HiPIMS-COATED NOVEL S(I)S MULTILAYERS FOR SRF CAVITIES

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Abstract

Pushing beyond the existing bulk niobium SRF cavities is indispensable along the path towards obtaining more sustainable next generation compact particle accelerators. One of the promising candidates to push the limits of the bulk niobium is thin film-based multilayer structures in the form of superconductor-insulator-superconductor (SIS). In this work, S(I)S multilayer structures were coated by high power impulse magnetron sputtering (HiPIMS), having industrial upscaling potential along with providing higher quality films with respect to conventional magnetron sputtering techniques (e.g., DCMS), combined with (PE)-ALD techniques for deposition of the *ex-situ* insulating layers. On the path towards formulating optimized recipes for these materials to be coated on the inner walls of (S)RF cavities, the research focuses on innovating the best performing S(I)S multilayer structures consisting of alternating superconducting thin films (e.g., NbN) with insulating layers of metal nitrides (e.g., AlN) and/or metal oxides (e.g., Al_xO_y) on niobium layers/substrates (i.e., Nb/AlN/NbN) in comparison to the so-called SS multilayer structures (i.e., Nb/NbN). This contribution presents the initial materials and superconducting and RF characterization results of the aforementioned multilayer systems on flat samples.

INTRODUCTION

The next generation compact particle accelerators require higher performances with reduced infrastructural and operational costs. In order to realize these goals, innovative solutions are needed to overcome the theoretical field limit along with the technological challenges set by the existing bulk niobium superconducting radio-frequency (SRF) cavity technology, which has been the leading accelerator technology for both high-energy and high-luminosity accelerator applications so far [1].

One of the promising candidates to push the limits of the bulk niobium is coating (S)RF cavities with alternating thin film-based multilayer structures in the form superconductor-insulator-superconductor (SIS).

According to the theory related to *the alternating multilayer structures* proposed by A. Gurevich [2], especially for bulk niobium cavities, the simplest alternating multilayer structures (SIS), made of superconductive thin films with thicknesses less than the London penetration depth of the cavity wall material, are expected to enhance not only the quality factor (Q_0) with lower surface resistance R_s , but also the vortex penetration field by means of the insulating layers, leading to sustain the Meissner state at higher accelerating gradients (E_{acc}), by delaying the RF vortex dissipation, and the related strong RF dissipation beyond the thermodynamic critical field of Nb (B_c (Nb)) so as to take advantage of higher T_c superconductors (e.g., B_1 and A_{15} compounds such as NbN and Nb_3Sn, respectively) without being penalized by their relatively small lower critical field (B_{c1}) with respect to the B_{c1} of Nb (200mT).

Theoretically, the SIS structure is a stronger candidate to increase the theoretical field limit as well as the onset of the vortex penetration thanks to the presence of the insulating layer, provided that the optimum layer thicknesses and material combinations are realised, as compared to the SS bilayer structure without any insulating layer; yet, the SS bilayers are also worth studying as being a simpler structure with promising RF performance of the SRF cavities by enhancing the onset of the vortex penetration to some extent owing to the SS boundary [3].

The emergence of the novel scalable sputtering technologies such as high-power impulse magnetron sputtering (HiPIMS) has provided the SRF community another tool to improve the quality of the deposited films by providing denser microstructures, more uniform morphologies, and homogenous crystalline phases in the recent years. Owing to its higher peak power magnitudes applied to the sputtering cathode in pulsed modes at certain duty cycles, HiPIMS technique yields highly ionized denser plasmas, as compared to conventional physical vapor deposition techniques, allowing more effective control of the kinetic energy of the sputtered species with high ionization fraction, arriving onto the substrate surface, by tuning the deposition parameters (i.e., substrate bias) [4].

Aside from HiPIMS, atomic layer deposition (ALD) has also drawn interest from the SRF community recently, especially for depositing insulating layers in multilayer structures [5].

The first investigations of the novel parameter space for HiPIMS-coated S(I)S structures, incorporating also (PE)-ALD-coated insulating layers as well as the further developments of the previously studied SIS structures, based on the QPR sample tests, are detailed in this paper.

EXPERIMENTAL METHODS

The multilayer S(I)S structures in the form of Nb/(AlN/Al_xO_y)/NbN, as illustrated in Fig. 1, were coated mainly onto silicon as witness samples in order to study in detailed the deposition parameters as well as onto the bulk Nb and the OFHC Cu substrates for comparison of the substrate material effects on the deposited films.

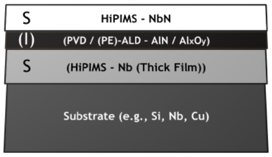

Figure 1: The schematic representation of the designed S(I)S structures together with their chosen deposition techniques.

The superconducting layers were coated by (reactive)-HiPIMS via a fully automated coating machine (CC800) of CemeCon AG GmbH at University of Siegen as shown in Fig. 2.

