

DEVELOPING RF STRUCTURES USING ATOMIC LAYER DEPOSITION*

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Abstract

An effort, centered at Argonne, has started to explore the use of Atomic Layer Deposition (ALD) to study and improve the performance of superconducting rf (SRF) accelerating structures. This effort has a number of parts: a survey the properties of ALD deposited films, a study of loss mechanisms of SRF structures, and a program of coating single cell cavities, to begin to optimize the performance of complete systems. Early results have included improving the performance of individual structures and, identification of magnetic oxides as a loss mechanism in SRF. We describe the program and summarize recent progress.

MOTIVATION

Many large accelerator facilities used for nuclear physics, photon and neutron sources and medical applications are based on the technology of superconducting rf linacs. While superconducting systems constructed from bulk niobium have been very successful and are used in many major facilities, the gradient of these structures is limited by a large number of individual mechanisms. At high gradients the Q can drop causing increased power consumption, for example, there can be field emission causing local hot spots, the Maxwell stress can cause deformation of the structure distorting the structure so it loses resonance, the cavity can quench due to a variety of mechanisms.

We are beginning to explore the use of thin films produced by Atomic Layer Deposition as an alternative mechanism for producing SRF linac structures. If these films can be made pure, conformal and with good superconducting properties, and can be uniformly deposited over large areas they seem to offer the possibility of addressing many SRF concerns, in addition to eliminating the requirements for large masses of niobium. In principle, the need to provide only a few London penetration depths ($\sim 40\text{nm}$) of material should loosen many mechanical, thermal and cost constraints in the design of these structures [1].

The proposal by Alex Gurevich to consider the use of layered superconductors also seems to open the possibility of using a variety of new materials, and operating at higher fields with lower losses, and lower

construction and operating costs. The layered structures require the ability to produce thin ($\sim 40\text{ nm}$) superconducting layers separated by equally thin insulating layers. This option seems to require a level of uniformity and precision that only ALD can provide [1].

We have coated a number of cavities with ALD films and with the general conclusion that the films are compatible with surface fields on the order of 70 – 80 MV/m, and seem to be mechanically robust enough to survive high pressure water rinsing and the thermal excursions required for rf operation at 2°K .

Our program has a number of goals: 1) surveying possible materials that could enhance SRF performance, 2) understanding loss / failure mechanisms in existing structures in order to suppress them, 3) demonstrating the viability of ALD technology in real systems, and 4) optimizing their rf performance.

SUPERCONDUCTING ALD FILMS

Ideally layered structures should require a material with a high H_{c1} for a substrate and a material with high H_{c2} for the layers. We have been surveying the superconducting properties of the large number of films that can be synthesized using ALD, beginning with niobium compounds. We have produced superconducting carbides, silicides and nitrides under a variety of conditions and stoichiometries using different precursors. The ALD process utilizes gaseous precursors, introduced into the vacuum chamber. An example is shown in Fig 1, where the sequential introduction of NbF_5 and trimethyl-aluminum gasses are used to grow NbC films.

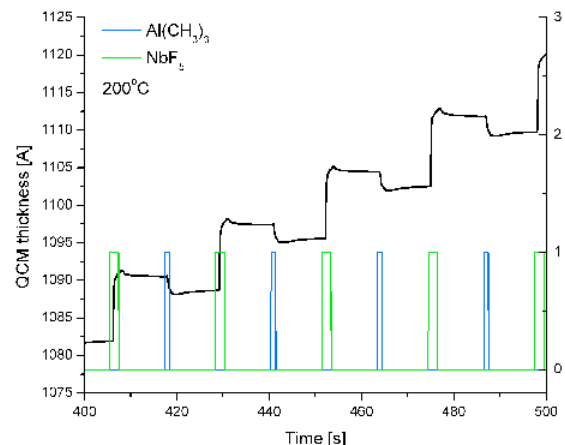


Figure 1: Quartz crystal microbalance measurements of ALD growth of NbC films.

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Preliminary results show that a wide variety of films have superconducting properties, with the best films showing T_c values equal to, and in some cases, greater than those expected from comparable thicknesses of good RRR niobium. So far, all ALD films have been produced using standard, thermal ALD. We are now beginning a program using plasma or radical enhanced ALD using a Cambridge Nanotech Fiji, PEALD system. It has been found that atomic and molecular radicals are able to enhance the ALD process by allowing reactions to proceed at lower temperatures, producing more dense films that have better metallic properties.

LOSSES DUE TO MAGNETIC OXIDES

The basic loss mechanisms in superconducting rf systems have not been not well understood. Recent effort has been devoted to understanding the nature of high field Q slope assuming magnetic impurities in the niobium oxide layer. The standard “cure” for this phenomenon is low temperature baking.

