ISBN: 978-3-95450-184-7 doi:10.18429/JACoW-IPAC2018-THPAL129

MAGNETRON SPUTTERING OF Nb_3Sn FOR SRF CAVITIES

Md. N. Sayeed[1,2], U. Pudasaini[3], H. E. Elsayed-Ali[1,2], M. J. Kelley[3,4,5], C. E. Reece[5], G. Eremeev[5]
[1]Department of Electrical and Computer Engineering, Old Dominion University, Norfolk, USA
[2]The Applied Research Center, Old Dominion University, Newport News, USA
[3] Applied Science Department, The College of William and Mary, Williamsburg, USA
[4]Virginia Polytechnic Institute and State University, Blacksburg, USA
[5]Thomas Jefferson National Accelerator Facility, Newport News, USA

Abstract

Nb_3Sn is a potential candidate for surface material of SRF cavities since it can enable the cavity to operate at higher temperatures with high quality factor and at an increased accelerating gradient. Nb-Sn films were deposited using magnetron sputtering of individual Nb and Sn targets onto Nb and sapphire substrates. The as-deposited films were annealed at 1200 °C for 3 hours. The films were characterized for their structure by X-ray Diffraction (XRD), morphology by Field Emission Scanning Electron Microscopy (FESEM), and composition by Energy Dispersive X-ray Spectroscopy (EDS) and Time of Flight Secondary Ion Mass Spectrometry (ToF-SIMS). The surface resistivity was measured down to cryogenic temperature to determine the superconducting transition temperature and its width. The composition of the multilayered films was controlled by varying the thickness of the Nb and Sn layers. The films showed crystalline Nb_3Sn phases with T_c up to 17.6 K.

INTRODUCTION

Nb_3Sn is an intermetallic compound which is considered as a replacement for Nb SRF cavities due to its high transition temperature compared to Nb [1]. However, due to the fragile nature and poor thermal conductivity of Nb_3Sn, it cannot be used as sheet material; instead Nb_3Sn films can be coated on Nb cavities to get better superconducting surface.

Since its initial synthesis in 1954 [2], several techniques have been used to deposit Nb_3Sn layers. The most successful and widely used coating technique is tin vapor diffusion technique which was developed at Siemens AG in the 70s [3] and utilized to coat superconducting Nb_3Sn RF cavities at Wuppertal University during 80s and 90s [4]. The technique is used by several research groups now to coat Nb_3Sn on the surface of RF cavities [5-7].

Besides the tin vapor diffusion process, another promising technique to deposit Nb_3Sn on Nb cavity is magnetron sputtering. The main advantage of magnetron sputtering over tin diffusion process is the control over the stoichiometry. By changing the thickness of Nb and Sn, different stoichiometry can be achieved which is hard to achieve in the tin diffusion process. Magnetron sputtering can be used in three ways to deposit Nb_3Sn film: sputtering from a stoichiometric Nb_3Sn target, sputtering from two different targets of Nb and Sn to make multilayers followed by annealing to produce the Nb_3Sn phase or co-sputtering of Nb and Sn on a heated substrate. All techniques have been utilized successfully to get the superconducting phase and research is going on to apply magnetron sputtering technique to coat large cavities [8-14].

We applied multilayer magnetron sputtering technique on Nb and sapphire substrates to study the coated film.

EXPERIMENTAL PROCEDURE

Multilayers of Nb and Sn were deposited using an ATC Orion-5 magnetron sputtering system (AJA International, Inc., USA) on Nb and sapphire substrates. A 2 inch Nb target has been used for DC sputtering of Nb films whereas a 2 inch Sn target is used as RF sputtering source for Sn. Previous study suggested different ranges of Nb-to-Sn thickness ratios to get stoichiometric Nb_3Sn [10-12]. We applied two different thickness ratios of 2 and 4.5 for our samples. The chamber was evacuated to 10^{-7} Torr before deposition. The deposition was performed at 3 mTorr with an argon flow rate of 20 SCCM. The deposition was done at room temperature.

Initially a thin Nb buffer layer was deposited on the substrate to avoid Sn diffusion through the sapphire substrate. The film deposition started with depositing Sn layer on the Nb buffer layer followed by Nb layer. Several layers were deposited to get a multilayered film. The outer layer of the film was Nb to avoid Sn evaporation during the annealing process. To obtain uniformly coated film, the substrate holder was rotated at 50 rpm throughout the deposition. The as-deposited films were annealed at 1200 °C for 3 hours in a different furnace. Due to incomplete diffusion of Sn into Nb, as found from the depth profile of the films, we reduced the individual layer thickness. The sample parameters are shown in Table 1.

