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ELECTROPLATING OF Sn FILM ON Nb SUBSTRATE FOR GENERATING Nb_3Sn THIN FILMS AND POST LASER ANNEALING*

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Abstract

Controlling film quality of Nb_3Sn is critical to its SRF cavity performance. The state-of-the-art vapor diffusion approach for Nb_3Sn deposition observed surface roughness, thin grain regions, and misfit dislocations which negatively affect the RF performance. The Sn deficiency and non-uniformity at the nucleation stage of vapor deposition is believed to be the fundamental reason to cause these roughness and defects issues. Thus, we propose to pre-deposit a uniform Sn film on the Nb substrate, which is able to provide sufficient Sn source during the following heat treatment for Nb_3Sn nucleation and growth. Here, we demonstrated successful electrodeposition of a low-roughness, dendrite-free, excellent-adhesion Sn film on the Nb substrate. More importantly, we further achieved a uniform, low-roughness (Ra = 66 nm), pure-stoichiometric Nb_3Sn film through thermal treatment of this electroplated Sn film in the furnace. Additionally, we provide preliminary results of laser annealing as a post treatment for epitaxial grain growth and roughness reduction.

INTRODUCTION

Nb_3Sn thin film is an important candidate for next-generation superconducting radio frequency (SRF) cavities due to its high superheating field together with high critical temperature [1,2]. However, the RF performance of current Nb_3Sn cavities obtained from a Sn vapor diffusion process [1,3] is greatly affected by surface roughness, grain boundaries, and crystal defects. Some of these issues are believed to arise from the low Sn flux (in vapour deposition) and the resulting Sn-deficiency at the early nucleation stage [4-6]. Recent results from Prof. Richard Hennig's group [7] demonstrate that sufficient Sn source can guarantee the stoichiometry of Nb_3Sn and overcome the Sn deficiency issue. Thus, we intend to pre-deposit a uniform Sn thin film on the Nb substrate which can be converted to Nb_3Sn films through heat treatment. A high-quality Sn film ensures (1) sufficient Sn source for Nb_3Sn nucleation and (2) no grain island growth.

Electroplating is a low-cost, fast, manufacturable, scalable, and possibly selective deposition approach. Electrodeposition of Sn film for generating Nb_3Sn thin films has been explored by using a Cu seed layer on Nb substrate [8]. However, possible Cu contamination would greatly reduce the critical current density of Nb_3Sn [9]. Without the Cu seed, direct Sn electrodeposition on a Nb substrate is challenging due to the frequently observed dendrite formation and peel-off issues. Hence, our motivation is to develop a direct electroplating process to deposit high-quality Sn films on the Nb substrate. Successful electrodeposition requires optimizing a variety of criteria: obtaining uniform films; reducing surface roughness; avoiding dendrite formation; and achieving good adhesion, which are demonstrated in this work.

Following the Sn electrodeposition, heat treatment is essential to facilitate the alloying of Sn film and Nb substrate. It requires a uniform conversion and generates a smooth Nb_3Sn film surface which can thereby demonstrate the theory of using a pre-deposited Sn film to encourage evenly distributed Nb_3Sn nucleation. In this work, we demonstrate the generation of smooth, perfect-stoichiometry Nb_3Sn films via furnace heating; and also propose a laser annealing technique to enable epitaxial growth of Nb_3Sn grains.

EXPERIMENTAL PROCEDURES

As illustrated in Fig. 1a, Sn electroplating was carried out using a standard 3-electrode setup with Pt counter electrode and saturated calomel reference (SCE) electrode. The starting Nb substrate was mechanically polished with a final surface roughness of Ra = ~70 nm as measured by Zygo optical profilometer (Fig. 1b). These electroplated Sn samples were then placed in a vacuum furnace at >1000 °C for 3 hours to generate Nb_3Sn films [2]. Laser annealing was studied using a 120 W CO_2 laser (λ = 10.6 μm) and another 980 nm wavelength diode laser.

After electroplating and heat-treatments, surface morphology, film uniformity, and dendrite formation were evaluated by scanning electron microscope (SEM, Zeiss Gemini 500). Surface roughness was measured and quantified using atomic force microscopy (AFM). Moreover, electron dispersive x-ray spectroscopy (EDS) and x-ray photoelectron spectroscopy (XPS, PHI Versaprobe III) were carried out to determine the elemental information

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and stoichiometry. X-ray diffraction was measured using a Rigaku SmartLab X-ray diffractometer (XRD) to determine the phase information and grain orientation. Lastly, tape tests were performed to confirm the strength of adhesion for the electroplated Sn film on the Nb substrate.

