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SUCCESSFUL Al_2O_3 COATING OF SUPERCONDUCTING NIOBIUM CAVITIES WITH THERMAL ALD

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Abstract

Surface modification of superconducting radiofrequency (SRF) cavities is mandatory to further push the limits in future accelerators. One strategy is the deposition of a multilayer of superconducting and insulating materials on top of the inner surface of a SRF cavity. Here we report on a successful low-temperature coating of a SRF cavities with insulating Al_2O_3 by thermal atomic layer deposition (ALD) without mitigating their maximum achievable accelerating field of more than 40 MV/m. Furthermore, an improvement of the surface resistance above 30 MV/m has been observed. Our results show that ALD is perfectly suited to conformally coat the interior of the cavity and to even modify and improve the properties of such devices.

INTRODUCTION

To achieve significant advancements in superconducting radio frequency (SRF) technology, it is crucial to explore new concepts that can enhance accelerating fields and improve cavity performance. These developments are necessary to address the demanding requirements posed by upgrades to existing accelerators or the construction of future ones [1, 2]. Our research focuses on a particularly promising direction known as the superconductor-insulator-superconductor (SIS) or multilayer approach. This approach involves coating the inner surface of Nb cavities with alternating layers of superconducting and insulating materials. The concept of this structuring was initially proposed by Gurevich in 2006 [3], and it has been further refined through theoretical studies to determine the optimal layer thickness for improved RF performance [4, 5].

In order to surpass the performance of Nb cavities, a promising approach involves coating the inner surface with thin films or multilayers of superconductors that have higher critical temperatures than niobium, such as NbN or NbTiN. These composite accelerator cavities are anticipated to exhibit lower RF losses and achieve higher accelerating gradients, potentially surpassing 100 MV/m [3].

To achieve this objective, it is important to tailor the deposited superconducting film. Specifically, the thickness of the superconducting layer, denoted as d_S, should be smaller than the London penetration depth (λ_L) of the coated superconductor, and d_S should be less than 100 nm. Moreover, the higher critical magnetic field (H_{c1}) of these multilayers should exceed the superheating field (H_{sh}) of niobium. This characteristic allows for the application of higher accelerating fields compared to niobium. The presence of the thin higher-H_c layers acts as a magnetic shield, preventing the penetration of vortices into the bulk superconducting cavity. This magnetic screening effect is further enhanced by the insulating layer, which typically has a thickness (d_I) ranging from approximately 5 to 20 nm.

In this work, we conducted a comprehensive study to examine the effects of introducing an insulating layer of Al_2O_3 on the inner surface of SRF cavities using thermal atomic layer deposition (ALD). The purpose of depositing insulating layers on the RF surface was to gain insight into the overall impact of the coating process on the RF performance. Achieving a uniform coating with an insulating layer, while maintaining or improving cavity performance, is a significant milestone on the path towards coating cavities with a multilayer structure. This investigation aimed to understand the influence of the insulating layer and ensure that it does not adversely affect the performance of the cavity.

EXPERIMENTAL DETAIL

The main part of the experiments and results summarized herein have been recently published in Wenskat et al., SUST [6]. The work was divided into three phases: first the recipe development on samples, then the transfer of the optimized recipe to a test (dummy) cavity, and finally coating of actual cavities. It is important to note that the cavity is not within a larger ALD chamber, but rather resembles the ALD chamber itself, and only the inner surface is coated.

The ALD processes for depositing Al_2O_3 coatings were carried out on an in-house developed thermal ALD system. The system maintained a base pressure of 10^{-3}mbar and utilized trimethylaluminum (TMA) and purified water (H_2O) as precursors.

To prepare the cavity for the coating process, it was initially evacuated until reaching the base pressure of 10^{-3}mbar. Following that, a constant flow of nitrogen 6.0 at a rate of 20 SCCM was introduced into the system as a carrier/purge gas. As a result, the working base pressure increased to approximately 1mbar. This controlled environment provided the necessary conditions for the ALD process to occur effectively, ensuring proper deposition of Al_2O_3 coatings on the cavity's inner surface.

In the initial stages of recipe development, we utilized SiO_2/(100)Si wafer substrates. These substrates allowed us to fine-tune the deposition parameters and optimize the process. Later on, we extended the recipe development to conical-shaped fine-grain Nb substrates. The deposition temperature during the optimization process ranged from 50 – 200 °C, with a focus on achieving target film thicknesses below 20 nm. These thicknesses align with the proposed insulator thicknesses for SIS structures. Detailed in-

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formation on the homogeneity of film thickness in this study can be found in [6]. We conducted additional optimization on a dummy cavity, as shown in Fig. 1(a). Reference Si substrates were placed beneath the cavity and at the top flange, while heating wires were used to control the temperature of the cavity. Additionally, a specially designed sample holder, tailored to fit the cavity, was utilized by placing reference substrates inside the cavity.