The crystal structures of the films were studied by XRD using a CuKα radiation. To observe the crystal phase formation, XRD was performed on coated films before and after annealing. The films were examined by FESEM with EDS. ToF-SIMS depth profile of the films were performed to observe Sn diffusion through Nb. Surface resistivity of the films were performed at low temperature to observe the transition temperature T_c of the films.

* Authored by Jefferson Science Associates, LLC under U.S. DOE Contract No. DE-AC05-06OR23177. The U.S. Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce this manuscript for U.S. Government purposes.

ISBN: 978-3-95450-184-7 doi:10.18429/JACoW-IPAC2018-THPAL129

Table 1: Coating Description of Deposited Films

Sample Name	Nb Isolating Layer Thickness (nm)	Nb Layer Thickness (nm)	Sn Layer Thickness (nm)	No. of Each Layers	Overall Thickness (nm)
S1, G8	20	202.5	45	12	2990
S3, G2	20	607.5	135	4	2990
S39, N9, N12	20	50	25	10	770
S41, N31, N40	20	50	25	15	1145

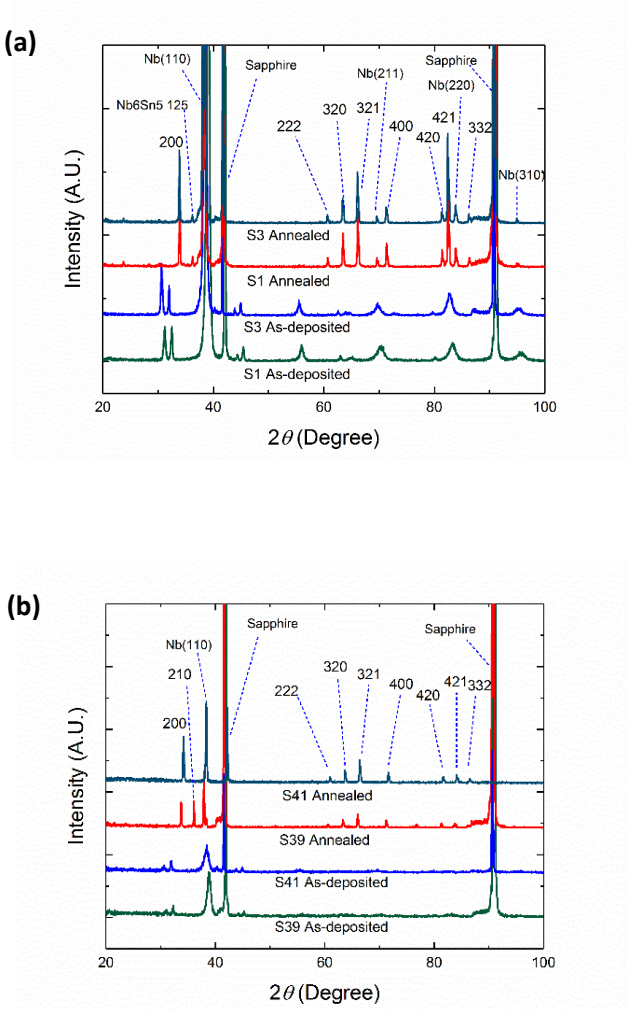

Figure 1: XRD patterns of the coated films with (a) thicker Nb-to-Sn layers and (b) thinner Nb-to-Sn layers.

RESULTS AND DISCUSSION

The XRD patterns of the films coated on sapphire substrates are shown in Fig. 1. The formation of Nb_3Sn phases can be identified clearly by comparing the XRD patterns of as-deposited films and annealed films. Several diffraction orders of the Nb_3Sn (200, 222, 320, 321, 400, 420, 421, 332) were observed on all annealed samples. XRD patterns of sample S1 and S3 showed formation of Nb_6Sn_5 phase after annealing which was not observed in S39 and S41. All samples showed no unreacted Sn phase. Sample S1 and S3 showed strong peak of Nb 110 diffraction order and weak peaks of 211, 220 and 310 orders whereas samples S39 and S41 showed only a weak Nb 110 peak. S1 and S3 had thick coating with thicker Nb-to-Sn thickness ratio of 4.5. S39 and S41, on the other hand, had thinner coating with a Nb-to-Sn thickness ratio of 2. Thinner layers minimize formation of phases other than Nb_3Sn.

