

# INSIGHTS INTO Nb<sub>3</sub>Sn COATING OF CEBAF CAVITIES FROM WITNESS SAMPLE ANALYSIS

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## Abstract

With the progress made in the Nb<sub>3</sub>Sn coatings on single-cell SRF cavities, development is ongoing to reproduce single-cell cavity results on practical structures such as CEBAF 5-cell cavities. During CEBAF cavity coating development, several changes from the single-cell cavity coating procedure to the coating setup and the heating profile were introduced to improve the quality of Nb<sub>3</sub>Sn films. To witness the properties of grown Nb<sub>3</sub>Sn films in different cavity locations, 10 mm × 10 mm samples were positioned in strategic places within the coating chamber. Composition and structure of the samples were analyzed with surface analytic techniques and correlated with sample location during coatings. Implications from sample analysis to Nb<sub>3</sub>Sn coatings on different geometries are discussed in this contribution.

## INTRODUCTION

Due to the promising results with the vapor-diffused Nb<sub>3</sub>Sn coating, Nb<sub>3</sub>Sn deposition system at JLab has been upgraded to coat CEBAF 5-cell cavities with Nb<sub>3</sub>Sn using vapor diffusion technique [1]. Early coatings of CEBAF 5-cell cavity coatings were observed to have up-down asymmetry. Despite non-uniformity, the quality factors above 3·10<sup>10</sup> at 4.3 K and above 10<sup>11</sup> at 2 K were measured in one of the cavities [1]. In the course of the cavity coatings, witness samples were used to investigate the structure of coated films. Analysis of these samples revealed interesting features of Nb<sub>3</sub>Sn process, which is presented and discussed in this contribution.

## 5-CELL CAVITY COATINGS

In order to investigate coating uniformity, samples were placed inside 5-cell cavities during cavity assembly in the cleanroom for Nb<sub>3</sub>Sn coating. The typical setup consisted of two 10 mm × 10 mm samples coated together with the cavity. One sample was placed on top of the tin crucible, which is attached to the cavity beamline port on FPC side. Another sample was hung at the top cover next to the secondary crucible. In one of the coating setups, 0.25" Nb rod was inserted along cavity vertical axis during cavity assembly for the coating. The rod extended from the top to the bottom crucible. During the coating, the rod witnessed tin supply variation along cavity axis and was coated accordingly. After the coating, the rod was cut into small samples for analysis.

Initially, coatings were done using a single tin source at

the bottom of the cavity. Post coating cavity inspections revealed the asymmetry in the cavity coatings. Cells at the bottom of the cavity, which were close to the tin and tin chloride source during coating, had the typical appearance of a well-coated Nb<sub>3</sub>Sn. The cells close to the top of the cavity showed irregular coating to a varying degree. In Fig. 1, optical images of coated IA110 are shown. Clear difference between the top and the bottom of the cavity is observed. Similar irregular coating was observed on some

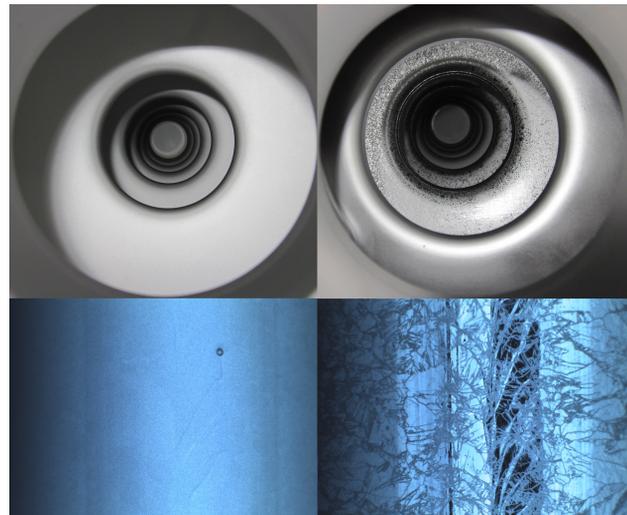


Figure 1: A picture of the inside of IA110 coated with Nb<sub>3</sub>Sn. The left top picture is taken through the beam tube on the fundamental power coupler side, which is at the bottom during coating. The right top picture is taken through the beam tube on the high order mode couplers side, which is at the top during coating. The left bottom picture is taken with KEK inspection system [2] looking at the equator of the cell 1, which is the cell closest to the tin source. The right bottom picture is taken with KEK inspection system looking at the equator of the cell 5, which is the cell furthest from the tin source. Note the difference in coating appearance at both sides.

