ISBN: 978-3-95450-227-1 ISSN: 2673-5490 doi:10.18429/JACoW-IPAC2022-TUPOTK013

PEALD SIS STUDIES FOR SRF CAVITIES

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Abstract

Recent technological advances and material treatments have pushed Nb superconducting radio frequency (SRF) cavities to their maximum RF performance. A novel approach for overcoming this limitation is the coating of multilayers by PEALD (plasma-enhanced atomic layer deposition) onto the interior surface of a cavity. Specifically, SIS (superconductor-insulator-superconductor) multilayers provide magnetic screening of the bulk Nb cavity, increase the field at which the vortex penetration starts, and lead as a consequence to higher quality factors of the cavity. Note, ALD is closely related to chemical vapor deposition and bases on sequential self-limiting gas-solid surface reactions facilitating conformal coatings with sub-nm precision even on complex substrates such as the interior of a cavity. As a preliminary study for potential SIS SRF cavities, we investigated AlN-NbTiN multilayers grown by PEALD in a supercycle approach. Different compositions and post-deposition thermal treatments have been investigated with respect to their superconducting properties, stoichiometry, and crystallinity.

INTRODUCTION

Over the past decades, bulk niobium has been the material of choice for SRF cavities, since it satisfies the requirement of having a high critical temperature (TcNb=9.2 K) and high lower critical field (Bc1Nb=170 mT), being widely investigated [1]. Different surface treatments have continuously improved the RF performance pushing up the accelerating field into the intrinsic material limit (B0≃200 mT; Eacc≃50 MV/m) [2]. This field limitation for SRF cavities is established by the superheating field Bs corresponding to the maximum magnetic field that the superconductor can withstand before the Meissner state becomes unstable and vortices penetrate at the superconductor surface defects which, at the low operating temperature, would develop a flux avalanche and cavity deterioration [3,4]. Thus, alternative superconductors with higher vortex penetration fields are needed in order to achieve higher acceleration gradients.

In this framework, an alternative approach proposed by A. Gurevich [5] may allow for applying higher accelerating fields while preventing vortex dissipation and revealing low RF surface resistance at the same time. This idea bases on the formation of alternating thin superconducting and insulating layers (SIS multilayers, see Fig.1) onto the inner surface of an SRF cavity (see Figure 1). Due to the strong increase of the first flux penetration in a thin film (where d<<λ), type II superconductors with $T_c > T_c^{Nb}$, and consequently lower surface resistance, can be used without being limited by their lower B_{c1}^{bulk}. Moreover, the SIS layers provide a significant magnetic shielding of the bulk cavity, block the propagation of local vortices and prevent avalanches which would cause a quench. Therefore, SIS structures improve the SRF cavity performance and can lead to an increase in the accelerating field and the RF performance. Specifically, A. Gurevich calculated this enhancement assuming a 50 nm of Nb_3Sn deposited on a Nb bulk cavity with an insulating interlayer would push up the field from B_0≈180 mT to B_0≈280 mT, and triple the quality factor Q.

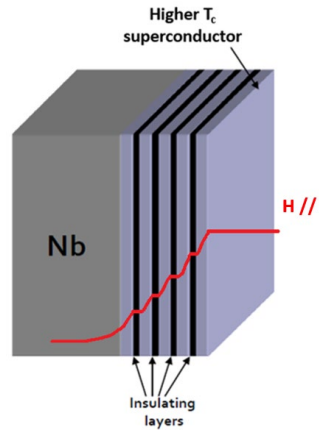

Figure 1: SIS multilayers for SRF cavities concept. The magnetic field is attenuated by the multilayers, leading to a reduction to a value that is lower than Bc1bulk for a bulk Nb cavity.

In addition, the use of other superconductors with lower BCS resistance, such as Nb3Sn, NbN or NbTiN, [6] offers the possibility of increasing the operating temperature to 4.2 K, resulting in a significant cryogenic cost reduction.