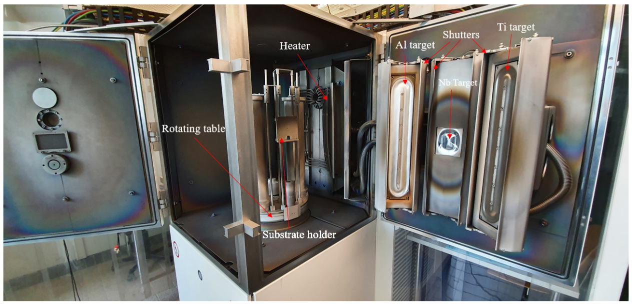

Figure 2: The overview of the sputtering machine (CC800) capable of DCMS and HiPIMS at University of Siegen (USI) - adapted from [6].

The coating machine contains three cathodes powered by ADL DC power supplies. The RRR 300 niobium sputtering target with dimensions of 88×100×10 mm^3, installed on the central cathode, which can be operated at either DCMS or HiPIMS mode, is used for the deposition of both Nb and NbN superconducting thin films under 100% argon gas (99.999 Vol-%) and argon/nitrogen reactive gas-mixture atmospheres, respectively. The aluminium target, with dimensions of 490×100×10 mm^3, located next to the Nb target, as shown in Fig. 2, was used to coat *in-situ* AlN insulating layers (*DCMS-AlN**) under 100% nitrogen gas (99.999 Vol-%) atmosphere similarly as detailed in [7]. The adjustment of the deposition pressures of both normal and reactive sputtering processes was done via the mass flow controllers and the resulted process pressures were monitored via the Baratron capacitance manometer. The programmable shutter system is used to protect sputtering targets from any unwanted contamination during the baking, MF etching, and target cleaning as well as the deposition procedures.

The realization of uniform thickness profiles along the inner walls of the cavities is indispensable for practical accelerator applications. In order to obtain uniform film thickness profiles, a special machine code has been developed via GUI programming for CC800 which allowed the rotatable table of the machine to be *rocked* at a certain *rocking angle* in front of the sputtering target during the deposition procedures. The measured distance is 55 mm when the table stays stationary opposite to the target system.

Based on the initial parameter studies [6] as well as some previous studies [7,8], the process temperature was kept constant at 400°C, corresponding to 180±5°C on the substrate surface. The HiPIMS parameters such as frequency [Hz] and pulse width [µs] were kept at constant values of 1000 Hz and 100 µs for most of the deposition processes of both Nb and NbN films based on the initial optimizations done in the range of 1000 to 2000 Hz, and 60 to 200 µs for the frequency, and the pulse width, respectively. The applied substrate bias was in DC mode in all deposition processes. The other parameters are detailed in Table 1.

Table 1: The Certain Window of the Novel Deposition Parameters for HiPIMS-coated S(I)S Structures

Material	Cathode Power [W]	Substrate Bias [V]	Deposition Pressure [mbar]	N_2 Content [%]
Nb	600-900	0-50	2.0×10^{-2}	0
(AlN*)	3500	0	6.0×10^{-3}	100
NbN	600-750	0-50	$2.0\text{-}2.7 \times 10^{-2}$	8, 9

The *ex-situ* insulating layers, AlN and Al_xO_y, were coated at CHyN / UHH by (PE)-ALD, and thermal ALD techniques, respectively.

The superconducting properties (e.g., the superconducting critical temperature (T_c), and the entry field ($B_{en} \sim B_{c1}$) of the deposited samples were characterized at IEE SAS via the vibrating sample magnetometer (VSM) technique.

The deposition parameters of HiPIMS-coated SIS and SS structures, analysed with Cu-QPR sample tests at HZB, are detailed in Table 2.

Table 2: The Deposition Parameters of S(I)S Structures

Material	Cathode Power [W]	Substrate Bias [V]	Deposition Pressure [mbar]	N_2 Content [%]
Nb	400	50	8.0×10^{-3}	0
(AlN*)	3500	0	6.0×10^{-3}	100
NbN	400	50	2.5×10^{-2}	10

The materials characterizations of all deposited films were done via SEM, EDX, and XRD at USI.

RESULTS AND DISCUSSION

The duty cycle of HiPIMS plays a crucial role in obtaining high quality thin films with desired morphological and microstructural properties. As shown in Fig. 3, the lower duty cycle depositions of HiPIMS-coated NbN films tend to promote better structural morphologies while leading to the formation of δ-NbN phase with (200) orientation compared to the higher duty cycle depositions which seem to result in less desired film morphologies and microstructures while enhancing δ-NbN (111) phase formation.