of the niobium covers used to cover cavity ports. One of the covers was inspected in SEM, Fig. 2. SEM inspection revealed that irregular coating is a mixture of well-coated regions and so-called patchy regions. Patchy regions have been observed to be single crystal Nb<sub>3</sub>Sn grain thinner than the surrounding coating [3]. EDS analysis of these regions in the coated cover indicates lower Sn content and is consistent with a thinner layer in these areas. Similar features

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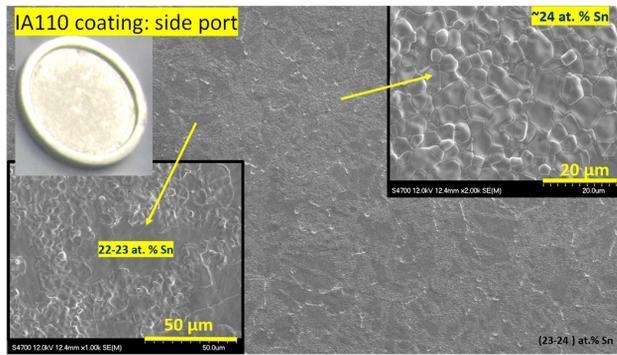


Figure 2: SEM image of the coated surface of IA110 pick-up port cover coated with  $Nb_3Sn$ . The surface appearance is the mixture of well-coated  $Nb_3Sn$  regions and patch regions.

were observed on one of the coated samples in our coating chamber in the past. A sample was supported at the edges inside the coating chamber during coating. The areas where the samples were supported have lower exposure to Sn flux due to lower conductance. These areas were clearly visually different from the rest of the sample, which was exposed to Sn vapors, Fig. 3. The visual non-uniformity was again linked to the higher density of patchy regions with a lower tin content. It is important to note that these visually non-uniform regions contain both regular  $Nb_3Sn$  coating and patchy regions. The inspections of irregular

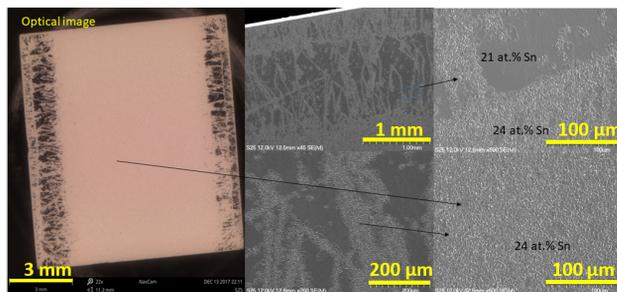


Figure 3: Optical and SEM images of a coated sample, which was supported at the edge. Note that the areas where the sample was supported have a different appearance.

coatings on 5-cell cavity covers and small samples indicated that such irregular coatings are caused by lower Sn and/or  $SnCl_2$  amount available to the coated surface. This finding was further corroborated by the inspection of the sample rod used in 5-cell cavity coating, where a niobium rod, positioned along the axis of a 5-cell cavity, was removed, cut into pieces, and inspected in SEM/EDS, Fig. 4. The bottom beam pipe and the two samples, corresponding to the first two bottom cells during 5-cell cavity coating, had a regular  $Nb_3Sn$  coating without any patchy regions. Starting from the sample, corresponding to the center cell, cell #3, patchy regions were observed along with the regular coating. The number of patchy regions increased progressively from cell #3 to cell #4 to cell #5. The sample, which was inside the top cell during the coating, had the highest density of patchy regions. While there was a clear trend observed in the

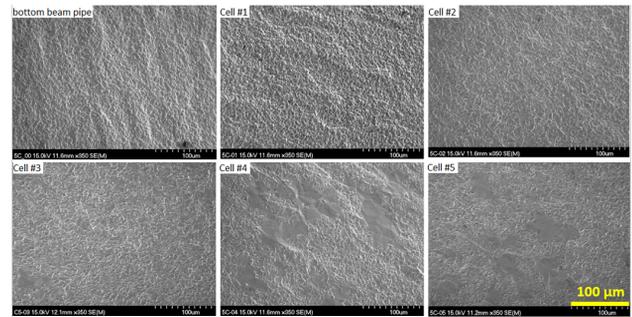


Figure 4: SEM images of a coated sample from niobium rod, placed before the coating inside 5-cell cavity along the cavity axis. Note absence of the patchy regions in the upper row and the presence of the patchy regions in the lower row.

abundance of patchy regions on samples collected from the sample rod, the grains were very similar in the areas, which had the regular  $Nb_3Sn$  coating, Fig. 5. It is worth noting that the distance from Sn and  $SnCl_2$  source did not significantly affect the grain size, and the samples at the source and away from the source exhibited a similar  $Nb_3Sn$  grain size.