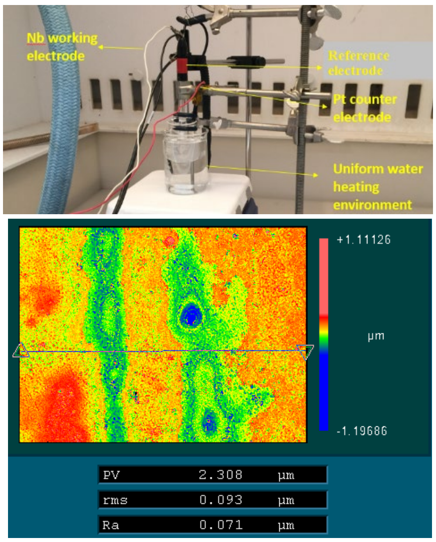
(a)
(b)


Figure 1: (a) Image of the electroplating setup. (b) Surface roughness of the starting Nb substrate.

RESULTS AND DISCUSSION

Electroplated Sn Film on Nb Substrate

The SEM image in Fig. 2a shows a high-quality Sn film on the Nb substrate from our optimized electroplating process. The film is uniform and its roughness is low (Ra = 41 nm and Rq = 10 nm). Also, we did not observe any dendrites on the film although some embedded "island" crystal appeared. Ensuring uniformity and surface smoothness of pre-deposited Sn films before heat treatment is critical to achieving evenly distributed nucleation events and thus a small roughness on the resulting Nb_3Sn film. Cross-section SEM images (Fig. 2b) indicate that the Sn thickness is around 4 µm after 10 min deposition. This thickness can be well controlled through monitoring the deposition time. The cross-section also suggests a layer-growth mode of the Sn film during electroplating.

Another important factor is the quality of adhesion for Sn films on the Nb surface. Peel-off issues were frequently observed, which made the electroplated Sn film to fail any cleaning or handling procedures. A tape test was performed on our electroplated Sn film, and results (Fig. 3) demonstrate excellent adhesion between the electroplated Sn film and Nb substrate.

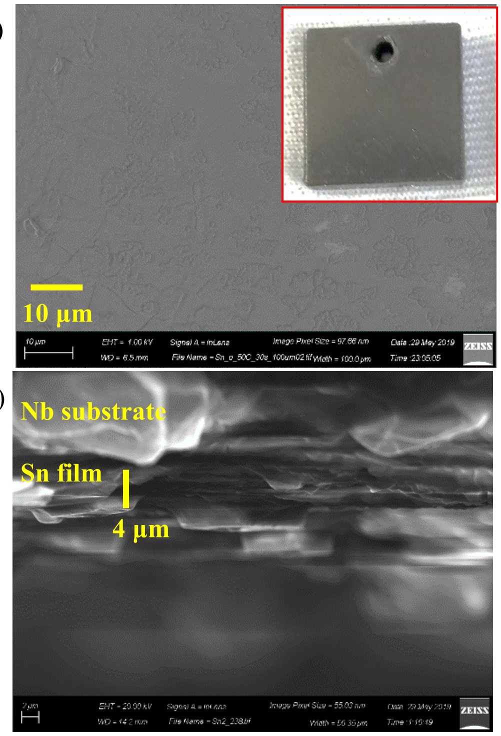
(a)
(b)


Figure 2: (a) Surface SEM image showing the uniform Sn film from electrodeposition. Insert is a picture of this electroplated Sn film on a 1 cm × 1 cm Nb substrate. (b) Cross-section SEM image indicating a film thickness of ~ 4 µm for 10 min electrodeposition.

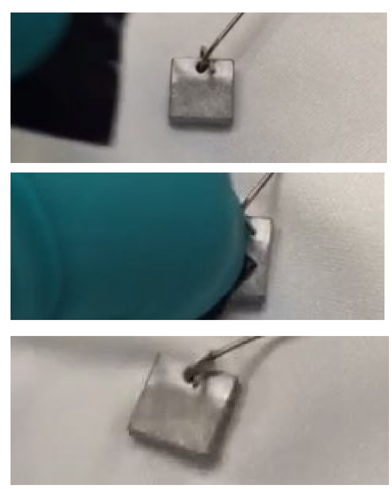


Figure 3: Snapshots presenting the electroplated Sn film before, during, and after tape test are shown from top to bottom.

Elemental determination and impurity analysis were carried out using EDS and XPS. As shown in Fig. 4a and 4b, both spectra confirm the predominant content of Sn element in the electroplated film. EDS spectrum exhibits the presence of C, O, and N impurities; however, the quantified values of these light elements using EDS are not accurate. XPS spectrum suggests low amount of C and N while

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some oxide is expected on the film surface where XPS primarily measured. Further accurate quantification of impurities will be performed in the future work.

Moreover, XRD results show that our electroplated Sn films have a body-centered tetragonal β-Sn structure and the growth is preferred in the [100] orientation.

