

PLASMA ETCHING OF A SINGLE-CELL RF CAVITY - ASYMMETRIC ELECTRONEGATIVE DISCHARGE*

J. Upadhyay[#], M. Rašković, L. Vušković, S. Popović

Old Dominion University, Department of Physics, Norfolk, VA, USA

L. Phillips, A.-M. Valente-Feliciano

Thomas Jefferson National Accelerator Facility, Newport News, VA, USA

Abstract

We are pursuing the use of environmentally friendly dry etching of SRF cavity in Ar/Cl₂ discharge. We have successfully demonstrated on flat samples that in a barrel-type reactor etching rates are comparable to the wet process, such as BCP or EP. The geometry of SRF cavities made of bulk Nb defines the use of asymmetric RF discharge configuration for plasma etching. The asymmetry in the surface area of a driven and grounded electrode creates a difference in the voltage drop over the plasma sheath attached to the driven electrode and the plasma sheath attached to the cavity surface. A specially designed single cell cavity is used to study these asymmetric discharges which contain 20 sample holder holes symmetrically placed over the cell. These sample holder holes can be used for both diagnostics and sample etching purposes. The approach is to combine radially and spectrally resolved profiles of optical intensity of the discharge with direct etched surface diagnostics to obtain an optimum combination of etching rates, roughness and homogeneity in a variety of discharge types, conditions and sequences.

INTRODUCTION

To achieve theoretically predicted values of accelerating fields, the surface of cavities must be prepared by a process that decreases surface roughness, produces surfaces with less prominent grain boundaries, and does not introduce additional impurities in the bulk of Nb. Plasma-based surface modification provides an excellent opportunity to achieve these goals. It is a crucial technology in the development of semiconductor circuit elements, and it has been applied in preparation of superconducting devices. However, it has not been considered so far as a viable alternative to the existing cost-intensive and environmentally unfriendly (liquid) acid-based technology.

Plasma etching in various combinations and geometries at low pressure has been extensively used in semiconductor material processing and fabrication of microelectronic devices [1]. Plasma-assisted etch processes can be highly selective with respect to direction and hence indispensable in patterned removal of surface material or in removal of material from non-flat surfaces. The etching process is anisotropic, and horizontal etch is easily controlled at zero level, which

leads to a homogeneous etching rate over wide surface dimensions.

Recently, plasma etching has been developed as the technology for removal of transuranic waste from contaminated sites [2]. In this case the use of non-thermal atmospheric pressure plasma allows in-situ exposure of the contaminated object, without building a special vacuum chamber for each site.

Plasma etching of Nb thin films is commonly used for production of Josephson junctions [3-19]. However, there is no data on plasma modification of bulk Nb surfaces where a high etching rate is critically required, in contrast to thin film plasma applications. The use of reactive gases containing fluorine or chlorine atoms was a natural choice for the plasma treatment of Nb, since Nb halogenides have a high vapour pressure and low boiling temperature [20]. All experiments on thin films were performed using RF discharges at low or moderate pressures. Depending on experimental conditions, etching rates vary from few nm/min in case of the physical sputtering process to a few hundred nm/min during reactive ion etching. The low etching rates are favourable for anisotropic etching and induce better etch pattern transfer, necessary for junction applications. Comparably higher etching rates were achieved using reactive gasses containing Cl atoms [6, 7, 10, 18, 19]. An etching rate of the order of $\mu\text{m}/\text{min}$ was reported in only one case [19].

Current understanding based on the analysis of cavity performances after wet acid etching [21] is that maximum gradients are obtained after removal of 100-250 μm of the bulk Nb surface that are mechanically damaged due to the Nb sheet manufacturing process. Without going into the argument that this depth is three orders of magnitude higher than the microwave skin depth and probably an order of magnitude larger than the residuals of the manufacturing process, we have adopted the same layer removal as the primary objective of the plasma etching process in our work, which we achieved successfully working with flat samples [22]. We have shown that etching rates of bulk Nb as high as 1.5 $\mu\text{m}/\text{min}$ can be achieved in a microwave glow discharge using Cl₂ as the reactive gas (See the schematic diagram in Figure 1). This is as a very promising result indicating that plasma etching could be a viable alternative to the environmentally problematic acid etching of SRF cavities due to the use of hydrofluoric acid in the chemical bath. A suggested process at high pressure (~ 1 Torr) and moderate input power density (~ 2 W/cm³) has proven to be efficient enough to be employed for accelerator cavities.

