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LOWER TEMPERATURE ANNEALING OF VAPOR DIFFUSED Nb_3Sn FOR SRF CAVITIES*

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Abstract

Nb_3Sn is a next-generation superconducting material for the accelerator cavities with higher critical temperature and superheating field, both twice compared to Nb. It promises superior performance and higher operating temperature than Nb, resulting in significant cost reduction. So far, the Sn vapor diffusion method is the most preferred and successful technique to coat niobium cavities with Nb3Sn. Although several post-coating techniques (chemical, electrochemical, mechanical) have been explored to improve the surface quality of the coated surface, an effective process has yet to be found. Since there are only a few studies on the post-coating heat treatment at lower temperatures, we annealed Nb_3Sn-coated samples at 800 ℃ - 1000 ℃ to study the effect of heat treatments on surface properties, primarily aimed at removing surface Sn residues. This paper discusses the systematic surface analysis of coated samples after annealing at temperatures between 850 ℃ and 950 ℃.

INTRODUCTION

Nb_3Sn is a potential alternative material to replace Nb in SRF cavities with a higher critical temperature T_c of 18.3 K, higher critical field, accordingly the lower surface resistance compared to Nb. Thus, Nb_3Sn could potentially give a lower dissipation than that of niobium at any given temperature. Nb_3Sn cavities can operate at 4 K delivering similar performance that of Nb cavities at 2 K promising significant reduction in the operational cost of SRF accelerators. Its superheating field of about 400 mT suggests a higher breakdown field for higher accelerating gradient, which can reduce initial cost to build SRF accelerators [1]. Because of brittleness and lower thermal conductivity, Nb_3Sn should be deposited as a thin film on RF structures. Among the available methods, the Sn vapor diffusion technique is successful technique pursued by several laboratories to coat Nb SRF cavities with different shapes and frequencies [2–6].

A Nb twin-axis cavity with complex geometrical shape was coated with vapor-diffused Nb_3Sn several times at JLab but resulting performance was not as good as expected for Nb_3Sn [1]. Further investigations pointed out that the poor performance was caused by mechanical deformation of the cavity during the cool down to cryogenic temperatures. Other factors that could have contributed to the performance degradations were potential non-uniformity of the coating at hard-to-reach areas, and surface quality of the thin-film. Later coating experiments with samples positioned at strategic positions revealed uniform and complete coating inside the cavity, but Sn residue were present in most of the cavity witness samples. Note that the presence of Sn-residues on the coated surfaces had been reported before [7]. Most of the post-coating treatment explored in the past to improve surface quality resulted in surface degradation [8] or were inconclusive.

We updated the coating parameters to minimize the accumulation of Sn-residues. It included Sn-supply optimization by reducing the amount of Sn, the size of the crucible, and temperature profile modifications (discussed later). Although the updated coating parameters with multi-step coating temperatures and gradual cool down below 900 ℃ reduced residue sizes, complete elimination was challenging.

One potential post-coating treatment to remove Sn-residue could have been annealing the coated sample without Sn. Previous studies at JLab have shown that annealing the coating at >1000 ℃ resulted in residue eliminations along with Sn-loss from the Nb_3Sn layer, and unusual grain faceting that resulted in performance degradation [9]. Temperature below 1000 ℃ was not explored, which could have avoided the Sn loss from the coated layer and mitigated the effect of Sn residues, which may help to improve the RF performance of Nb_3Sn-coated cavities. In this contribution, we report surface studies of Nb_3Sn samples which received post-coating annealing at 850-950 ℃.

EXPERIMENTAL

Sample Preparation and Nb_3Sn Coating

Samples used in this study were 10 mm × 10 mm coupons cut from high RRR, 3mm thick Nb sheet, similar to one used to fabricate SRF cavities. These samples received 100 µm of Buffered Chemical Polishing (BCP) to remove damaged layer from the surface. They were then heat treated at 800 ℃ for 3 hours for hydrogen degassing. Finally, these samples were subjected to 5 µm BCP removal before Nb_3Sn coating.

Typically, the samples for this study were coated using similar coating process used to coat single-cell cavities recently at JLab. The temperature profile included 1 hour of nucleation at 500 ℃ followed by about 3 h of coating (~ 40

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min at 1200 °C, 45 min at 1150 °C, and 85 min 1100 °C). Following the coating, the temperature was ramped down at 1 °C/min to 800 °C before the heater turned off, similar to the twin axis cavity coating. Samples were coated inside a Nb chamber using 0.9 g of Sn loaded into 0.5" diameter crucible and 0.5 g of $SnCl_2$ packaged inside Nb foil [1, 8]. Each sample was coated uniformly with expected Sn residues similar to twin axis cavity coating.