For precised temperature control and improved homogeneity during the process, a custom-made heating jacket, designed to match the shape of the cavity, was employed. This further facilitated precise temperature management and improved the uniformity of the coating process.

In the ALD process, the first precursor used was TMA, which was pulsed into the cavity volume. To ensure precise control over the reaction, a stop-valve was employed to disconnect the rotary vane vacuum pump from the deposition chamber during a specified exposure time. This allowed for ligand-exchange reactions to occur and the completion of each half-cycle reaction. Following the TMA exposure, the chamber underwent a purging step. In this step, the pumping line was reconnected to the deposition chamber, and any excess precursors and byproduct gases were removed from the system. This purging process ensured the removal of any remnant that could interfere with subsequent deposition steps. After purging, H_2O pulse was introduced into the system. The H_2O reacted with the surface, resulting in the formation of a hydroxylated monolayer of Al_2O_3.

RESULTS

Recipe Development

The aim of this part of our study was two-fold: (i) to identify a working set of parameters to achieve a homogeneous coating by an Al_2O_3 layer for the large cavity surface, while (ii) at the same time minimize the associated annealing process by reducing the thermal budget, which is understood as the area under the temperature-time-curve, to reduce the impact of diffusion processes on the RF performance.

Sample Studies

To optimize the ALD process, we focused on the process time for individual pulse, exposure, and purge steps. The operational temperature was fixed at 120 °C, a nitrogen (N_2) flow rate of 20 SCCM was used and 100 process cycles.

To evaluate the effectiveness of different recipes in achieving full and homogeneous coverage of the cavity, a dummy cavity was used. Si substrates were strategically placed at the bottom and top of the cavity, as shown in Fig. 1(a). This configuration allowed us to determine the recipe that resulted in the most uniform coating across the entire cavity surface. Multiple recipes were tested and are summarized in Table 1. The film thicknesses of samples coated with these recipes are shown in Fig. 1(b). Among the tested recipes, Recipe III demonstrated the same film thickness at both the bottom and top of the cavity within the shortest duration per cycle.

To further assess the homogeneity of film thickness within the dummy cavity, a dedicated sample holder matching the cavity shape was designed. Applying Recipe III for 84 cycles in two separate coating runs on the dummy cavity resulted in a homogeneous coating of (18 ± 1)nm along the cavity surface, as depicted in Fig. 1(c). These findings validated the excellent conformality and reproducibility of the ALD process. Based on the successful optimization and homogeneity achieved, Recipe III was selected for coating three niobium SRF cavities. These cavities were coated at a constant film thickness of 18,nm using Recipe III at an operational temperature of 120°C.

Table 1: Recipes tested for 120 °C and a N_2 flow of 20 SCCM. Duration given for each cycle phase Fig. 1(b).

	TMA			H_2O			Cycles
	Pulse /ms	Exposure /s	Purge /s	Pulse /ms	Exposure /s	Purge /s	
I	50	12	120	50	12	120	100
II	250	45	120	500	45	120	100
III	500	60	120	500	60	120	100
IV	500	60	240	500	60	240	100

To investigate the interface between the coated layer, the native niobium oxide, and the niobium substrate, secondary-ion mass spectrometry (SIMS) was employed. The SIMS measurement confirmed the presence of an 18 nm Al_2O_3 layer, as shown in Fig. 2(a).

Mechanical Film Stability

In order to asses the coated layer withstand high pressure rinsing (HPR) and to detect any potential damage such as cracks or delamination appear, two coated niobium samples were investigated with scanning electron microscope (SEM) before and after multiple HPR. The SEM inspection could not reveal any obvious defects (cracks, flakes, delamination) in the layer after the HPR, down to a resolution of 60 nm, Fig.2(b) and EDX analysis confirmed the existence of the Al_2O_3 layer before and after the HPRs see Fig. 2(c). These results indicate that the coating successfully withstood the HPR process without exhibiting any noticeable damage or degradation.

Cavity Coating

1Z1 :

The cavity achieved an accelerating field of 20 MV/m with a hard quench limiting the performance. The quality factor is above $2x10^{10}$ till the quench, and hence a good test cavity, as it is sensitive to any fundamental problem due to the coating procedure. The cavity underwent the baseline measurement, received a first 18 nm coating of Al_2O_3 and was measured again to be compared to the baseline. After that, it received another 18 nm coating, underwent the preparation procedure once more and was tested again. The overall cavity performance does not change after the coatings and is detailed in Ref. [6].

