured. In Fig. 3 the plot shows the resonant frequency change as a function of temperature for a Nb sample and for our Nb₃Sn coated sample. The frequency change is proportional to the field penetration change and shows the temperature dependence expected from the theory. The mid-point transition temperature of coated Nb₃Sn was measured to be 17.85 K.

Prior studies on the Nb₃Sn compound has established empirical functional dependencies between the transition temperature, energy gap, lattice parameter, and low temperature normal conducting resistivity, which were reviewed in [14]. From the empirical relation between the transition temperature and atomic Sn concentration, we derive that critical temperature of 17.85 K corresponds to $\beta = 24.94$ atomic percent of Tin on the sample's surface. And from the empirical relation between the atomic Sn percent and low temperature normal conducting resistivity, we derive that the surface is expected to have $\rho = 4.9 \mu\Omega\text{cm}$, which is about only 40 percent higher than the best measured value of $3.4 \mu\Omega\text{cm}$.

The best flat sample was measured in the JLab Surface Impedance Characterization (SIC) system. The results show the transition temperature of about 17.9 K consistent with coaxial sample measurements, and surface resistance was about $20 \mu\Omega$ at 4 K, which increases to about $40 \mu\Omega$ at 10 K [13].

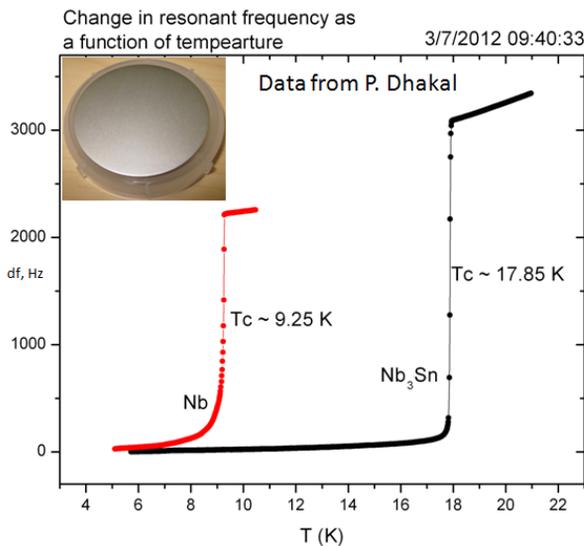


Figure 3: Test results on the coaxial sample. The data shows that the transition temperature of the coaxial sample was 17.95 K. In the top left corner the surface of the best sample is shown. Note, that on the y-axis $\Delta f(T) = f(T_0) - f(T)$, where $f(T_0)$ is the resonant frequency at about 4 K, is plotted.

CAVITY DEPOSITION SYSTEM

Since at the project inception there was no vertical furnace available at Jefferson laboratory for the project (both Wuppertal and Cornell group used vertical furnace), it was decided to build a horizontal insert that can be later converted to be used in a vertical furnace ordered from T&M vacuum company. For horizontal runs we used the so-called 'Big Blue' furnace. Following suggestion from Stimmell's dissertation we decided to implement a vapor guide to control vapor distribution. The crucible has cooling a He gas line, and a heater is planned to vary the Sn crucible temperature ± 100 °C from the furnace temperature. The insert design as well as the as-built insert are shown in Fig. 4. For the first runs we built a niobium cavity that hosts 10 samples. The two halves of the cavity are spot welded together, so that the cavity can be easily taken apart. The first run in the horizontal furnace was done with

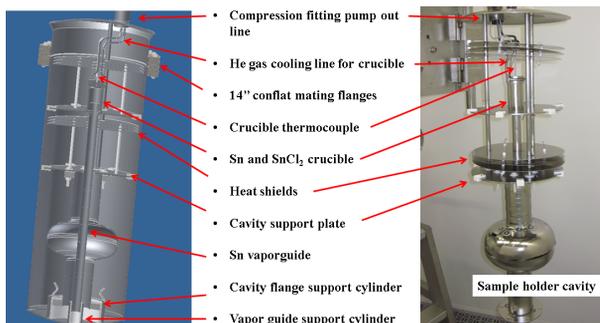


Figure 4: In this plot the insert design (to the left) and as-built insert without niobium retort (to the right) are shown.

out Sn. The run was composed of several ramp and hold temperature steps. Eventually, the furnace was run at 1200 °C for two hours. At the highest temperature, room temperature helium gas was run through the cooling line around the crucible. The thermocouple on the crucible indicated about 50 °C drop when He gas was run through the cooling line.

Following the Sn-free run, the insert has been cut apart by wire EDM, degreased, loaded with Sn and SnCl₂, and welded back together for Nb₃Sn sample coating in the 'Big Blue'. Ten samples have been arranged on the inside of the test cavity inside the insert.

Concurrently with the horizontal insert testing, we have procured a new vertical furnace from T&M vacuum company designed for Nb₃Sn coating using the built insert. The vertical furnace bought from T&M vacuum for investigations of Nb₃Sn coatings on 1.3 GHz 1-cell cavities was delivered in the end of August 2013 and is being commissioned.

SUMMARY

Alternative BCS superconductors with the superconducting critical temperature higher than that of niobium theoretically surpass the limitations of niobium. The feasibility of technology has been demonstrated at 1.5 GHz with Nb₃Sn vapor deposition technique at Wuppertal University. The benefit at these frequencies is more pronounced at 4.2 K, where Nb₃Sn coated cavities show RF resistances an order of magnitude lower than that of niobium. At Jefferson Lab we started the development of Nb₃Sn vapor diffusion deposition system within an R&D development program towards compact light sources. Coatings on niobium samples showed promising results and 1.3 GHz 1-cell cavity insert has been built and tested in the existing horizontal furnace. A new furnace for Nb₃Sn coating development has been procured and is being commissioned at Jefferson Lab.

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