Because of the reactivity of niobium atoms, there are many stable niobium oxides, which have a variety of electrical and magnetic properties. Some oxides have been known to have magnetic properties for many years, and because of the strength of the magnetic interaction, these oxides can break up the Cooper pairs in superconducting currents and produce SRF losses [2].

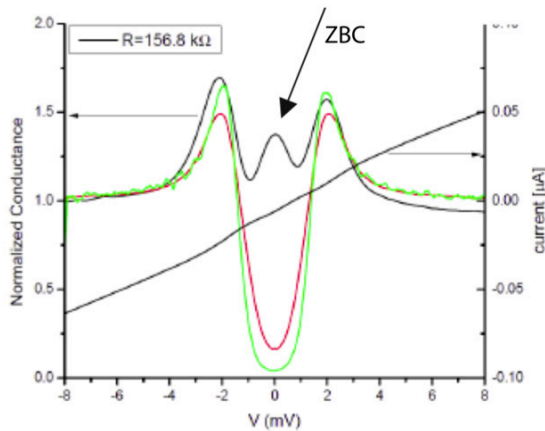


Figure 2: Point Contact Tunneling data showing the Zero Bias Conductance (ZBC) at hot spots [3].

Magnetic oxides were first detected using Point Contact Tunneling, (PCT). From the shape of the conductance vs. bias it is possible to identify the presence of magnetic oxides described by the Shiba model and reject a number of competing processes such as inelastic tunnelling, strong coupling effect or proximity effects. Experiments have shown that low temperature baking reduced the zero bias conductance, in agreement with measurements in rf structures. The effect of mild baking seems to be to slightly alter the stoichiometry of the boundary between Nb_2O_5 , containing the magnetic oxides, and the NbO and NbO_2 layers, enabling them to better shield the magnetic oxides from the superconducting currents.

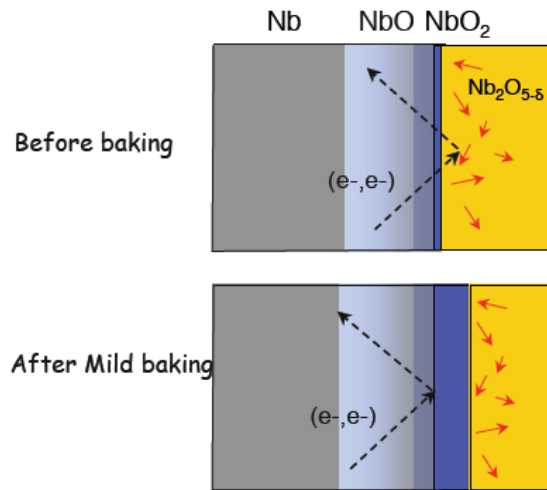


Figure 3: Effect of low temperature bake on oxides.

The presence of magnetic impurities in native niobium oxides has been confirmed by PCT spectroscopy, SQUID magnetometry and Electron Paramagnetic Resonance (EPR). All niobium measurements displayed a small impurity contribution to the magnetic susceptibility at low temperatures which exhibited Curie-Weiss behavior, indicative of weakly coupled localized paramagnetic moments. By examining Nb samples with widely varying surface-to-volume ratios (rods, foils, wires, powders) it was found that the impurity contribution is correlated with surface area. PCT tunneling measurements which utilize the native oxide layers as barriers exhibit a zero-bias conductance peak which splits in a magnetic field $> 4T$, consistent with the Appelbaum-Anderson model [1] for spin flip tunneling. Viewed together, the experiments strongly suggest that the native oxides of Nb are intrinsically defective, and consistently exhibit localized paramagnetic moments, likely caused by oxygen vacancies in Nb_2O_5 .

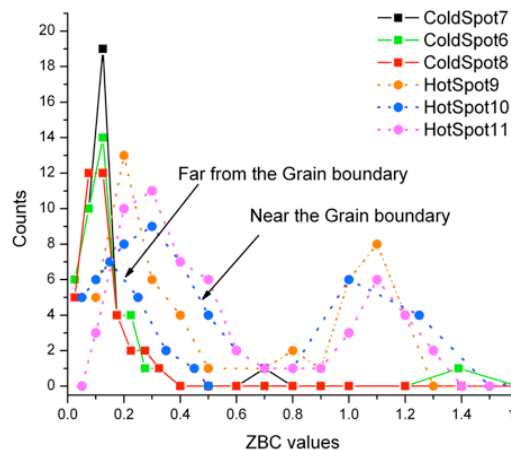


Figure 4: ZBC measurements of hot and cold spots in a working cavity [3].