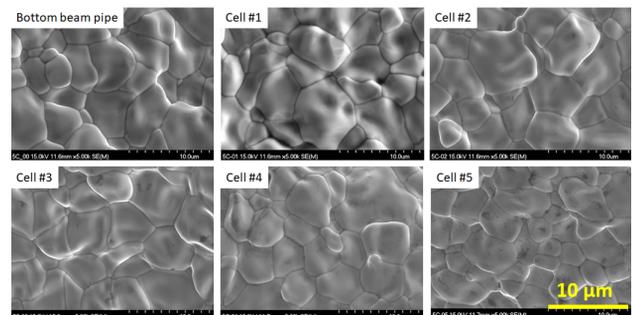


Figure 5: Higher resolution SEM images of coated samples from niobium rod, placed before the coating inside 5-cell cavity along the cavity axis. Note the similar grain size among all samples.

To increase the amount of Sn reaching the surface in the cavity areas away from the tin source, another tin source was suspended from the top cover close to the center cell. The addition of the secondary source improved coating uniformity, which was reported in [4].

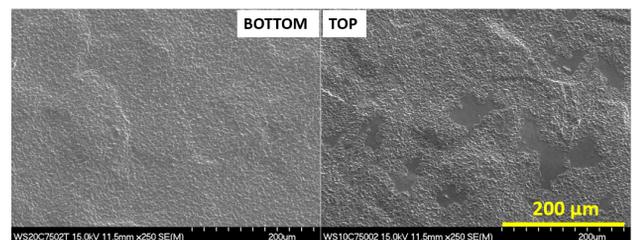


Figure 6: SEM images of witness samples coated with 5-cell cavity 5C75-J-002. Note the absence of patchy regions in the sample from the bottom and a few patchy regions in the sample from the top of the cavity

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Witness samples, which were coated with the 5-cell cavities using two tin sources, were inspected with SEM, Fig. 6. The witness samples at the bottom close to the primary tin source showed a regular  $Nb_3Sn$  coating, expected for the temperature and duration used in the process. The witness sample at the top close to the secondary tin source showed a regular  $Nb_3Sn$  coating, sometimes with a few patchy regions. EDS analysis of the top witness sample showed very simi-

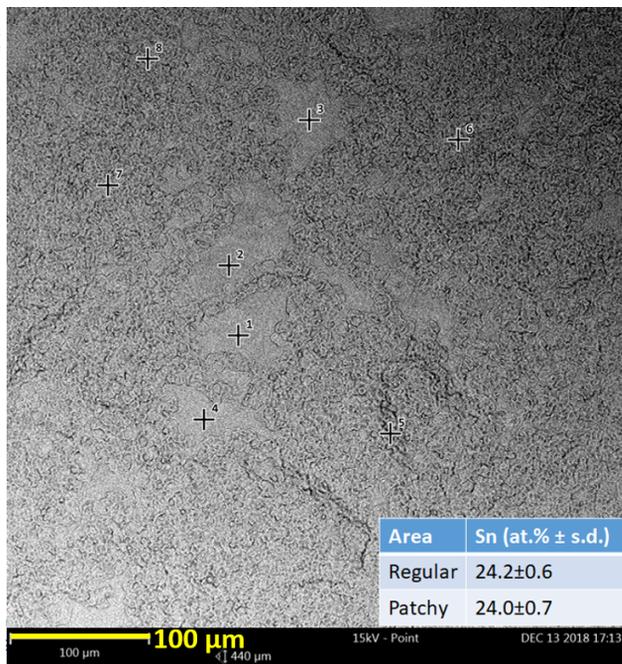


Figure 7: SEM image and EDS analysis of the coated surface of the top witness sample. Note the similar composition between areas with regular coating and patchy regions.

ilar composition between the regular  $Nb_3Sn$  coating and the patchy regions, Fig. 7. Typically, patchy regions are found to contain less tin, which is explained by thinner thickness of the coating in these areas. Similar tin content suggests that patchy regions are about  $1 \mu m$  thick, which is the information depth for 15 keV voltage used in EDS measurements. Longer coating process time of 24 hours, which was applied in these 5-cell cavity coatings, is likely the cause for the thicker coating in the patchy regions.

