ATOMIC LAYER DEPOSITION FOR SRF CAVITIES

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Abstract

We have begun using Atomic Layer Deposition (ALD) to synthesize a variety of surface coatings on coupons and cavities as part of an effort to produce rf structures with significantly better performance and yield than those obtained from bulk niobium, The ALD process offers the possibility of conformally coating complex cavity shapes with precise layered structures with tightly constrained morphology and chemical properties. Our program looks both at the metallurgy and superconducting properties of these coatings, and also their performance in working structures. Initial results include: 1) results from ALD coated cavities and coupons, 2) new evidence from point contact tunneling (PCT) showing magnetic oxides can be a significant limitation to high gradient operation, 3) a study of high pressure rinsing damage on niobium samples.

INTRODUCTION

Steady progress in superconducting radio-frequency (SRF) accelerator cavity development has recently resulted in demonstration of operating field gradients of 45-52 MV/m on prototype Nb cavities [1-5], for which the peak equatorial RF fields 180-190 mT have almost reached the dc depairing limit set by the thermodynamic critical field $H_c(0) \approx 200$mT of Nb. In turn, these remarkable achievements indicate that the Nb cavity technology may have reached its fundamental intrinsic limit, so the traditional incremental empirical refinement of the Nb cavities may unlikely produce further significant increase of the RF field gradients and decrease the accelerator cost. Since the theoretical maximum gradient is proportional to H_c, going from Nb (H_c~200 mT) to Nb_3Sn (Hc~540 mT), for example, could almost triple the limiting gradient. In addition to theoretical limits, the manufacture of real cavities usually implies a number of other practical limits, from particulates, high field Q drop, field emission, multipactor, that must be addressed.

In SCRF applications, ALD seems to offer a number of useful options. Capping or insulating layers can be applied to protect the surface from impurities coming either from the surface or from the bulk. The surfaces produced by ALD can be nano-smooth to avoid field emission, applied in protective layers to avoid quenches, chemically pure to avoid contamination or defects, applied on almost any structure, which allows design freedom, and applied *in-situ*, to avoid contamination during assembly. Since only the top few hundred monolayers of the superconductor are active parts of an RF superconductor, the ability to produce these surfaces precisely and reproducibly will be valuable. Since ALD coatings can be relevant to almost all aspects of SCRF limiting behaviour, we feel a productive experimental program on this topic can be realized [1].

Our initial effort has been directed at understanding the materials science issues associated with superconducting rf, and the chemistry of the synthesis processes required to produce an "ideal" rf surface.

**Work supported by the Office of High Energy Physics USDOE
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ALD-COATED CAVITY RESULTS

We have begun an experimental program to evaluate the promise and problems associated with this technique and have preliminary results from a number of studies.

Tests of Compatibility

This program has produced a number of significant results, both from both cavities and coupons. We will present a summary of the RF test of ALD coated cavities. Three cavities from Jefferson laboratory has been coated at Argonne National Laboratory. Alumina and Niobia with different thickness were chosen as starting materials, as proof of principle. Three cavities have been coated at Argonne, and all of them reveal the full compatibility of ALD made dielectric layers with the high field environment inside the cavities.

Figure 1 shows as an example the second ALD coated cavity at Argonne, After the first coating RF test (blue) reveal significant field emission, probably due to defective ALD layers or dust included during the ALD synthesis. A second HPR (green) shows a clear degradation of the cavity performance, indicative of an irreversible effect of the field emission on the ALD layers. After the second coating (black) the Quality factor Q increased and the Q-slope disease disappeared. This last result shows the "self-healing" mechanism of ALD, and indicates that the drastic changes in performance are confined to tenth of a nanometer on the surface of Nb cavities. Because of these results we have begun to study the effects of high pressure water rinsing on both ALD and bulk niobium surfaces.

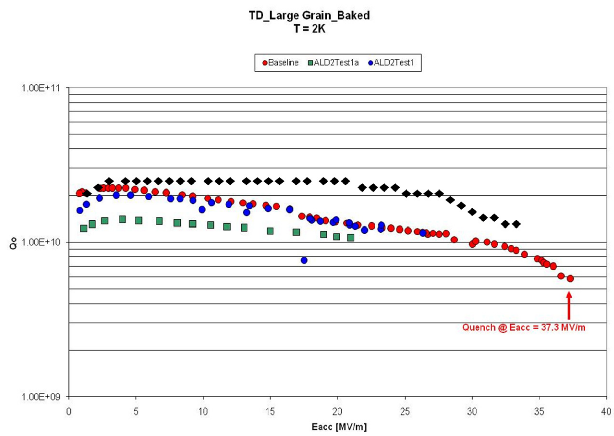

Figure 1: Tesla/ILC shape 1.3GHz cavity. RRR> 300, large grain Nb from Tokyo-Denkai. BCP and in-situ mild baked. In Bleu after the first coating with 10 nm of Al2O3 and 3 nm of Nb2O5. In green after the second HPR. In Black after the second ALD coating with 5 nm of Al2O3 and 15 nm of Nb2O5.

