

# EXPERIMENTAL STUDY OF NUCLEATION FOR Nb<sub>3</sub>Sn DIFFUSION COATINGS ON NIOBIUM SRF CAVITIES\*

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## Abstract

Nb<sub>3</sub>Sn has the potential to achieve superior performance both in terms of operating temperature (4.2 K vs 2 K) and accelerating gradient resulting in significant reduction in both capital and operating costs of SRF linacs. Cavity interior surface coatings are obtained by two step vapor diffusion: nucleation followed by deposition. To gain more understanding of nucleation and its effect on the subsequent coating, we investigated the effect of varying parameters in a typical tin/tin chloride process. We report findings obtained by SEM/EDS, AFM, SAM and other materials characterization approaches.

## INTRODUCTION

Nb<sub>3</sub>Sn is an alternative superconducting material that has potential to replace traditional niobium in SRF cavities. Critical temperature and predicted superheating field of Nb<sub>3</sub>Sn are nearly twice that of niobium which offers the possibility of attaining higher operating temperature and accelerating gradient resulting in significant reduction in both capital and operating cost. Nb<sub>3</sub>Sn is a challenging material because of lower thermal conductivity and brittleness. That restricts its application as a cavity material to only coating form. Fabrication of Nb<sub>3</sub>Sn cavities by deposition of Nb<sub>3</sub>Sn diffusion coatings on niobium cavities dates back to 1970's [1-3], and is the most successful technique so far. Recent performance results of such cavities are very promising, attaining quality factor >10<sup>10</sup> operating at 4.2 K with gradient more than 10 MV/m [4].

The majority of research institutions working currently to develop Nb<sub>3</sub>Sn coated cavities use the diffusion coating technique [5-7]. The essence of the process is to transport tin vapour to the niobium substrate, and provide the high temperature environment to form Nb<sub>3</sub>Sn phase, which is determined by binary phase diagram [8]. This coating process is favourable to complicated shaped cavities. It involves two steps: nucleation and deposition. Tin chloride, used as a nucleation agent evaporates at about 500°C to deposit tin on niobium surface which later acts as Nb-Sn nucleation sites to form Nb<sub>3</sub>Sn phase [9]. It is assumed that these nucleation sites grow with influx of tin vapour at higher temperature. An example of Nb<sub>3</sub>Sn coating process at Jlab is shown in Figure 1. The first temperature plateau at nucleation temperature of 500°C is dedicated to nucleation process whereas second temperature plateau for deposition.

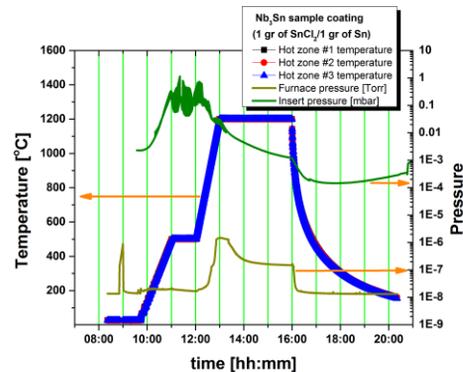


Figure 1: Temperature profile used for coating Nb<sub>3</sub>Sn on niobium samples.

The purpose of this work was to gain insight into the nucleation process, and to determine the effects of changing nucleation parameters. We varied nucleation temperature and/or duration at which nucleation temperature was kept constant for same amount of tin and tin chloride. We also investigated the effect of changing the amount of tin chloride in few cases.

## EXPERIMENTAL DESCRIPTION

### Materials

The samples here were high purity niobium coupons prepared by EDM cutting 3mm thick, high RRR (~300) sheet material of the type used to fabricate SRF cavities. All were subjected to buffered chemical polishing (BCP) with expected minimum removal of 50 μm. These samples further received metallographic polishing, also known as nanopolishing (NP) to obtain smoothest possible surfaces. The average roughness of NP samples was ~2 nm which was measured from 50 μm x 50 μm scan areas using an atomic force microscope (AFM).

### Experiments

The same deposition system that is available at Jlab to coat Nb<sub>3</sub>Sn on niobium cavities was used in this work. Detailed description of deposition system is available [5]. Experimental setup was very similar to the one used before to deposit Nb<sub>3</sub>Sn coatings [10]. One gram of tin and required amount of tin chloride were packaged in niobium foil, and placed on the niobium plate which covered the bottom side of sample chamber. Both ends of the sample chamber were covered by niobium plates after mounting coupons inside, before installing into the furnace insert. The insert was pumped down to attain pressure of 10<sup>-5</sup> – 10<sup>-6</sup> Torr range before the heating profile was initiated. The temperature inside the insert was raised at the rate of

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6 °C/min until it reached the nucleation temperature of 300 °C, 400 °C, 450 °C or 500 °C. These temperatures were maintained constant for different time periods before ceasing the heating process. Once cooled down to room temperature, the deposition system was purged with nitrogen to regain atmospheric pressure. The sample chamber was then taken out to remove the nucleated samples.