(a)

(b)

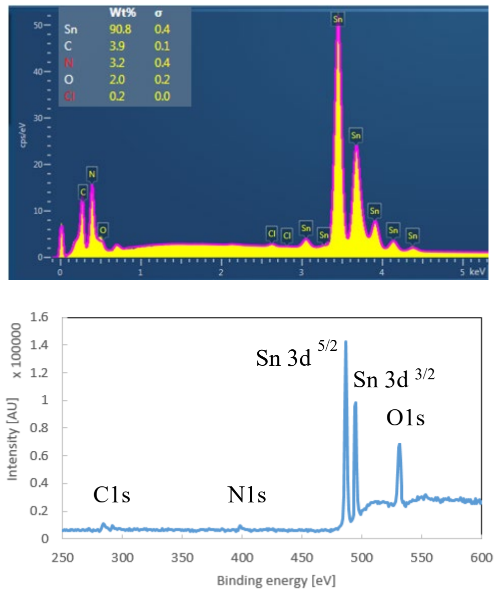

Figure 4: (a) EDS spectrum from a random point on the electroplated Sn film. (b) XPS spectrum confirming a low amount of impurities.

Nb_3Sn Generation from Electroplated Sn Films

After furnace heating electroplated Sn films at >1000 °C with the Nb substrate, we observed the generation of a uniform Nb_3Sn film as shown in Fig. 5a. The grain size is around 500 nm. The stoichiometry as determined by EDS is perfectly 3:1 (Fig. 5b).

Indeed, the surface roughness of the obtained Nb_3Sn film is comparably low (Ra = 66 nm, Rq = 9 nm) as the electroplated Sn film or the starting Nb substrate. This observation demonstrates the theory that sufficient and uniform Sn source during Nb_3Sn alloying can significantly reduce the surface roughness of the resulting film.

Preliminary Results on Laser Annealing

Laser annealing technique has been reported to epitaxially grow surface layer or thin films [10,11]. We intend to enable epitaxial growth of Nb_3Sn grains or achieve Nb_3Sn conversion from Sn films. Preliminary results showed that Nb_3Sn surface failed to absorb sufficient laser energy using a 120 W, 1064 nm wavelength CO_2 laser and a 250 W, 980 nm wavelength diode laser. The reflection spectra (Fig. 6) exhibited a below 5% absorption at these wavelengths. Continued efforts will be made in the future work.

(a)

(b)

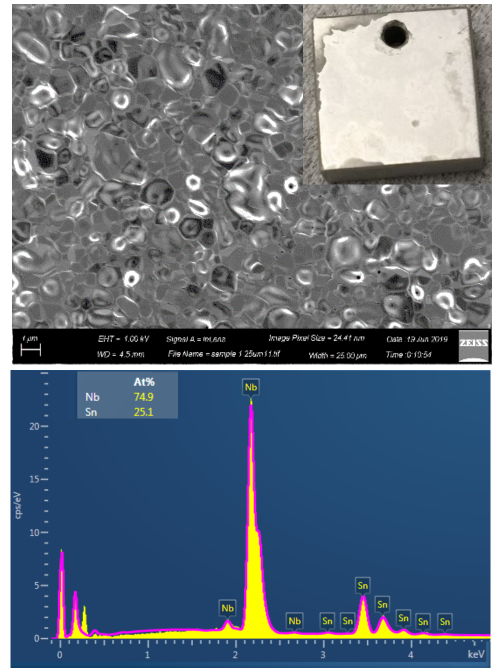

Figure 5: (a) SEM image showing a uniform converted Nb_3Sn film. Insert is a picture of this Nb_3Sn film converted from the electroplated Sn film. (b) EDS spectrum confirming perfect stoichiometry of Nb_3Sn.

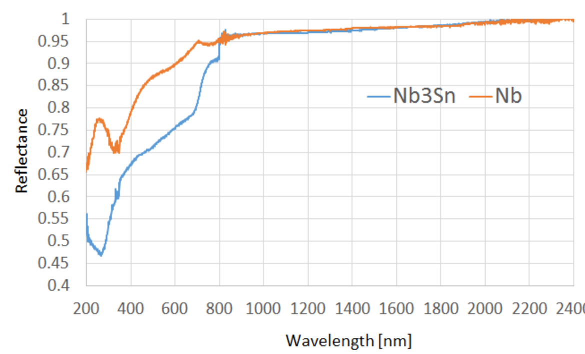

Figure 6: Reflection spectra of Nb and Nb_3Sn.

CONCLUSIONS

In conclusion, an electroplating process was successfully developed to deposit high-quality Sn films on Nb substrate. The electroplated Sn film shows excellent uniformity, very low surface roughness (Ra = 41 nm), no dendrite formation, and great adhesion to the Nb substrate. This high-quality electroplated Sn film is demonstrated to provide sufficient Sn source during Nb_3Sn nucleation and growth; as a result, we effectively converted the electroplated Sn film with the Nb substrate to a uniform, low-roughness (Ra = 66 nm), pure-stoichiometric Nb_3Sn film. Laser annealing can be a promising approach for epitaxial growth of Nb_3Sn film, and further investigation will be carried out in the future work.

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