The parameters of the second coating were exactly the same as for the first coating, this strongly indicate that impurities introduced during the first coating were responsible for the strong field emission observed. Even if the HPR removed them, irreversible effects (cracks with sharp edges, may have damaged the ALD layers...) prevent the cavity from reaching its initial performances. The second coating didn't introduce particulates and cured the existing layers by burying them underneath the new layer.

We believe the introduction of particulates can be avoided systematically by building a cavity-dedicated ALD system in a clean room environment that would enable the control of the deposition history.

High Temperature Baking

As we showed in a previous paper, a high temperature baking on ALD coated cavity grade niobium samples improves the superconducting properties of niobium, as probed by point contact tunnelling spectroscopy, by creating an abrupt and oxide-free interface between the Nb and the Alumina. We will now present preliminary results on one of the ALD coated and baked cavity (Fig. 2). This cavity has been coated with 15 nm of alumina first, sent to J-lab HPR then tested (red curve), then a high temp baking, HPR and tested again (green curve).

The RF test after baking of cavity 1 for 20 hrs at 450 C (green) reveals a degradation of the cavity performance with a multipacting wall and strong field emission at 20 MV/m, not mitigated by successive HPR.

This multipacting surface was also present after the same high temperature baking of cavity # 3 (not shown). These results indicate that the outermost Nb_2O_5 when deposited prior to the high temperature baking is not the limiting factor but rather the Al_2O_3 layer.

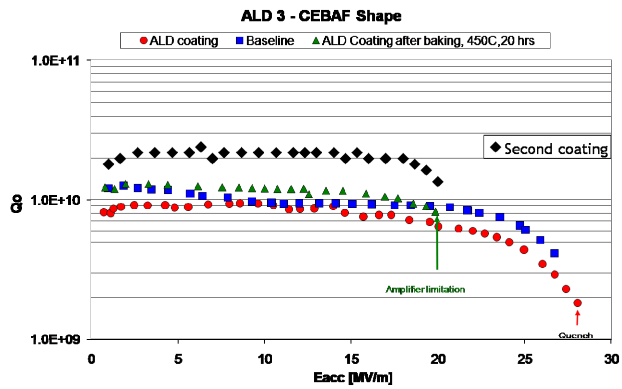

Figure 2: High temperature baking on ALD coated cavity#1. In bleu the ALD baseline, in red the RF test after the 450C-20hrs baking, in pink after the first HPR and in green after the second HPR.

We believe that cracks in the ALD layers can appear due to the temperature excursion of the baking and cooling process and changes of the structure of the native niobium oxide layer as oxygen diffuses into the bulk. These cracks will create asperities where the local field is enhanced and thus field emission points. They can also destroy the diffusion barrier properties by creating weak points through which an external source of oxygen can diffuse. HPR done after each test is the most probable source of oxygen: this hypothesis has been confirmed by complementary experiments not shown here.

These results seem to show that tests of entire cavities are more sensitive than experiments of surface superconductivity measured on cavity grade niobium coupons. Cavity tests are nonetheless a step forward in the understanding of the residual resistance of the surface layers that seems the factor preventing cavities from reaching their intrinsic limits. We believe a solution to prevent the injection of oxygen into the bulk through the cracks is to have an in-situ baking capability on the cavity dedicated ALD system. This will allow us to control the chemistry evolution of the coated surface layers and to recoat the cavity walls after the HTB without HPR, as well as the use of materials such as TiN that require higher growth temperature (>400C), and that cannot be done with the current ALD set up.

ALD NEW COMPOUNDS AND NEW POINT CONTACT RESULTS.

We have shown using Point contact tunneling spectroscopy (PCT) the influence of magnetic impurities in the niobium oxides on the superconducting properties [2]. Recent results by PCT and electron paramagnetic resonance EPR (at IIT) confirm the presence of these localized magnetic moments in the niobium oxides and quantify more precisely their amplitude and influence on the superconductivity: for instance, we showed that dissipation, or heat is generated by these moments in a RF magnetic field that could trigger pair breaking and vortex entry.