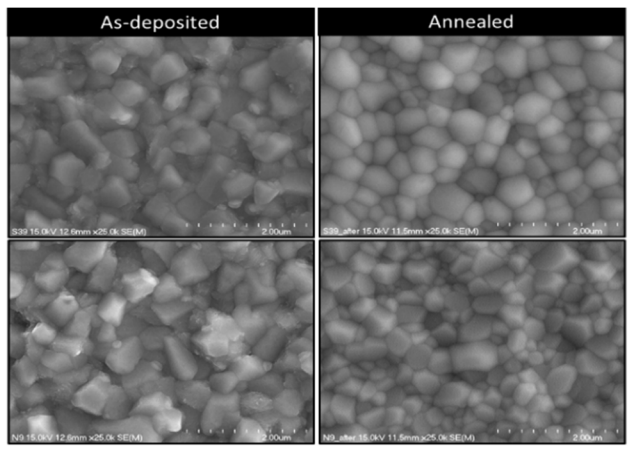

Figure 2: FESEM image of films on both sapphire and Nb substrates. Top two images show film on sapphire substrate (S39) whereas the bottom two images show film on Nb substrate (N9) before and after annealing.

Table 2: EDS Data showing Sn Concentration of the Films

Sample Name	At. % of Sn Before Annealing	At. % of Sn After Annealing
S1	14	10
S3	10	6
S39	23	19
S41	22	19

The surface images obtained from FESEM are shown in Fig. 2. For both Nb and sapphire substrates, uniform coating of Nb and Sn were observed. The annealed films showed grain structures like Nb_3Sn grain structure. A significant problem of Nb_3Sn films grown by conventional Sn diffusion process is formation of patchy regions and voids between Nb_3Sn grains in the grown film that degrades the performance of SRF cavities [15]. No such features are found in the sputtered surface images. The annealed films grown on sapphire substrate showed uniformly distributed grains of average grain size ~ 400 nm whereas the films on Nb substrate had grains from 200 nm to 800 nm with a high

ISBN: 978-3-95450-184-7 doi:10.18429/JACoW-IPAC2018-THPAL129

amount of smaller grains. The atomic concentration observed in EDS are shown in Table 2. We observed rich Nb on our first two samples (S1 and S3), however the films coated with thinner Nb-to-Sn ratio showed up to 23 At. % of Sn. For all samples, significant amounts of Sn deficiency were observed after annealing.

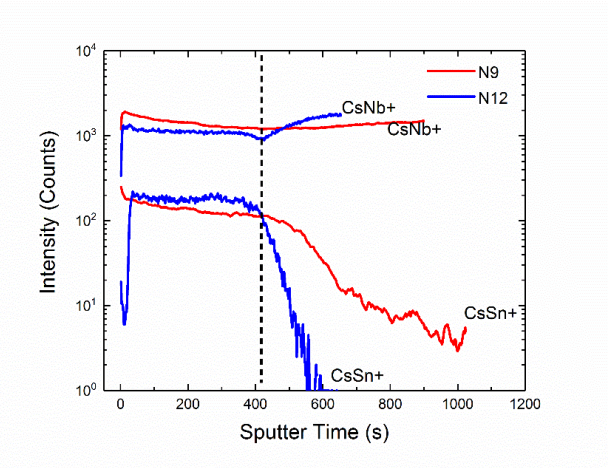

Figure 3: ToF-SIMS depth profile of as-deposited (N12) and annealed (N9) samples.

ToF-SIMS depth profile was performed on the films on Nb substrates (N9, N12) to observe Sn diffusion after annealing. Depth profile was performed on both unannealed (N12) and annealed (N9) films to observe variation. Figure 3 shows the depth profiles of the films. Due to the rough surface of the films, the individual layers on the unannealed samples were hard to distinguish. In case of unannealed samples, a sharp fall of Sn signal is observed at the inter-face whereas annealed samples showed a tail after the interface region that indicates Sn diffusion through Nb.

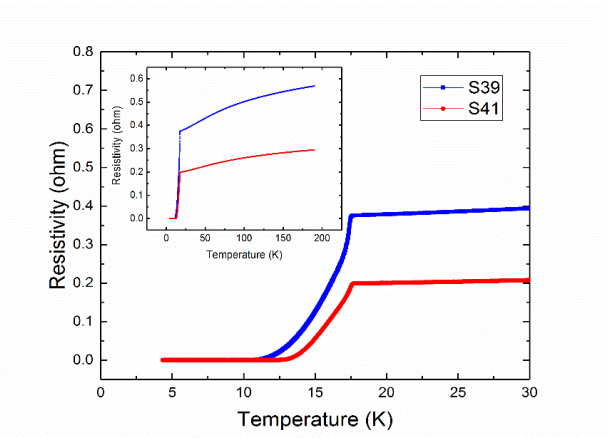

Figure 4: Surface resistivity of S39 and S41 as a function of temperature.