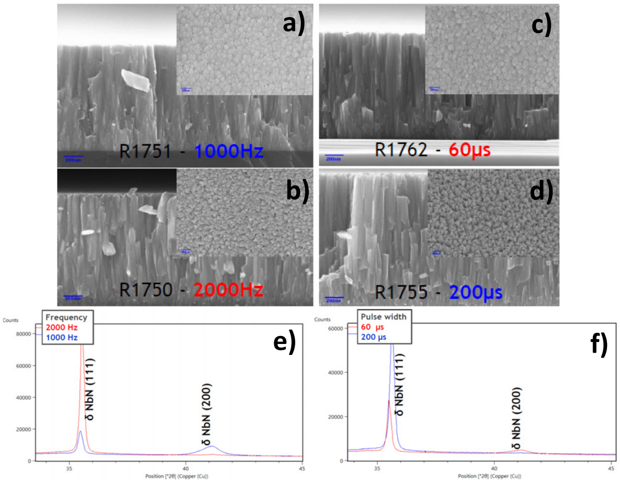

Figure 3: The SEM images of HiPIMS-coated NbN at: a) 1000 Hz, b) 2000 Hz, c) 60 µs, d) 200 µs. The X-ray diffractograms of the aforementioned films as a function of: e) frequency, f) pulse width.

As expected, the application of the negative substrate bias seems to enhance the density of the films relatively compared to the unbiased HiPIMS coating of SS structures (NbN films on Nb layers); however, the crystalline structure transits into completely non-superconducting phase (e.g., Nb_5N_6) from the desired δ-NbN (111) phase, as shown in Fig. 4, in particular at these high cathode power-depositions (the bottom layer: Nb (at 900W) / the top layer: NbN (at 750W)).

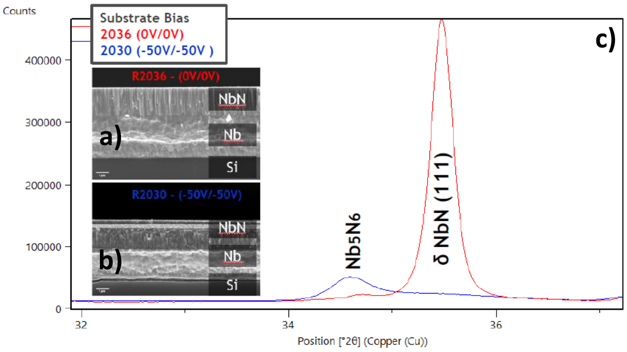

Figure 4: The SEM images of SS structures: a) at 0 V, b) -50 V. c) The crystallinity shift as a function of bias.

Along with the surface quality of the substrate (i.e., cavity surface), onto which thin films are to be coated, the material of the substrate is paramount important too as it changes not only the material properties, but also the superconducting properties significantly as shown in Fig. 5 and Table 3. The best crystallinity as well as the highest critical superconducting temperature for HiPIMS-coated NbN films was observed on bulk copper substrate. Both *ex-situ* insulating layers, thermal ALD-coated Al_xO_y and PE-ALD-coated AlN, seemed to suppress T_c values compared to the bulk metallic substrates in correlation to the induced poorer crystallinity in HiPIMS-coated NbN films.

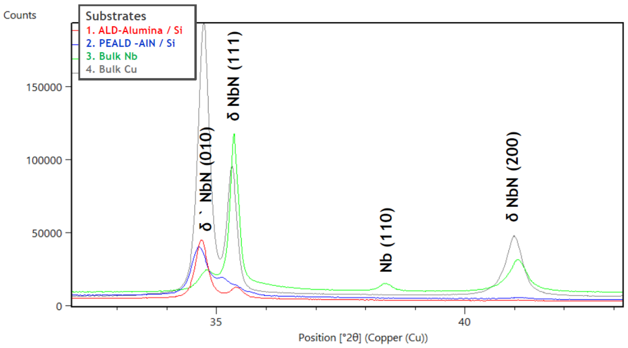

Figure 5: The substrate material effects on the crystallinity of the HiPIMS-coated NbN films.

Table 3: The VSM Results of HiPIMS-NbN coated on Different Substrates

Substrate	B_{en} [Oe]	T_c [K]
1. ALD-Alumina / Si	80	15
2. PE-ALD-AlN / Si	100	15
3. Bulk Nb	1060	15.8
4. Bulk Cu	110	16.1

As seen in Fig. 6, the insulating layer changes the morphology of SIS as compared to the SS structure.

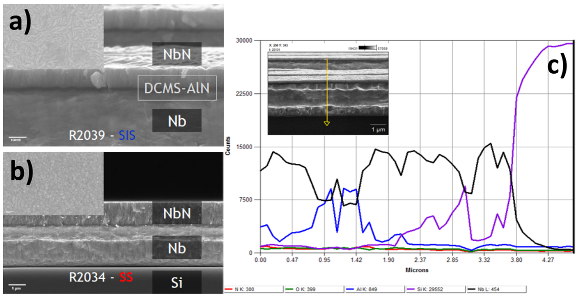

Figure 6: The SEM images of surface and cross-section of a) SIS, and b) SS structures. c) The EDX data of SIS.

CONCLUSION

The recent studies regarding the RF performance of HiPIMS-coated SIS versus SS structures with the improved quadrupole resonator (QPR) yielding higher resolution measurements of flat samples [9], point out that both multilayer structures enhance Q_0 similarly [10]. Further investigations are needed to establish stronger correlations between S(I)S structures and their SRF performance.

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