A number of PCT measurements of sections of rf cavities known to produce abnormal losses during high power tests were made in order to try to identify differences in the structure of the superconductor. Six samples were used, three from identified hot spots and three from areas with normal losses. It was found that the regions with hot spots showed significantly higher Zero Bias Conductance (ZBC), consistent with the presence of magnetic impurities [3]. The data in Fig 4, shows a correlation between local rf loss mechanisms and the local concentration of magnetic impurities consistent with magnetic impurities as the primary loss mechanism.

RESIDUAL RESISTANCE

The residual resistance of real superconductors can be modeled assuming that the surface impedance of magnetic impurities is the primary perturbation on the BCS theory. The computation of the surface impedance (R_s) in presence of magnetic impurities in the Shiba approximation reveals the saturation at low temperature of R_s , suggesting that magnetic impurities are responsible for the so-called residual resistance. These properties may have an impact on Nb based superconducting devices and shine a new light on the origin of the paramagnetic Meissner effect (PME) [4].

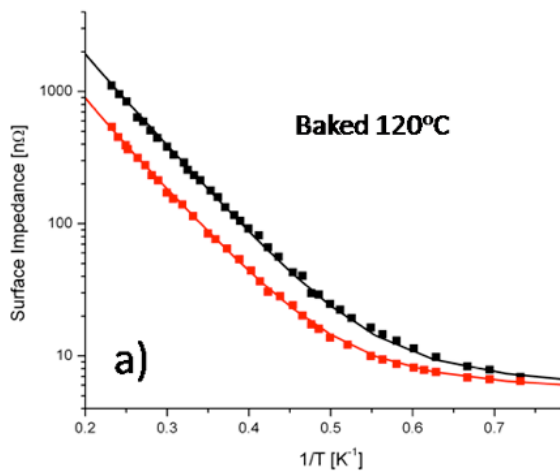


Figure 5: The effect of low temperature baking on the residual resistance of niobium (red before, black after) showing the effects.

CAVITY COATING TESTS

In the past we have coated a number of niobium single cell cavities with a variety of films to determine if the ALD technology might be useful high gradient SRF environment. These tests have been successful, showing that the ALD films are compatible with surface fields on the order of 75 MV/m, water rinsing and with thermal excursions from 2 – 700^oK, in some cases improving the Q, in other cases the performance at maximum gradient.

FIELD EMISSION

Field emission can be a problem for both normal and superconducting structures. In both cases this process seems to be due to small geometrical asperities that cause local fields can be in the range of 5 – 10 GV/m. In warm cavities these asperities can be a source of breakdown, and in SRF structures they cause field emission that can limit the ultimate gradient.

We have been exploring the ability of ALD films to conformally coat these asperities in warm cavities and will be doing an experiment using warm cavities to see if conducting coatings can eliminate active asperities and the problem of field emission in all cavities.

SUMMARY

We are studying the applicability of Atomic Layer Deposition (ALD) to SRF systems. We have begun a survey of the superconducting properties of materials deposited using thermal ALD using a variety of precursors, stoichiometries, temperatures and thicknesses. This work will continue using plasma enhanced ALD.

Studies of losses in SRF materials strongly suggest that the native oxides of Nb are intrinsically defective, and consistently exhibit localized paramagnetic moments, likely caused by oxygen vacancies in Nb₂O₅. The computation of the surface impedance (R_s) in the presence of these magnetic impurities in the Shiba approximation reveals the saturation at low temperature of R_s , suggesting that magnetic impurities are responsible for the so-called residual resistance.

We will continue cavity coating tests on SRF structures and also warm cavities in order to optimize their performance.

ACKNOWLEDGEMENTS

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REFERENCES

- [1] T. Proslie, Y. Ha, J. Zasadzinski, G. Ciovati, P. Kneisel, C. Reece, R. Rimmer, A. Gurevich, L. Cooley, G. Wu, M. Pellin, J. Norem, J. Elam, C. Antoine, Proc. of PAC09, Vancouver, CA. May 4-8, (2009)
- [2] T. Proslie, J. Zasadzinski, L. Cooley, C. Antoine, J. Moore, J. Norem, M. Pellin, K. E. Gray, App. Phys. Let. **92**, 212505 (2008)
- [3] Th. Proslie, J. Norem, J. Elam, M. Pellin, J. Zasadzinski, G. Ciovati, Proc. of SRF2009, Berlin Germany, Sept. 20 – 25 (2009)
- [4] Th. Proslie, M. Kharitonov, M. Pellin, J. Zasadzinski, G. Ciovati, Proc. of ASC2010, Washington D. C. Aug. 1-6 (2010)