Typically, a mesh or a molybdenum diffuser is used to cover bottom tin crucible. Molybdenum diffuser was made out of 99.95% molybdenum (tungsten  $\leq 300$  ppm; other impurities  $\leq 50$  ppm). It was speculated that such cover will improve homogeneity of tin flux leaving molten tin pool. Molybdenum diffuser was inspected in SEM after coating, Fig. 8. No coating or irregular structures were seen on the surface. EDS analysis did not show any tin or chlorine. Molybdenum diffuser has routinely been used in our coatings, including some of the best coatings [5]. Microscopic analysis and cavity results suggest that molybdenum is benign to the coating process and can be used as a part of reaction chamber. Another benefit of a mesh or a molybdenum diffuser was observed, when attempts were made

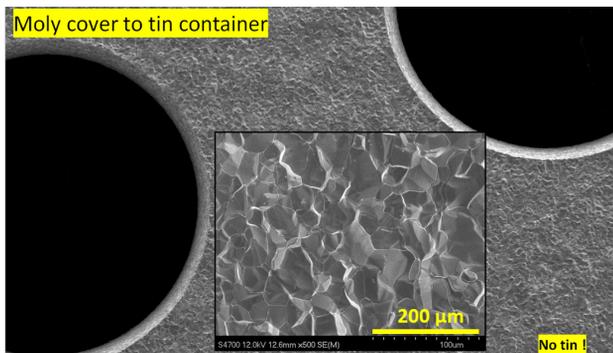


Figure 8: SEM image and EDS analysis of molybdenum diffuser. Note the absence of tin or chlorine on the surface.

to coat cavities without them. In an attempt to further increase tin flux, bottom tin crucible was left open to the cavity interior without a mesh or a diffuser in several coatings. Inspections of the coated surfaces revealed uniformly coated  $Nb_3Sn$ , but also small spots on the surfaces close to the tin source. These spots were speculated to be tin splatter. The mechanism responsible for formation of these spots is not clear, and in a few cases such spots were observed inside the cells away from the tin source.

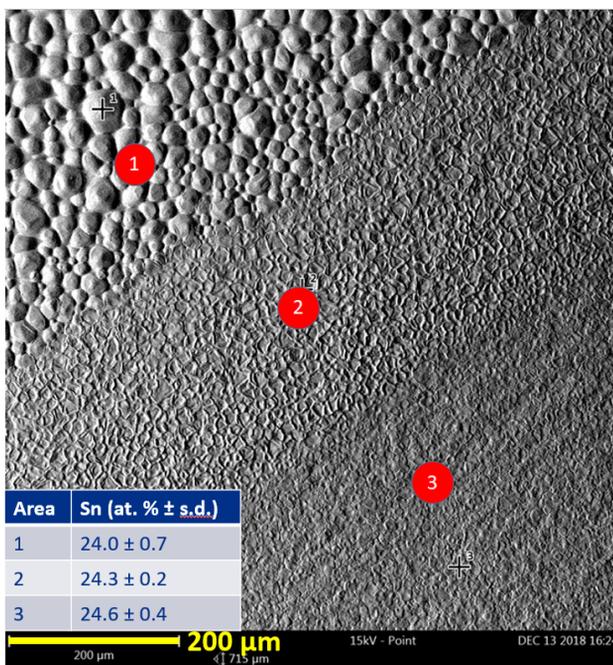


Figure 9: SEM image of the coated  $Nb_3Sn$  surface close to the edge of a spot. Note the three distinct regions and the sharp boundaries between them.