* Authored by Jefferson Science Associates, LLC under U.S. DOE Contract No. DE-AC05-06OR23177. The U.S. Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce this manuscript for U.S. Government purposes.

[#] valente@jlab.gov

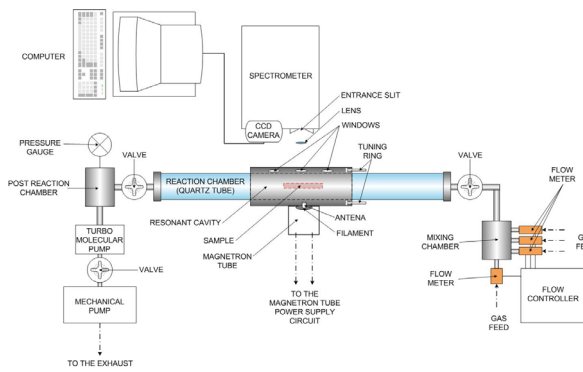


Figure 1: Schematic diagram of experimental setup for flat sample etching.

Based on the flat sample etching experience, we can define the chlorine budget in processing SRF cavities. We could calculate the usage of chlorine at 8.6 g per 1 g of etched Nb layer. By scaling, a seven cell cavity would require about 10 kg of Cl, which is transformed into environmentally friendly Cl compounds in the scrubber wherefrom they could be easily removed.

STANDARD CELL PROCESSING

Plasma etching in single cell cavity and the RF performance of this cavity has to be tested by vertical testing method. We used a bell jar vacuum system (See Fig. 2) which has completely covered the SRF Nb single cell cavity. Discharge was generated inside the cell, in two regimes: (a) High Voltage (HV) (2 kV) a.c. discharge with an Nb rod as the powered electrode; (b) microwave (MW) discharge excited by the magnetron antenna inside the resonant cavity. The plasma sheath was homogeneously distributed over the inner surface of the cavity in both cases (See Figure 3). However, the estimated sheath voltage was of the order of 500 V in the case of the a.c. discharge, and not more than 50 V in the case of the MW cavity discharge.



Figure 2: Single cell experimental set up.

A specially designed diagnostic cell has been used for preliminary testing on homogeneity of plasma and surface processing performance. The cell has a set of 20 sample holder holes that can be used as plasma observation windows or small sample holders for etching tests. After completion of the tests with the diagnostic cell, a set of standard single cells will be prepared for vertical RF performance testing.

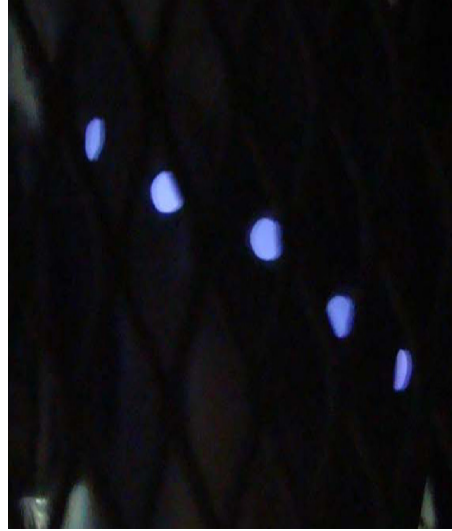


Figure 3: Plasma through the cavity holes.

CHARACTERISTICS AND DIAGNOSTICS OF PLASMA DISCHARGE

Asymmetric Discharge

Due to the differences in the surface area of driven and grounded electrodes, there is an asymmetry in the voltage between the plasma and the driven electrode and between the plasma and the grounded electrode. It can be easily shown through the capacitor model [1] (See Figure 4).