*Characterization*

A field emission scanning electron microscope (FE-SEM) with energy dispersive X-ray spectroscopy (EDS) was used to examine the nucleated samples. SEM images were taken and visible features were analyzed with EDS. We used a commercial AFM in tapping mode to obtain information of surface at higher resolution. Surface sensitive elemental analysis was done using X-ray photoelectron spectroscopy (XPS), and also used scanning Auger microscopy (SAM) in some samples.

**RESULTS AND DISCUSSION**

*Temperature Variation*

Nucleation temperature was varied from 300 °C to 500 °C keeping a constant nucleation time of an hour. All the tin chloride (1 g) was evaporated when the nucleation temperature was 450 °C or higher, otherwise surplus tin chloride was found following the experiment. Variation of nucleation temperature was found to produce different surfaces. SEM images of some of those obtained surfaces can be seen in Figure 2.

Nucleation temperature of 300 °C only created thin particles on niobium surface. EDS was not able to detect anything other than niobium. Surface looked very different when the sample treated at 400 °C for an hour, often showing some large tin particles. Nucleation temperature of 450 °C and 500 °C created similar looking surfaces with visible tin particles. Size and distribution of tin particles was slightly different between these two temperatures. EDS was unable to detect any tin in between these visible tin particles. Calculated surface coverage of these particles from SEM images was always less than 10%.

Samples from each experiment were examined using XPS with 200 μm x 200 μm survey scans. The surfaces were always found to be covered with carbon and oxygen which is expected to come from post-experiment handling. The samples were then pre-sputtered lightly and scanned again. Obtained result from samples with nucleation temperature 400°C or higher showed an average of (20-30) % tin indicating more tin in between tin particles shown in Figure 2. XPS normally accounted more tin than SEM/EDS analysis on each sample. XPS analysis was not able to detect any of chlorine in any of those samples.

We analyzed samples from the experiment with nucleation temperature of 500°C for an hour using high resolution SAM. Elemental mapping of tin in SEM image SAM is shown in Figure 3, which shows tin coverage consistent with the XPS result, and shows that there is tin on the surface in between the visible particles in SEM image. Background between tin particles was found more

tin enriched than XPS analysis in some cases. One such example of elemental comparison of tin particle and background is also shown in Figure 3.

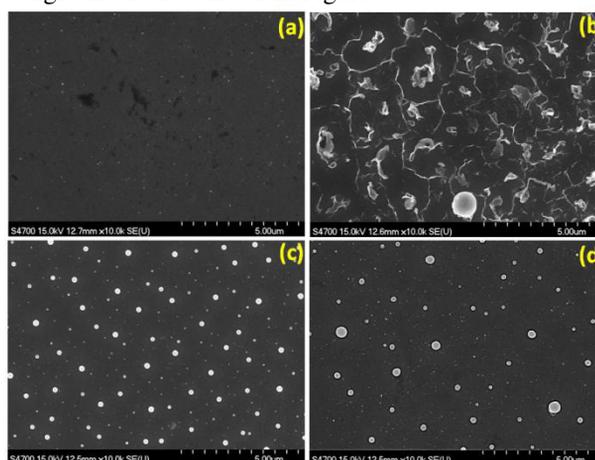


Figure 2: SEM images obtained from samples activated at nucleation temperature of (a) 300 °C, (b) 400 °C, (c) 450 °C, and (d) 500 °C with constant nucleation time of an hour.

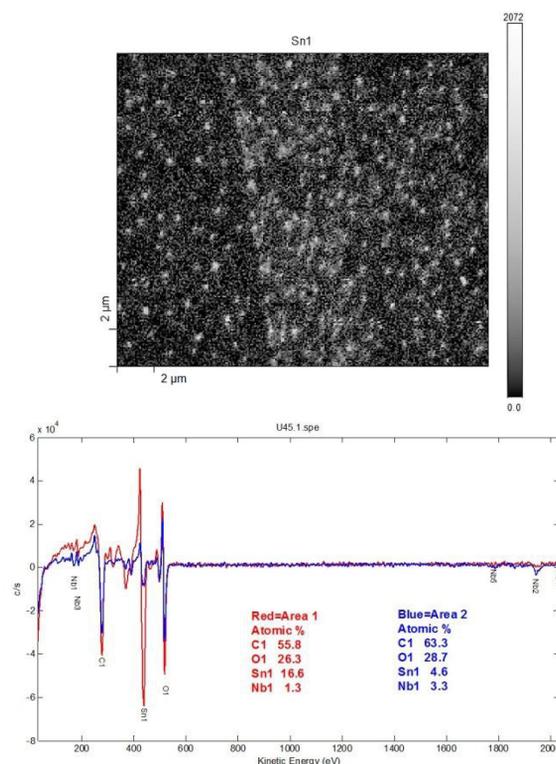


Figure 3: SAM elemental mapping of tin coverage after sputtering for one minute is shown in top image. The brighter areas are richer in tin than darker area as shown in intensity scale at right. Elemental composition comparison of particle (red area) and background (blue area) is shown in bottom spectra.