Annealing Experiments

First, we annealed samples at (950 ± 5) °C for 45 minutes to get an idea of suitable temperature profiles for annealing studies. As shown in the Fig. 1, it developed an unusual facet in Nb_3Sn grains which was similar in samples annealed at higher temperatures. EDS analysis of the samples showed no Sn loss like we observed during the high temperature annealing at 1100 °C, reported before [8] indicating significantly lower or no Sn loss at 950 °C. Based on these results, we explored annealing between 850 °C and 950 °C for shorter time periods.

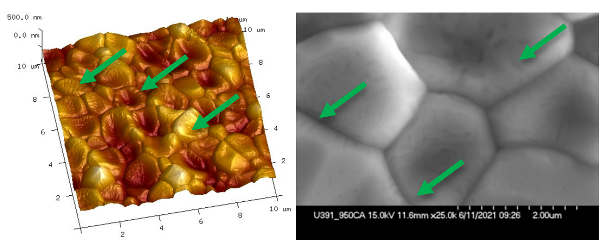

Figure 1: AFM (left) and SEM (right) images from samples annealed at 950 °C for 45 minutes. Note faceting features developed on the coated surface after the annealing as marked with the arrows.

The coated samples were annealed at (850 ± 5) °C, (900 ± 5) °C and (950 ± 5) °C for 10 min and 20 min. The samples were then analysed with several characterization techniques to assess study post-annealing effect on composition, microstructure, topography, and surface features. The analysis was also focused on size and distribution of Sn residue. We will discuss results only from 20 min annealed samples here.

MATERIAL STUDIES

The coated and annealed samples were examined using Atomic Force Microscopy (AFM), Scanning Electron Microscopy (SEM) and X-Ray Diffraction (XRD).

AFM

The topography of each sample was analyzed out with a Digital Instruments Nanoscope IV AFM in tapping mode. Samples were scanned at 3 randomly chosen locations with 5 µm × 5 µm, 10 µm × 10 µm, 20 µm × 20 µm, and 50 µm × 50 µm scan sizes.

The representative AFM images from each experiment are shown in Fig. 2. As expected, a uniform distribution of Sn residue was observed on coated sample as shown in the top left figure. Similarly, the topography of annealed samples is shown below. Note that there are no annealing features like we observed in Fig. 1. Sn residues with different sizes were present varying from a few nm to several tens of nm. The heights of many Sn residues were measured from each sample. The average height of these residues is tabulated below (see Table 1). The density and the height of Sn residue appear to be reduced after the annealing, more with increasing temperature of annealing.

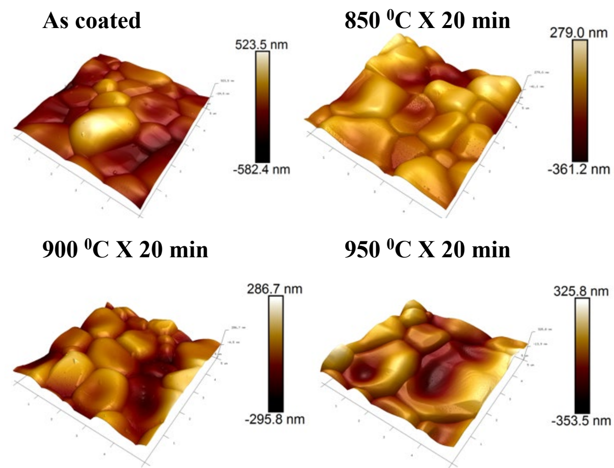

Figure 2: AFM images of the as coated and annealed samples for 20 minutes at different temperatures.

Table 1: Average Height of the Sn Residue

	As coated	**850 °C ×20 min**	**900 °C ×20 min**	**950 °C ×20 min**
Height (nm)	10.8±3.1	5.1±2.4	4.6±2.4	2.8±1.8

The surface roughness measured from each sample is shown in Table 2. The RMS roughness of analyzed as–coated and annealed samples were very similar for 50 µm × 50 µm scans. Sample annealed at 950 °C appears to be rougher than those annealed at lower temperatures in 10 µm × 10 µm and 20 µm × 20 µm AFM scans. Discrepancy of roughness variation in different scan sizes may have come from the inclusion of Nb grain boundaries.

Table 2: RMS roughness of the coated and annealed samples for 5 µm, 10 µm, 20 µm and 50 µm scan sizes. Note that about 10% error is expected for each estimation.