Good superconductors for SIS multilayer SRF applications need to have higher Tc, larger Δ, and lower normal conducting resistivity than bulk Nb. Some of the compounds which satisfy the aforementioned requirements are A15-compounds (such as Nb3Sn, Nb3Al, and V3Si) and Nb B1-compounds (such as NbN and NbTiN). In particular, our studies are based on previous results on binary and ternary Nb nitrides, since both present high Tc (T_c^{NbN}=17.3 K and T_c^{NbTiN}=17.8 K) [6]. However, the binary NbN cubic δ-phase (phase of interest) is metastable at room temperature, and has a very high normal conducting resistivity,

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while the incorporation of Ti contributes to the stabilization of the cubic phase and reduction of the normal conducting resistivity.

NbTiN alloys can be deposited by physical vapor deposition (PVD), chemical vapor deposition (CVD), and atomic layer deposition (ALD), being the latter the deposition technique chosen for our studies. ALD bases on a sequence of self-limiting gas-solid surface reactions and allows for conformal and smooth coating of highly structured, three-dimensional substrates without shadowing effect and with sub-nm thickness resolution, which makes it particularly interesting for coating the internal surface of SRF cavities. The deposition process of NbTiN by thermal ALD alternates metal chlorides precursors ($NbCl_5$ and $TiCl_4$) and NH_3. However, it has been shown that the reducing power of NH_3 is insufficient [7] and an additional Zn pulse—as source of Zn contaminants—as a reducing agent is needed to obtain high-quality films. In addition, the use of metal chlorides precursors can introduce chlorine contamination [8] on the deposited films. Finally, as vaporous HCl is formed as by-product which is highly corrosive. Therefore, plasma-enhanced ALD (PEALD), which enables the use of metalorganic precursors, may improve the quality of the deposited films, can lower deposition temperature, and could be a potentially better technique for the deposition of NbTiN on SRF cavities than thermal ALD. Moreover, PEALD enables AlN deposition, which is a promising candidate for the insulating layer, to enhance the superconducting properties of NbTiN films [9].

EXPERIMENTAL DETAILS

We investigate the deposition of superconducting $Nb_xTi_{1-x}N$ and insulating AlN films grown on Si wafer by PEALD utilizing metalorganic precursors and a H_2/N_2 plasma. The precursors used were trimethylaluminum (TMA) for Al, tetrakis(dimethylamino)titanium(IV) (TDMAT) for Ti, and (t-Butylimido)tris(diethylamino)niobium(V) (TBTDEN) for Nb, and were maintained at room temperature, 70 ºC and 90 ºC respectively. The deposition temperature was set to 250 ºC and the plasma power to 300 W.

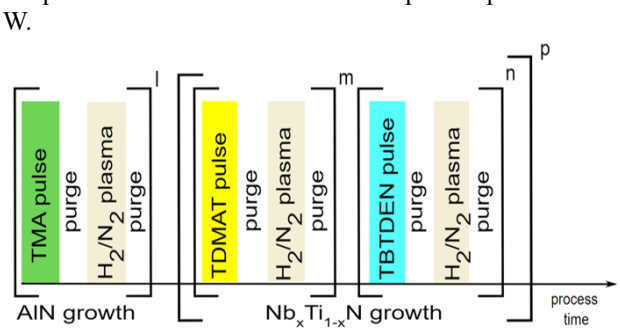

Figure 2: Schematic illustration of deposition cycles of AlN and supercycles of $Nb_xTi_{1-x}N$.

The deposition process is sketched in Fig. 2 and begins with the AlN PEALD cycle, which consists of the alternation of TMA precursor and plasma exposures separated by purge steps. This AlN cycle is repeated *l*-times, to reach the desired thickness. Subsequently, PEALD of$Nb_xTi_{1-x}N$ is performed in supercycle fashionconsisting of PEALD cycles for the deposition of NbN (alternation of TBTDEN pulse and plasma exposure) and TiN cycles (alternation of TDMAT pulse and plasma exposure). Hence, the composition of the deposited $Nb_xTi_{1-x}N$ films can be modified by varying the ratio of NbN cycles to TiN cycles run within the PEALD supercycle. Herein, we studied eight different Nb:Ti composition ratios ranging from pure TiN up to a Nb-rich NbTiN thin film.