Surface resistivities of the films were measured by four-point probe method down to cryogenic temperatures. Figure 4 shows the superconducting transition regions of S39 and S41 with an inset image of resistivity change as a function of temperature measured. The resistivity decreases slowly as the sample temperature decreases till it reached 17.6 K where the resistivity drops to zero. However, relatively wide transition widths were observed. S1 and S3 also showed superconducting properties with a transition at 15.52 K and 17.35 K respectively.

CONCLUSION

We coated multilayers of Nb and Sn and annealed it to fabricate Nb_3Sn. The films examined by different characterization tools identified Nb_3Sn formation. Depth profile images confirmed Sn diffusion through Nb after annealing the films. The films showed good superconducting properties with T_c up to 17.6 K. The surface results showed formation of multiple phases if thicker Nb-to-Sn layers are used. All films showed Sn loss after annealing which needs to be solved to achieve good superconducting property.

REFERENCES

[1] Godeke A. Nb3sn for radio frequency cavities. Ernest Orlando Lawrence Berkeley NationalLaboratory, Berkeley, CA (US); 2006 Dec 18.

[2] Matthias BT, Geballe TH, Geller S, Corenzwit E. Superconductivity of Nb3Sn. *Physical Review*. 1954 Sep 15;95 (6):1435.

[3]Hillenbrand B, Martens H. Superconducting Nb3Sn cavities with high quality factors and high critical flux densities. *Journal of Applied Physics*. 1976 Sep; 47(9):4151-5.

[4] Kneisel P, Roeth R, Piel H, Mansen D, Pouryamout J, Müller G. Nb3Sn Layers on High-Purity Nb Cavities with Very High Quality Factors and Accelerating Gradients.

[5] Eremeev G, Clemens W, Macha K, Park H, and Williams R. "Commissioning of Nb3Sn Cavity Vapor Diffusion Deposition System at JLAB.", in *Proc. IPAC'15* (2015).

[6] Posen S, Merio M, Romanenko A, Trenikhina Y. Fermilab Nb3Sn R&D Program, in *Proc. SRF'15*. 2015 Dec.

[7] Posen S, Liepe M. RF Test Results of the First Nb3Sn Cavities Coated at Cornell. InTUP087, in *Proc. SRF'13*, Paris, France.

[8] Wu CT, Kampwirth RT, Hafstrom JW. High-rate magnetron sputtering of high T c Nb3Sn films. *Journal of Vacuum Science and Technology*. 1977 Jan; 14(1):134-7.

[9] Rosaz G, Taborelli M, Leaux F, Mydlarz Z, Motschmann F, Calatroni S, Vollenberg W. Development of Nb3Sn coatings by magnetron sputtering for SRF cavities. 2016 Jan 26.

[10] Rossi AA, Deambrosis SM, Stark S, Rampazzo V, Rupp V, Sharma RG, Stivanello F, Palmieri V. Nb_3Sn films by multilayer sputtering, in *Proc. SRF'09* (Berlin,). 2009.

[11] Vandenberg J, Gurvitch M, Hamm R, Hong M, Rowell J. New phase formation and superconductivity in reactively diffused Nb_3Sn multilayer films. *IEEE transactions on magnetics*. 1985 Mar; 21(2):819-22.

[12] Sosa Guitron S, Chang Beom E, Gurevich A, Sundahl C, Eremeev G, Delayen J. Measurements of RF properties of

ISBN: 978-3-95450-184-7 doi:10.18429/JACoW-IPAC2018-THPAL129

thin film Nb_3Sn superconducting multilayers using a calorimetric technique.

[13] Xiao L, Lu X, Tan W, Xie D, Yang D, Yang Y, Yang Z, Zhao J. The Study of Deposition Method of Nb_3Sn Film on Cu Substrate, in *Proc. SRF'17*, Lanzhou, China, pp: 131-133.

[14] Tan W, Li B, Lu X, Xiao L, Xie D, Yang D, Yang Y, Yang Z, Zhao J. Nb_3Sn Thin Film Deposition On Copper By DC Magnetron Sputtering, in *Proc. SRF'17,* pp: 512-515.

[15] Eremeev G, Pudasaini U, Tuggle J, Reece C, Kelley M. Progress with multi-cell Nb_3Sn cavity development linked with sample materials characterization.