1DE18 :

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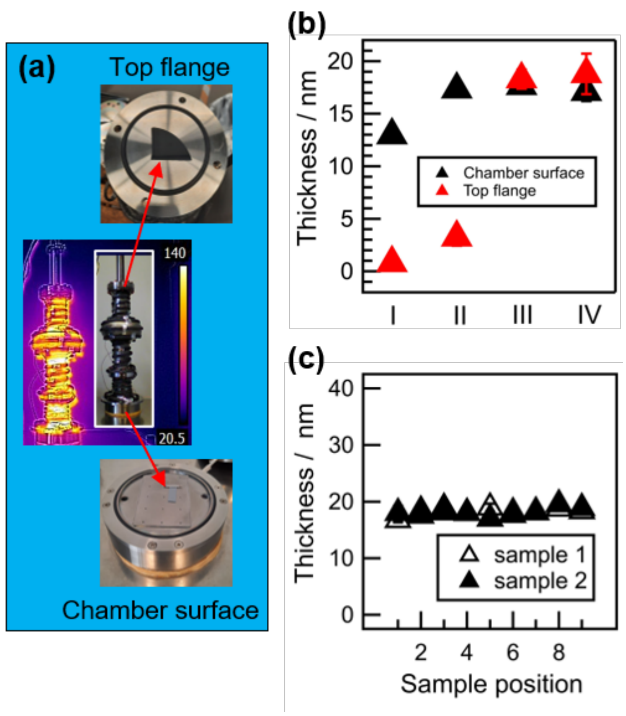

Figure 1: (a), Infrared image taken of a dummy cavity with heating wires around during a parameter scan. The inner surface area of a 1.3 GHz single-cell TESLA cavity which has to be coated is about 0.11 m^2. The Si samples are placed at the bottom and top of the cavity. (b), Homogeneity of the film thicknesses compared for the top and bottom samples, placed as shown in Fig. 1(a), for the four different recipes described in Table 1. Thickness of the Al_2O_3 layer along the cavity surface for two coatings according to recipe III. Sample 9 is a the upmost position, sample 1 at the lowest position in the cavity.

After successfully confirming the feasibility of the process in the proof of principle test of 1Z1, we coated a cavity which achieved a maximum accelerating field of 40 MV/m in the baseline measurement. With this, we intended to show that the coating maintains high accelerating fields without creating field emission. Furthermore, we wanted to study other possible additional loss mechanisms of the coating at higher fields.

Figure 3(a) presents a comparison of the quality factor (Q-factor) at 2 K for the baseline measurement before coating to the measurement after the 18 nm coating .

The remarkable finding of the study was the absence of any degradation in the cavity's performance after the coating process. This was evident in the preservation of the extraordinary performance of the cavity, as no deterioration was observed. To further analyze the effects of the coating, additional measurements were conducted to determine the residual resistance (R_{res}) and the reduced gap (Δ/k_bT_c). The surface resistance was measured as a function of temperature, as depicted in Fig. 3(b). Note, the contribution from R_{BCS} is suppressed at lower temperatures (i.e., 1.5 K) and

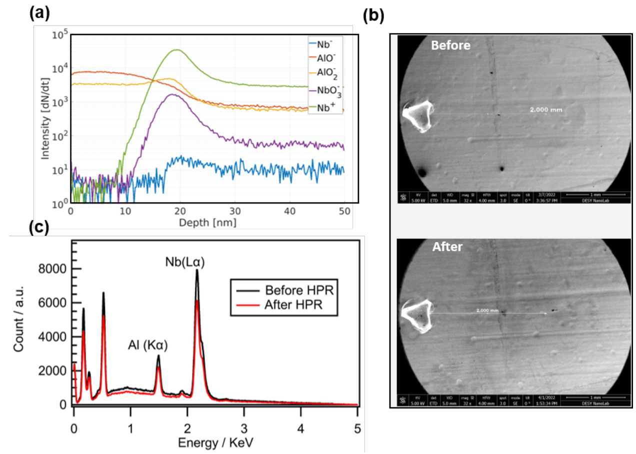

Figure 2: (a) Secondary ion concentration as a function of depth obtained by SIMS measurement of a coated niobium sample. (b) Exemplary SEM images of the sample before (top) and after (bottom) undergoing seven HPR. The triangular shaped marker dent is seen. No defects in the layer were found. (c) Energy-dispersive X-ray spectra taken before and after seven HPR. The Al peak is obvious.

the measured surface resistance is predominantly influenced by R_{res} as shown in Fig. 3(c).