After the PEALD, thermal annealings have been performed at 1000 ºC in pure N_2. Two different annealing procedure have been studied: (i) with a ramping rate of 60 ºC/min at a base pressure of 1E-3 mbar, which is denominated rapid thermal annealing (RTA), (ii) with 3.33 ºC/min at 2.5E-6 mbar, denominated slow thermal annealing (STA).

High pressure rinsing (HPR), which is a key process for the surface preparation of high field superconducting cavities, has been performed on a Nb conical substrate coated with 15 nm AlN and 30 nm NbTiN films to explore the adhesion of the multilayer film on the Nb bulk material.

RESULTS

The structural, compositional, and superconducting properties of 25 nm $Nb_xTi_{1-x}N$ films on an AlN layer has been studied as a function of the ratio of the relative number of Nb cycles inside the PEALD supercycle and in dependence on the post-deposition treatment.

The film composition was analysed using EDX, for eight different $Nb_xTi_{1-x}N$ compositions defined by the supercycle approach. The linear relationship between the Nb/Ti film composition and the NbN/TiN ALD cycles demonstrates the well-defined control of the elemental composition of our films (see Fig. 3). Note, the slightly smaller value than unity for the compositional ratio as a function of time indicates a slightly smaller growth rate for NbN compared to TiN which is expected because of the larger and more bulky molecule TBTDEN.

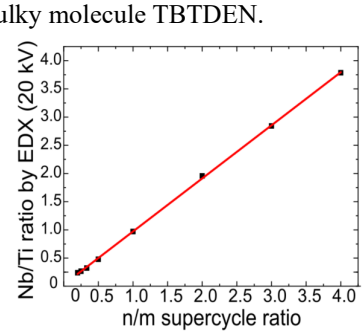

Figure 3: Elemental composition ratio of Nb to Ti in $Nb_xTi_{1-x}N$ films as a function of the Nb to Ti supercycle ratio, measured by EDX. The red line represents the linear relationship between the elemental composition and the PEALD supercycle with slope of 0.941 and R^2=0.999.

Superconducting properties of the thin films have been measured in an MPMS Dynacool system. In general, the resistance tends to increase as the temperature decreases resembling a semiconductor behavior, more prominent for

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larger amounts of Nb present in the NbTiN compound as highlighted in Fig. 4. Note, that the composition which reveals the highest T_c as-prepared is $Nb_{0.66}Ti_{0.33}N$, with a T_c of 7K.

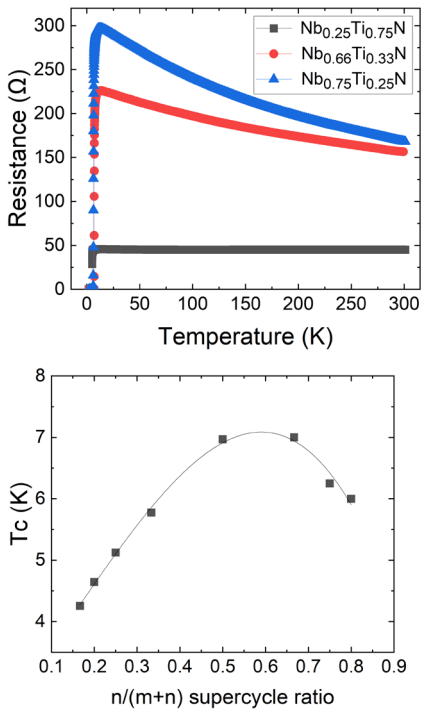

Figure 4: Top: resistance as a function of temperature for different $Nb_xTi_{1-x}N$ compositions. Bottom: critical temperature as a function of the relative number of Nb cycles inside the supercycle.