Sharp Metal/Oxide Boundaries

Since metallic niobium is highly reactive, it is always present with a coating of oxides that seem to be associated with rf losses. We have shown that it is possible to eliminate the layer of niobium oxides by "capping" niobium metal with a thin, ALD deposited alumina layer and baking the sample at a temperature high enough so that the oxides diffuse into the bulk, where the oxygen density is negligible [3]. Higher baking temperature, up to 700C was recently achieved and the XPS spectrum revealed an even more pure niobium at the interface with the Alumina. Point contact tunneling will be conducted soon on these samples.

Synthesis of Nb Metal

We have begun to synthesize Nb metal using thermal ALD. Initial tests have shown that it is possible to produce a oxide free Nb metal (Fig.3) capped by alumina or SiO_2. However these tests show a slight contamination by Fluorine and our efforts are aimed at reducing this contamination. The resistivity of these films is very close to the value of bulk niobium and the critical temperature Tc is 3.1K for a 30 nm thick film. Point contact tunneling measurements are being conducted to measure the T_c and the gap of these layers.

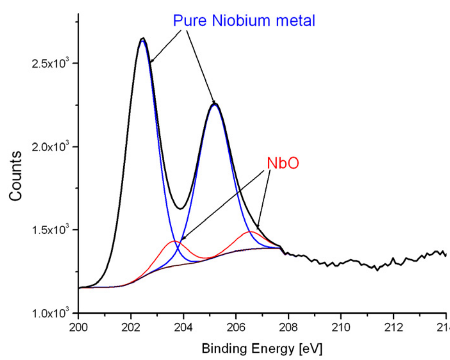

Figure 3: XPS spectrum of ALD made Niobium metal.

Synthesis of NbN

In order of make significant improvements in rf performance, it is necessary to synthesize materials other than niobium. In principle NbN, MgB_2, Nb_3Sn, Nb_3Ge, etc., can provide higher critical temperatures, and thus higher critical fields. In order to explore these options we have begun to synthesize superconducting layers of NbN. The initial attempts show a critical temperature below the best bulk measurements and we are beginning to optimize the ALD process to produce better performance. Fig.4 show the last NbN growth attempt with a new precursor.

For the best material properties (mean free path, impurities, superconducting gap...) the Plasma ALD (PEALD) technique is the way to go. PEALD is still ALD which mean conformality, atomic layer control... and **has nothing** to do with PECVD.

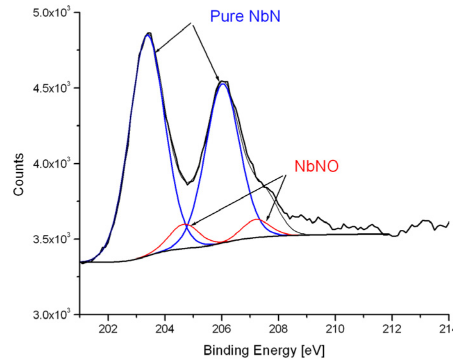

Figure 4: XPS measurements of NbN coupons.

Study of Hot Spots for Cavities

In collaboration with J-lab we begun systematic studying by point contact tunneling spectroscopy of hot and cold spots niobium samples cut from the cavity walls. We already can distinguish quite different behaviour of the superconducting and niobium oxide properties near a grain boundary and far from it on a hot spot sample. We also show that in average for hot spots samples, conductance curves taken above T_c reveal the presence of magnetic impurities in higher concentration. This study is under progress.

High Pressure Water Rinsing

In order to determine the sensitivity of ALD coatings to aggressive high pressure water rinsing, we have done tests with coupons that were coated with thin ALD layers of alumina and Nb_2O_5 of various thicknesses, and studied these with ellipsometry, atomic force microscopy (AFM) and XPS. The data show that ALD layer both Alumina (above 2 nm) and Niobia thickness and chemical composition are unchanged by a HPR for 15 s (time comparable per point to a real cavity HPR).

In order to study the damage mechanisms involved in water rinsing, we have obtained coupons heavily damaged by extended water rinsing from J-lab and have begun a program of analysis of these samples using SEM, Raman scattering, AFM and XPS. Initial results show that the composition of these oxide is a mixed of NbO_x with $2<x<2.5$, Nb_2O_5 being predominant in the first 10 nm. The depth profiling and thickness of these oxides has been studied thoroughly at IIT and Argonne.

REFERENCES

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[2] Th. Proslier et. al. Appl Phys Lett. 92 21505 (2008)

[3] Th. Proslier, et. al. Appl. Phys Lett. 93, 192504 (2008)