PROGRESS REPORT
Project Name: "Investigation of Plasma Etching for Superconducting RF Cavities Surface Preparation"

Personnel and Institution Requesting Funding
L. Vušković, Old Dominion University
S. Popović, Old Dominion University
M. Rasković, Old Dominion University
Collaborators
A.–M. Valente, Thomas Jefferson National Accelerator Facility
L. Phillips, Thomas Jefferson National Accelerator Facility
Project Leader
Leposava Vušković
Vuskovic@physics.odu.edu

Project Overview

Preparation of cavity walls has been one of the major problems in superconducting radio-frequency (SRF) accelerator technology. Accelerator performance depends directly on the physical and chemical characteristics at the SRF cavity surface. The primary objective of this project is to develop a cavity surface preparation process which is superior in terms of cost, performance, and safety to the wet chemical processes currently in use. Plasma based processes provide an excellent opportunity to achieve these goals and, in addition, offer a unique opportunity to provide an immunity to subsequent exposure to atmosphere through plasma passivation processes while the etched surface is still under vacuum and in an oxide free state. Additionally, a plasma etching process may consist of different sequential etches in the same process for the purpose of providing a fast niobium removal of many tens of microns if required, followed by a slower etch tailored to provide a finished surface optimized for the best RF performance. This can be accomplished by applying sequentially the effects of various types of electric discharge plasmas to minimize surface roughness and eliminate or minimize deterioration of cavity performance by oxygen, hydrogen and other chemical contaminants.

In the first and second year of the project, we have built and operated two different electric discharge systems where we have been investigating effects of various plasma regimes on surface roughness, and the efficiency of electric discharges in eliminating the adverse effects of oxides and hydrides on the cavity performance. The results on the proof-of-principle tests on bulk Nb discs were published in a peer reviewed journal [1] and several conference proceedings [2-4]. The analysis of processed samples was focused on surface roughness, where the comparison with conventional buffered chemical polishing (BCP) was readily available. The obtained results are very promising, since the electric discharge plasma etching has proven to be superior to BCP both in the size of features and sharpness of the boundaries between individual features at the surface. Analysis performed with various surface characterization techniques show a consistent tendency of the electric discharge plasmas to increase etching rates in the regions of enhanced electric field. This natural smoothing behavior leads to oblique shapes of the domains and elimination of sharp edges. The Surface composition of the sample is not covered in this report, since the surface topology was our priority in this phase of work.

Based on promising results of plasma etching on the disc shaped samples, we designed a single-cell plasma-etching apparatus, which is being constructed and tested in the third year of this project. This system is a barrel type reactor with a flexible design to accommodate pulsed D.C. or continuous microwave (MW) power input. The SRF cavity will be the grounded electrode/conductor and the discharge is designed to operate in the sheath mode so it follows the curvature of the cavity.

Obtained results make us confident that this project, upon completion, will provide a very valuable alternative to current SRF cavity surface processing technology.

Progress Report

1. Introduction

The primary objective of the present research is to minimize surface roughness and the presence of Nb-oxides and other contaminants that result in the degradation of SRF cavity characteristics. The experiment is devoted to the use of metal-to-discharge interfaces to generate an adequate environment for transformation of Nb oxides or other impurities from the surface of SRF cavity into removable volatile compounds. The gas mixtures employed for the surface treatment (BF_3/Ar and Cl_2/Ar) contain reactive gases that are toxic and/or corrosive, but concentrations used in plasma etching processes are much smaller than acid concentrations used in BCP or electrochemical (EP) polishing [5] and are easily eliminated by automatic scrubbing.

2. BF_3 reactive etching

In the first year, we exposed a limited number of Nb disc shape samples to reactive-ion surface modification in a repetitively pulsed D.C. diode system in order to explore the possibilities of developing the new technique [1]. This reactor is designed and operated by Varian Inc. as a doping system. The principle of operation and diagnostics are described in more details in [6]. In the present study, this system has been used for proof-of principle demonstrations of bulk Nb plasma etching. The applied operational mode was the sheath mode, in which the sample is connected to the cathode. The sheath mode enhances the physical sputtering of surface over the chemical etching. Process control in the plasma-etching system consisted of plasma diagnostics and residual gas analysis. More details about the equipment used and the experimental conditions during the samples' exposure can be found in [1].

All samples were transported to the diagnostic system exposed to room temperature air. The plasma-processed samples were compared with (*i*) unprocessed samples, and (*ii*) BCP processed samples by the means of several surface characterizing techniques. We used the scanning electron microscope, digital video-microscope, profilometer, and scanning probe microscope for the inspection of the sample surface topology, as in this phase of work we were focused on surface morphology.

The preliminary results of this study show that plasma-treated samples are comparable or superior to a BCP sample, both in the size of features and sharpness of the boundaries between individual features at the surface. Images obtained with the optical microscope, the scanning probe microscope, and the scanning electron microscope show the consistent tendency of electric discharge plasmas to increase etching rates in the regions of enhanced electric field. This natural smoothing behavior leads to oblique shapes of the features and elimination of sharp edges.