In order to prove the validity of the multilayer deposited by PEALD for SRF cavity coating, we have studied whether these films can resist the typical cavity surface preparation process. Hence, a Nb substrate coated by the 15 nm AlN and 30 nm NbTiN on top has been gone under high pressure rinsing (HPR). The EDX spectrum after performing 7x HPR (standard cavity treatment) presents Al, Ti and Nb indicating that the coating remains after such a surface treatment (not shown here).

A post-deposition thermal annealing at 1000 ºC has been studied in order to enhance the superconducting properties of 75 nm NbTiN films. Two different thermal treatments have been investigated, and in both cases, the T_c and the resistance of the films have been improved (see Fig. 5). The $Nb_xTi_{1-x}N$ composition with the highest T_c after thermal annealing was $Nb_{0.75}Ti_{0.25}N$ which is in contrast to the as-deposition values. The highest T_c observed in our study amounted to 15.9 K after STA of $Nb_{0.75}Ti_{0.25}N$ thin films, higher than the values so far obtained by thermal ALD (8.5 K [10]) and by PEALD (13.2 K [11]).

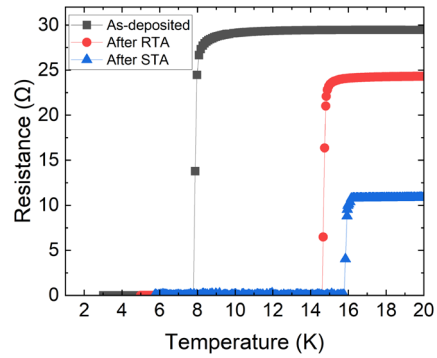

Figure 5: Comparison of the superconducting transition of as-deposited, after RTA and after STA 75 nm NbTiN films.

Electron backscatter diffraction (EBSD) has revealed that the crystallization after thermal annealing of the films depends on the annealing procedure, namely STA and RTA. No signal for Kikuchi lines can be identified in Fig. 6 for the as-deposited thin film, while after applying RTA recrystallization can be observed and even more significantly after STA.

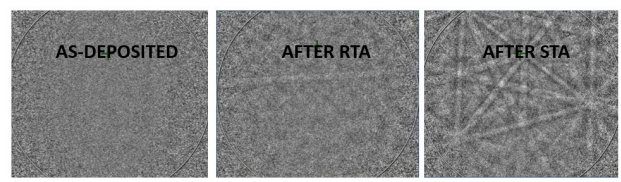

Figure 6: Electron diffraction patterns (Kikuchi lines) for as-deposited, after RTA, and after STA samples, where one can clearly see no signal for the as-deposited ones, a weak signal for the RTA samples, and a remarkable signal for the STA samples.

CONCLUSIONS

Insulating AlN and superconducting NbTiN films have been synthesized by PEALD. The characterization of such multilayers serves as a first stepping stone on the path towards SIS multilayers deposited by PEALD on SRF cavities. The effect of their composition and post-deposition annealing on the T_c has been studied. Two different annealings, with different temperature ramping rates and base pressures, namely rapid thermal annealing (RTA) and slow thermal annealing (STA), have been investigated. The results show an improvement of T_c as well as resistance for both kinds of annealing. However, STA reveals better film properties with the highest T_c of 15.9 K and the lowest resistance above T_c for a 75 nm thin film. Kikuchi lines obtained from electron diffraction show that the crystallinity of the films is enhanced by the post-deposition annealing. The $Nb_xTi_{1-x}N$ composition which presents the highest T_c, changes for the as-deposited, being the composition $Nb_{0.66}Ti_{0.33}N$, while after annealing it is the $Nb_{0.75}Ti_{0.25}N$.

The EDX spectrum exhibits Al, Ti and Nb peaks, demonstrating that the thin films survived seven high pressure rinsing processes, treatment typically done for surface preparation cavities.

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