Similar spots were sometimes observed on the witness samples and niobium foils, used to contain tin shots. In Fig. 9, an SEM image of the area close to the edge of one of the spots is shown. Three distinct regions with a sharp boundary between them can be identified in SEM image. Area #1 is the area where molten tin was located. Area #2 is the transition region, and Area #3 has the appearance of

the regular vapor diffused Nb<sub>3</sub>Sn. In Fig. 10, SEM images of two different resolution are shown from each of these regions. The region, where molten tin was available, has large spherical Nb<sub>3</sub>Sn grains with gaps between the grains. The transition regions has the appearance somewhat similar to vapor diffused Nb<sub>3</sub>Sn, but with brighter grains and grain edges, which could indicate them to be sharp and elevated near the grain boundaries. The third area has the regular appearance of vapor diffused Nb<sub>3</sub>Sn. An SEM image of area #4

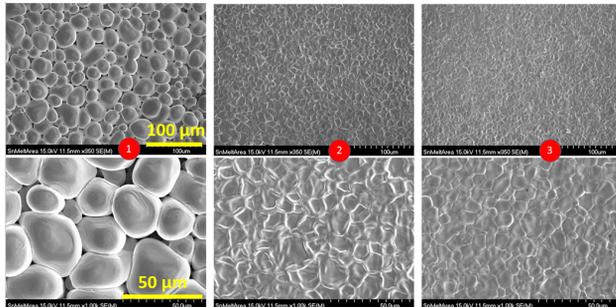


Figure 10: SEM images from the different areas in Fig. 9. Note the drastic difference between Nb<sub>3</sub>Sn coating, where molten tin was present, and the regular vapor diffused Nb<sub>3</sub>Sn coating.

is shown in Fig. 11. The substrate in this case was niobium foil, which was used to contain tin shots before the coating. This area is believed to have contained molten tin during the coating process. Areas #1 and #2 were observed close to the area #4, which appears distinct from the other areas. The coating is irregular with several islands containing small Nb<sub>3</sub>Sn grain and the rest of the film have a continuous coating appearance. Sharp boundaries were observed between all these areas, which is likely pointing to different growth mechanisms behind formation of each area. Even when a

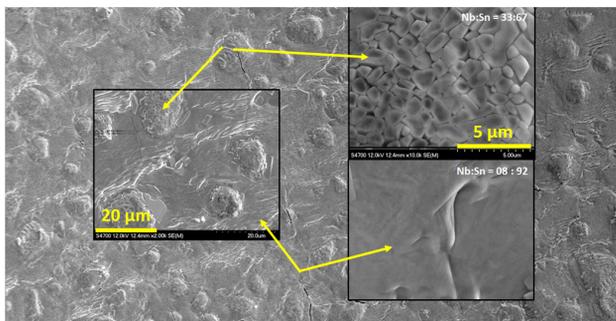


Figure 11: SEM image of the regions, where molten tin was likely available during the process.

mesh or a molybdenum diffuser were used, tin nanoresidue was found after coating in several samples, when two tin sources were used, Fig. 12. Similar tin nanoresidue was found in some of the single cell cavity coatings [5]. These particles are believed to form during cooldown phase of Nb<sub>3</sub>Sn coating process from the residual tin vapor inside the reaction chamber. Several post treatments on the coated samples were found to reduce or eliminate tin nanoresidue,

but the experiments are ongoing to check the efficacy of such treatment in coated SRF cavities [5].

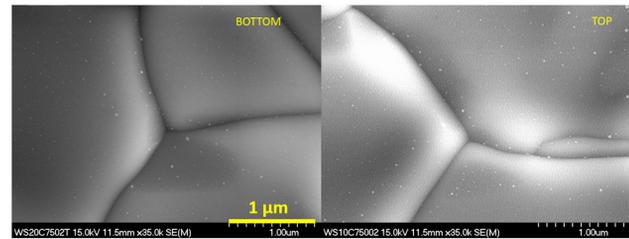


Figure 12: High resolution SEM image of coated witness samples. Note that tin nanoresidue is present in both samples.

## SUMMARY

Analysis of the samples coated together with 5-cell cavities shows that development of patchy regions is linked to the low tin supply. Under such conditions Nb<sub>3</sub>Sn layer grows as a mixture of the regular Nb<sub>3</sub>Sn areas and patchy regions. The density and amount of patchy regions increases with the reduction of tin supply.

Excess of tin on the other hand could result in “tin” spots, sometimes, significant distance away from the tin source. Such spots, which grow in the presence of molten tin, have loosely-connected spherical grains significantly larger than vapor diffused Nb<sub>3</sub>Sn grown under the same temperature profile. Several distinct coated regions are observed next to the spots, where molten tin believed to be present, with sharp boundaries between them suggesting different growth mechanisms behind different areas.

99.95% molybdenum hardware has been used inside the reaction chamber. Molybdenum does not appear to affect the coating quality.

## ACKNOWLEDGMENTS

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