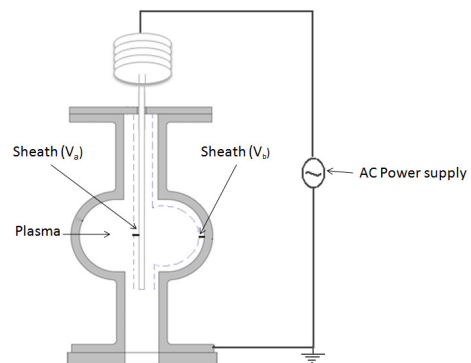


Figure 4: Scheme of asymmetric discharge.

As smaller area has a smaller capacitance so it has a larger voltage drop. On the other hand, the larger area

electrode has a large capacitance and a small voltage drop. The scaling of the voltage drop in the plasma sheath with the surface area of electrode is

$$\frac{V_a}{V_b} = \left(\frac{A_b}{A_a} \right)^{5/2}$$

As the ion bombarding energy is the sum of total energy gained by the sheath potential and the energy ion already had before entering into the sheath. The ion energy does not just depend on the electron temperature but it also depends on the source geometry and the application of a bias voltage. So the ion kinetic energy lost at the surface is [1]

$$E_i = V_s + \frac{T_e}{2}$$

where
$$V_s = \frac{T_e}{2} \ln \left(\frac{M}{2\pi m} \right),$$

Where T_e is the electron temperature, M and m are the ion and electron mass, respectively. We designed our electrode in such a way that sheath voltage V_s is approximately 10 V at the grounded electrode sheath to ensure an efficient etching of SRF cavity surface, which is grounded in our case.

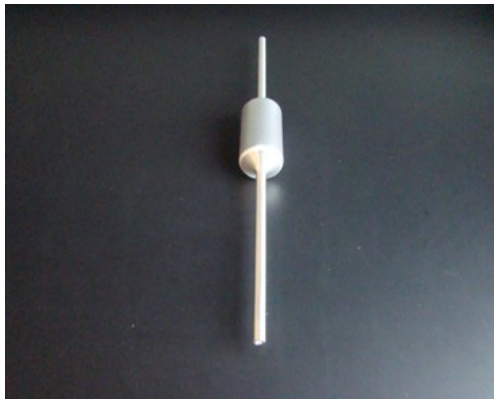


Figure 5: Specially designed electrode.

Fibre Optics Diagnostic System

To verify the non-uniformity of the plasma in the cavity, a fibre optic diagnostic system is developed. The driven electrode was 2.6 mm in diameter 17.2 cm long niobium rod and the cavity was grounded. An optical fibre of 1 mm diameter was placed with the help of a feed through at the middle hole in the equatorial region. The optical intensity passing through the fibre is measured at the other end with the help of a photomultiplier tube which was attached to an oscilloscope. The photomultiplier tube was powered through a high voltage power supply. The schematic of the experiment is represented in figure 6.

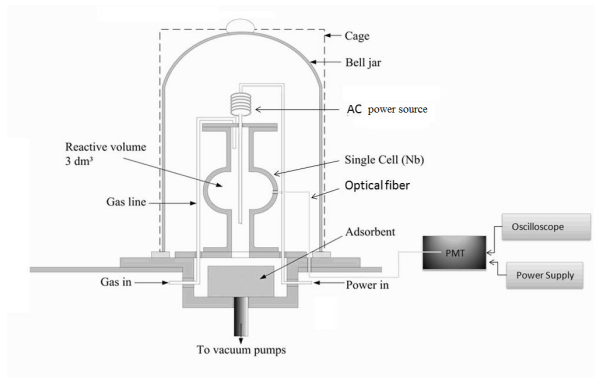


Figure 6: Schematic of the optical fiber experiment.

The vacuum in the bell jar chamber was obtained with a turbo pump coupled with a roughing pump. The base vacuum obtained was 5.5×10^{-5} mbar. For this preliminary experiment we maintained 1×10^{-1} mbar pressure with 145.5 sccm gas flow of argon with the help of mass flow controller. The effective voltage at the driven electrode was varied from 400 V to 700 V. We took the measurements at the different positions from the cavity wall representing the two sheaths. The distance versus spectral line intensity profile at fixed effective voltage of 600 V is presented in Figure 7.