Additional sets of experiments are being planed for the second year to determine the contribution of physical sputtering relative to chemical etching in sheath operating mode conditions. Prior to exposure all samples' surfaces were characterized in regard of morphology (SEM, profilometry, AFM, SFEM) and composition (XPS, SIMS). Exposure is being done with pure Ar or pure BF_3 gas under the same operating plasma conditions and the influence on the surface roughness and composition is being investigated.

3. Cl_2 reactive etching

At the beginning of the second year, the MW reactor was constructed and used to plasma etch 1"- diameter samples. Our microwave cavity discharge, described in [2], is a typical "barrel" reactor [7] capable of plasma etching at pressures up to 1 Torr. Compared to an RF discharge, a microwave discharge has a more efficient transfer of energy from the microwave electric field to the gas and has, as a consequence, higher electron densities and higher radical densities than RF sources used for plasma etching of Nb thin films [7]. These plasma conditions are more favorable for plasma etching than for sputtering processes. Also, the higher gas temperature in the plasma contributes to a higher rate of chemical reaction and vaporization of products of chemical reaction. The samples are placed on ceramic holders in the central part of the reaction chamber. The reactive gas mixture containing Cl_2 and Ar is introduced into the reaction chamber. High constant flow rates are necessary so that the reactive species depleted in the chemical reaction can be replenished and the products of the chemical reaction removed from the sample. The reactor allows for the option of biasing the sample. Biasing the sample can increase the small value of the sheath potential. In both experimental systems, exhaust gas purifiers ("scrubbers") have been applied.

Flat sample treatment and analysis is an important subject for the second year activity. The first group of activities is to optimize the plasma etching process with respect to the surface roughness and to eliminate sharp edges that could contribute to field emission in SRF cavities. The parameters that are being optimized include source power, gas flow, pressure, bias potential, and exposure time. The optical method for etching rate detection is being developed. This will be followed by the analysis and treatment of the surface contaminants. This activity will include a search for plasma composition (reactants and radicals) and conditions that provide the minimal formation of oxygen and hydrogen compounds on the surface and minimize their diffusion through the bulk of Nb.

4. Kinetic model for plasma treatment

A better knowledge of the reaction mechanisms of Nb plasma etching would facilitate a better control of the process and would lead to better etch rates with smaller energy and feed gas consumption. Therefore, we have initiated the development of a comprehensive kinetic model for plasma-surface interaction in order to complement process diagnostics and enhance its interpretation [2, 8]. Challenges facing this activity are multifold. The necessary cross sections and rate coefficients for electron-impact ionization of Nb based molecules are not available in the literature. We calculated these data by employing *ab initio* quantum chemistry models and semi-empirical approximations, such as Binary-Encounter-Bethe model [9], for cross-section calculations. The validation of the obtained data is limited by the lack of experiments in this area, and by the limited number of basis sets available for the description of transition metal atoms. All calculated data for elementary collision processes are currently being implemented in the plasma kinetic model. Available mass spectrometry and emission spectroscopy data were used for the validation of the assumed reaction mechanism. Obtained results are in preparation for publication.

5. Concluding remarks and future work

In the course of this project, we have demonstrated that plasma etching of Nb surfaces is a viable alternative to the currently used technology. We anticipate its use as either a substitutive or a complementary process, which would minimize the residual roughness, provide surface cleaning, and eliminate the adverse effects of oxide interfaces. An important result is that the plasma technology is compatible with the SRF cavity geometry regardless of its complexity. We have constructed a barrel type plasma reactor that uses the existing reference cavity and an umbrella electrode system that will provide plasma sheath structure, which follows the curvature of the cavity (See Figure 1). Using a dc bias, the sheath voltage can be varied to provide enough energy for the reactive etching ions.