Scan size (µm× µm)	**RMS Roughness (nm)**			
	As coated	**850 °C ×20min**	**900 °C ×20min**	**950 °C ×20min**
10 × 10	64.14	67.2	67.3	114.2
20 × 20	92.1	89.2	80.4	118.4
50 × 50	122.3	144.5	130.8	134.9

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SEM

The elemental composition of each sample was analyzed by energy dispersive x-ray spectrometry (EDS) with Hitachi 4700 field emission scanning electron microscope (FE-SEM) or Phenom ProX. The Sn content in the coating was not changed after the annealing for any annealing profile studied (see Table 3). SEM was used to examine the microstructure of as-coated and annealed samples. SEM images were used to estimate the grain size and analyze Sn particles on the surface. The representative SEM images from each sample is shown in Fig. 3. Note that nanoscopic Sn residue is not visible in this magnification. The estimated average grain size for each sample is presented in the Table 3. The grain size variation was within the error bar.

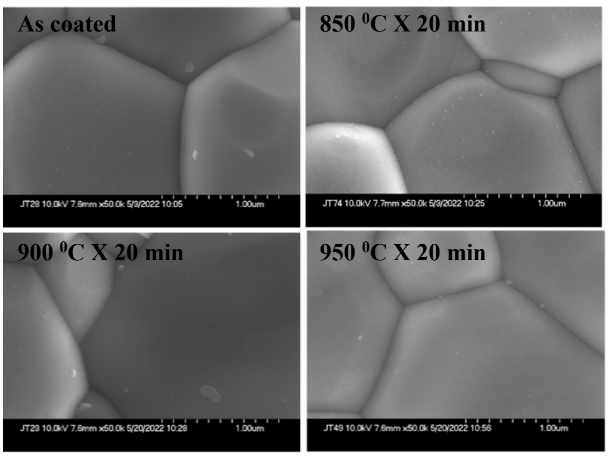

Figure 3: SEM images of the as coated and annealed samples for 20 minutes at different temperatures.

Table 3: Average grain size and the Sn composition of the samples SEM Images of the as-coated and annealed samples for 20 minutes at different temperatures.

	Average grain size (µm)	Average Sn %
As coated	1.23±0.01	24.90±0.56
850 °C × 20 min	1.26±0.05	24.93±0.22
900 °C× 20 min	1.19±0.07	25.07±0.28
950 °C× 20 min	1.21±0.10	24.98±0.11

XRD

X-Ray Diffraction (XRD) pattern of the coating before and after annealing were obtained from Rigaku Miniflex II X-ray diffractometer with Cu-α radiation. As expected only Nb3Sn peaks were observed from as coated sample (see Fig. 4). Some new peaks were evolved on each annealed samples, mostly corresponding to Nb. Some of the peaks observed only in 850 °C and 900 °C annealed samples were hard to discern between Nb or Nb-Sn (2θ at 70°) to other than Nb_3Sn. Note the strong Nb peak revealed annealing at 950 °C. Considering the annealing features in Fig. 1, The XRD peak may indicate the Sn loss from the shallow surface similar to annealed samples at higher temperatures. The data may suggest Sn loss with higher temperature or longer annealing. Similar analysis is in progress for samples annealed for 10 minutes at each temperature studied which may provide more insights on the Sn loss and optimized annealing profile.

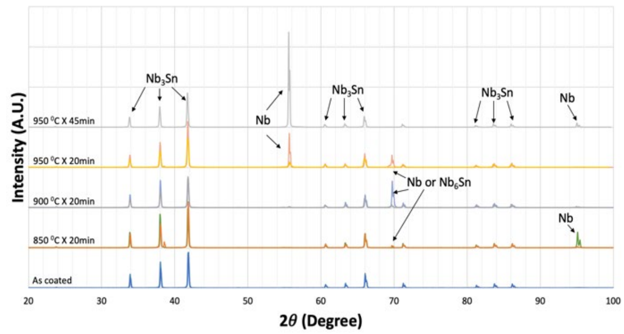

Figure 4: XRD peaks of the samples.

SUMMARY AND OUTLOOK

Sn residue is the limiting factor in the performance of the Nb_3Sn cavities. Sample study is conducted to see how the low temperature annealing affects the surface Sn residue and microstructure in the Nb_3Sn coatings as higher temperature annealing removes Sn from the Nb_3Sn coating and degrade the quality of the film. We annealed Samples at 850 °C, 900 °C and 950 °C for 20 minutes and analysed the samples. The height of the Sn residues measured seems to decrease with the annealing temperature, but we could not see any trend in the grain size, Sn composition or the surface roughness. For the samples annealed at 950 °C there were Nb peaks appeared in the XRD, and further analysis is needed to confirm more details. Since we suspected Sn loss from the surface with 950 °C annealing, we are investigating short annealing at each temperature. We have coated a single cell cavity with coating process described in this paper to anneal it with optimized temperature profile to study the effect of annealing on RF performance.

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