The residual resistance at (4 ± 0.5) MV/m remains unchanged: It was (2.9 ± 0.2) nΩ and (2.8 ± 0.3) nΩ before and after the coating, respectively. In addition, the reduced gap was unchanged, too: The values amount to 1.76 ± 0.05 and 1.76 ± 0.07 before and after ALD coating, respectively, indicating no significant alteration due to the ALD coating process. Noteworthy, and contrary to the fact that the residual resistance was unchanged within uncertainty at low accelerating fields, we observed a distinct difference at higher accelerating fields (>30 MV/m). In detail, at low accelerating fields, the measurement almost equaled the regular R_s vs. T measurement. But the surface resistance of the coated cavity decreased slower with increasing accelerating field than the cavity before coating. At 30 MV/m, the surface resistance, i.e., residual resistance at this temperature, was improved by (0.7 ± 0.3) nΩ by the coating. This improvement can be attributed to two factors: First, modification of native Nb_2O_5 by Al_2O_3 coating as TMA precursor can potentially reduce the substrate surface [7]. Second, 120 °C baking during ALD process can cause modification of the mean free path within the RF surface [8] and a trapping of hydrogen at interstitial oxygen atoms, preventing the formation of lossy niobium hydrides at cryogenic temperatures [9, 10]. Thus, it is obvious that high accelerating fields can be maintained with a coating of Al_2O_3.

The measurement of the cavity frequency as a function of temperature (Fig. 3(d)) revealed two important observations. Firstly, an increase of the frequency shift compared to the baseline and, secondly, a not altered T_c. Assuming the frequency shift solely comes from a change of the resonating volume, the shift can be converted into a change of the pene-

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tration depth $\Delta\lambda$ of the RF field and an effective penetration depth λ_0. This assumption is modeled by using Slater's Theorem and the Gorter-Casimir model [11]. Calculating the effective penetration depth yields to $\lambda_0 = (67 \pm 1)$ nm for the coating, meaning the cavity is in the Pippard Limit and the resulting mean free path is in the range of typical values for cavity surfaces. In contrast, the effective penetration depth for the baseline only amounts to $\lambda_0 = (8 \pm 2)$ nm, which is unusually low. As this value shows an extreme clean surface, no mean free path can be derived.

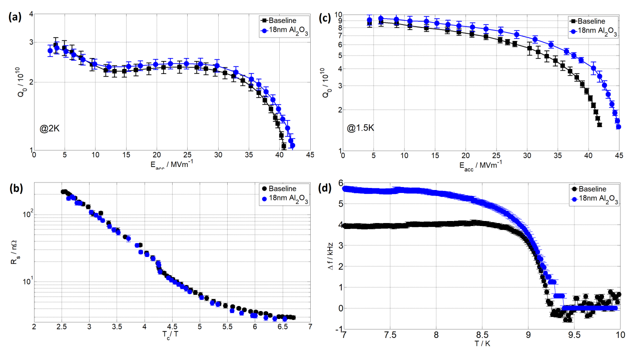

Figure 3: (a) Quality factor vs. accelerating field of 1DE18 measured at 2 K. The lines serve as guide to the eye. The baseline (black) compared with the coating of 18 nm Al_2O_3 (blue). (b) Surface resistance vs. inverse temperature for 1DE18 measured at (4 ± 0.5) MV/m. (c) Quality factor vs. accelerating field of 1DE18 measured at 1.5 K. (d) Frequency shift of the resonance frequency vs. temperature.

1DE10 :

In addition to Ref. [6], anther cavity, namely 1DE10 has been coated with 18 nm of Al_2O_3 with the same process parameters as the other cavities, proving the successful repeatability of our coatings . The comparison of the quality factor at 2 K for the baseline measurement before coating to the measurement after the 18 nm coating is shown in Fig. 4. This cavity also maintained its extraordinary performance after coating and achieves a higher gradient. The observed improvement is still under investigation, but is most likely due to different test environments.

CONCLUSION

The RF results of three Al_2O_3-coated SRF cavities from our laboratory, including two Al_2O_3-coated cavities to achieving more than 40 MV/m, are presented here. We showed that the coating does not have a detrimental effect on the accelerating field of the cavities, and even a reduction of the residual resistance above 30 MV/m is found in one case. The origin of this improvement can be explained by either an oxide layer reconstruction or an enhanced oxygen diffusion into the lattice. The studies and results presented here are a proof-of-principle experiment for further SIS studies and might pave the way to future experiments achieving an improved RF performance of cavities.

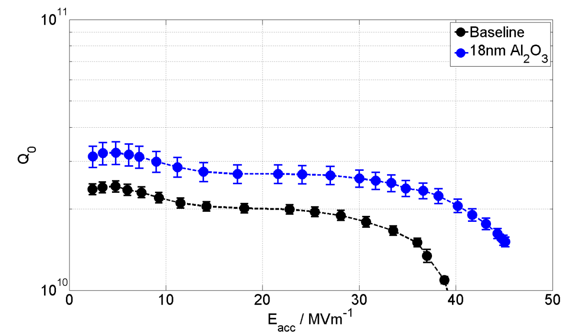

Figure 4: Quality factor vs. accelerating field of 1DE18 measured at 2 K. The lines serve as guide to the eye. The baseline (black) compared with the coating of 18 nm Al_2O_3 (blue).

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