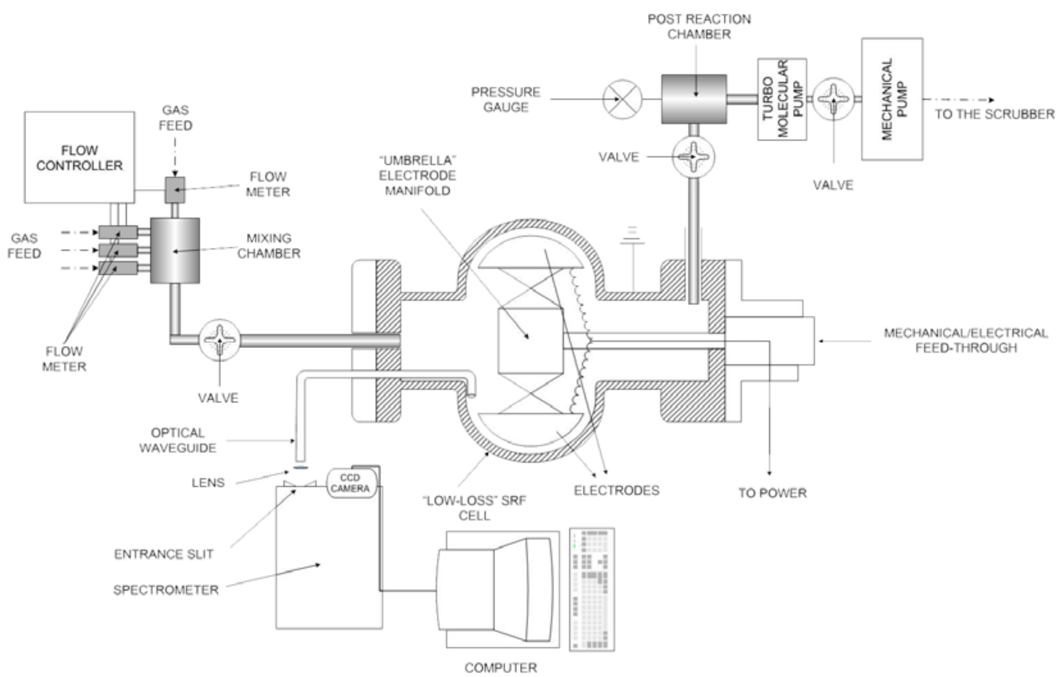

Figure 1. Schematic diagram of the SRF cell plasma etching reactor.

Using both constructed plasma reactors for cavities and disc-shaped samples, the future exploratory work, planned for the next funding period, will focus on enhancing the oxygen transport out of the bulk Nb. For this purpose the currently used, relatively simple, gas mixtures will be complemented with nitrogen or carbon-based gases to increase the oxygen reduction on the surface and enhance oxygen transport from the plasma.

All plasma mixtures that are currently being used are oxygen-free and the plasma treatment can provide the indirect resolution of the current dilemma about the origin of the oxygen in niobium – whether it is interstitial, or process related. For this purpose sample analysis to determine the dynamics of oxygen transport will implement more sophisticated techniques, such as XPS or SIMS with the aim to determine the difference (if any) in oxygen transport after wet (BCP) and dry (plasma) etching processes. In order to obtain the full insight of complex chemical dynamics at the surface and near-surface interfaces between various oxide phases, plasma-chemical kinetic and oxygen transport simulation models will be developed.

Plasma technology has its own complexity and the optimization of the process will require studies of geometry and time sequencing. An ultimate goal in this sub-task is to assure permanent and stable chemical and structural characteristics of the surface, which is believed to be one of the crucial requirements for adequate performance of the cavities.

6. References

[1] M. Raskovic, et al., Nuclear Instruments and Methods in Physics Research A 569 663–670 (2006).
[2] L. Vušković, et al., , Plasma Treatment of Bulk Niobium Surface for SRF Cavities, EPAC, June 26-30, 2006, Edinburgh, Scotland, Proceedings, pp. 493-495.
[3] M. Rašković, et al., Plasma Treatment of Bulk Niobium Surface, 59th Annual Gaseous Electronic Conference, October 10-13, 2006, Columbus, Ohio pp. 20.
[4] M. Rašković, et al., The Niobium Surface Modification in Microwave, 58th Annual Gaseous Electronic Conference, October 16 – 20 2005, San Jose, California pp. 20.
[5] K. R. Williams, K. Gupta, and M. Wasilik, J. Microelectromech. Syst. **12**, 761 (2003).
[6] S. Radovanov, et al., J. Appl. Phys. **98**, 113307 (2005).
[7] B. Chapman, *Glow Discharge Processes – Sputtering and Plasma Etching*, John Willey&Sons, New York (1980).
[8] M. Rašković, et al., Electron-impact Ionization Rates of Niobium Oxides, DAMOP Meeting 2006, May 16-20 2006, Knoxville, Tennessee pp.92.
[9] W. Hwang, et al., J.Chem.Physics. **104**, 2956 (1996).

Final Year Project Activities and deliverables

Project activities in the third year will include testing and optimization of the single-cell cavity plasma etching reactor. It is a barrel-type reactor, which operates in the sheath mode. Therefore, the outside conductor is the cavity itself, which is grounded and acts as a cathode in the pulsed d.c. operation. In microwave operation, the system acts as a coaxial waveguide. In both cases, the sheath follows the curvature of the inner surface of the cavity cell, and the bulk plasma region is wide enough so that the shape of inner conductor (or anode) is irrelevant. It is fed electrically through an isolated feedthrough. A reactant gas mixture flows in and out through separate ducts in the flanges. The beam pipes of the cavity are etched in an additional step using the same coaxial principle, but with a shorter inner conductor.

In this phase, the optimum plasma conditions will be determined. A large part of the third year will be devoted to the characterization of the RF properties and measurements of power losses in the single cell treated with the electric discharge plasma. Finally, the project will conclude with the proposed scenario for plasma treatment and passivation of the cavity inner surface to prevent oxidation after treatment.

Results achieved in this phase will be reported in a mid-year progress report. At the end of the project's third year a final report will be delivered.