AFM images were captured from samples with different nucleation profiles. AFM images were able to reveal more features in between tin particles consistent with SAM than SEM observation as shown in Fig. 4.

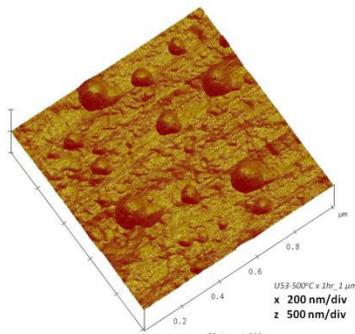


Figure 4: AFM image obtained from sample treated at 500 °C for an hour. Note that scan size is only 1 μm x 1 μm.

### Variation of Time at 500 °C

We varied nucleation period keeping nucleation temperature constant at 500 °C. It was found that five minute at 500 °C was not enough to evaporate all the tin chloride (1 g), but was able to produce some tin particles already. The surface was covered with frizzy features as shown in Figure 5, image to the right. Result obtained after an hour at the same temperature is discussed already in previous section. Surface produced after four hour at 500 °C is also shown in Figure 5. Comparing with result obtained after an hour at 500 °C, four hour at same temperature appears to produce more tin particles, but size of these particles are smaller with more coverage on surface. EDS was only able to detect tin from visible tin particles, but once again XPS analysis showed more tin than SEM/EDS analysis accounted.

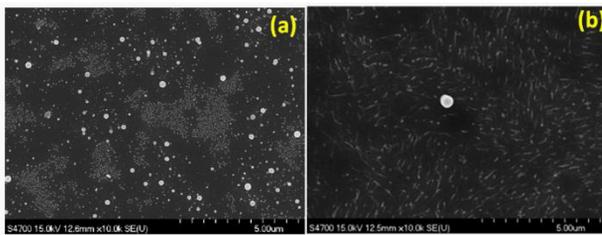


Figure 5: SEM images from samples obtained from experiments with nucleation temperature of 500 °C for (a) 4 hours and (b) 5 minutes.

### Low Vs High Amount of Tin Chloride

The diffusion coating of Nb<sub>3</sub>Sn coating recipe was first developed at Siemens AG where a very small amount of tin chloride was used to avoid nucleation problem. Recent recipes from in Jlab and Cornell use different amount of tin chloride to produce Nb<sub>3</sub>Sn coatings. We compare the nucleated surfaces produced by using Jlab recipe (3 mg/cm<sup>2</sup> of tin chloride, 500 °C for an hour, temperature ramp at 6 °C/min) and Cornell recipe (7 μg/cm<sup>2</sup> of tin chloride, 500 °C for five hours, temperature ramp at 3 °C/min) and corresponding final coating, as shown in Figure 6. SEM images shows uniform distribution of particles produced with low amount of tin chloride. These particles are very thin and small compared to the nucleation produced with a higher amount of tin chloride. XPS analysis shows comparable tin coverage in nucleated

samples obtained from both of those experiments. It is clear that more tin chloride produces bigger tin particles with similar tin coverage on niobium surface. The longer nucleation time in Cornell recipe is speculated to have an important role to produce such a good coverage of tin with a small amount of tin chloride. Examined by SEM/EDS, composition and structure obtained by both recipes were found very similar.

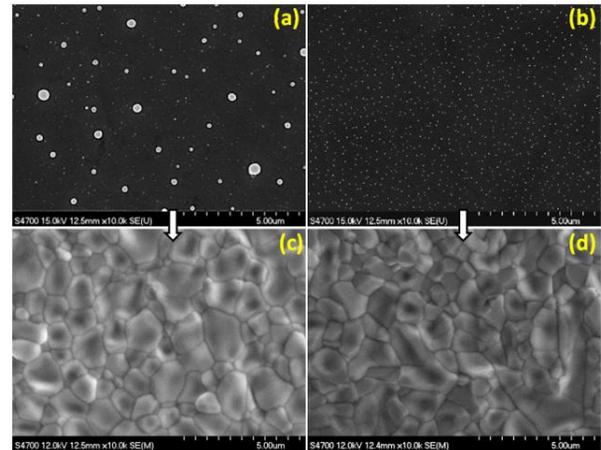


Figure 6: SEM images of nucleated surfaces obtained by using (a) Jlab and (b) Cornell recipes are shown in first row. Note the small tin particles produced by Cornell recipe. Second row shows final coatings obtained by corresponding nucleation profiles. Note that  $(23.5 \pm 2)$  atomic percent tin was observed in both coating.

## SUMMARY AND OUTLOOK

Nucleation parameters were varied to study the nucleation process that is considered to be an important step in Nb<sub>3</sub>Sn diffusion coating. We studied the nucleated produced surfaces with different set of parameters by using different characterization tools. No chlorine was detected in any experiment. XPS and SAM analysis on nucleated samples indicated the presence of tin particle and/or tin film in nucleated samples. Amount of tin chloride is found not to be crucial for tin coverage on niobium surface, but larger amount of tin chloride appears to create tin particle of bigger size. We are in process of investigating the final coatings produced with different set of different nucleation parameters.

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