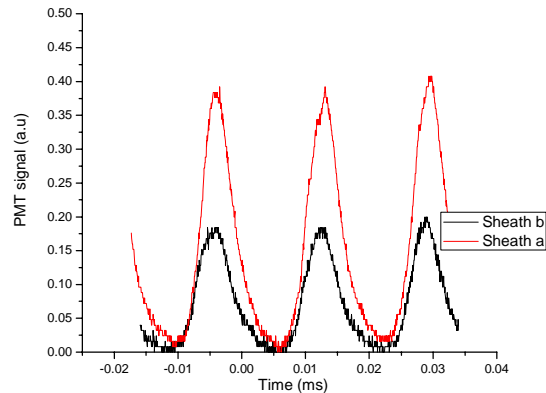


Figure 7: Time waveforms of spectral line intensity at both sheaths in the discharge.

There is a qualitative relationship between the intensity of Ar spectral lines and the sheath voltage, which is useful for illustration of the sheath distribution in the asymmetric discharge. As shown in Fig. 6, the emission from the Ar spectral line is much stronger near the driven electrode.

Relative concentrations of reactive species in the gas mixtures were monitored using emission spectroscopy. The emission from the discharge directed through a lens on the entrance slit of the imaging spectrometer. A CCD camera was connected to the spectrograph to record simultaneously the spectral line intensities from chlorine radicals and from argon buffer. The concentrations of molecular Cl_2 under different experimental conditions can be connected to relative intensity of emission of electron

impact excitation of Cl₂ at 306 nm. The relative intensity of the atomic (Cl I) and the ionic (Cl II) form of Cl was recorded in the spectral region 465 - 485 nm together with the atomic (Ar I) and the ionic (Ar II) carrier gas emission. Data from these spectra can be used for derivation of degree of dissociation of chlorine in the plasma and for correlation with etching rate in order to determine the dominant mechanism of etching [22, 24].

CONCLUSION

In view of the relatively complex technological challenges facing the development of plasma-assisted surface treatment, we have adopted a three-step approach:

1. Step one: Work with flat samples, with the objective to fulfil the requirements for etching rates, surface roughness, and to demonstrate environmentally friendly and cost-reducing aspect of the plasma-assisted process.
2. Step two: Work with a single-cell cavity to establish optimum conditions for an asymmetric electronegative discharge in cavity geometry, to demonstrate the uniformity of surface treatment, and to perform the RF performance test compatible with existing standards.
3. Step three: Work with multiple-cell cavities to demonstrate final performance of the process, to establish treatment protocol, and to define the process monitoring procedure.

Based on highly encouraging results with flat Nb samples and on a relatively straightforward transition to the cavity wall processing, we can state confidently that efficient plasma etching of Nb superconductive RF cavities can be developed into a low-cost, environment-friendly technology to replace the “wet process”, which uses a strong mixture of acids, including HF. This paper describes the current status and issues of the second step - to prove comparable RF performance of cavities etched with “dry” (plasma) process by etching the standard (“low-loss”) 1.5 GHz single-cell cavities, which would be RF tested. The RF performance is the single feature that remains to be compared to the “wet” process, since all other characteristics of the “dry” technology, such as etching rates, surface roughness, low cost, and non-HF feature, have been demonstrated as superior or comparable to the currently used technologies.

The main issue is that the geometry of the inner surfaces of cavity implies that the plasma discharge has to be asymmetric, with much higher sheath voltage at the driven electrode. This is in contrast to the usual parallel-plate electrode configuration of thin film wafer treatment. In order for the asymmetric discharge to be effective, the lower sheath voltage at the treated surface (large area, undriven electrode) has to be at least equal or higher to the plasma floating potential at every point of the surface. When this condition is satisfied, one should expect a uniform etching and a satisfactory global RF performance of the cavity.

ACKNOWLEDGMENT

This work is supported by the NSF/DOE collaborative effort through the Office of High Energy Physics, Office of Science, Department of Energy under Grant No. DE-FG02-05ER41396. Thomas Jefferson National Accelerator Facility, Accelerator Division supports M. Rašković and J. Upadhyay through fellowship. We thank the Surface Characterization Lab, College of William and Mary Applied Research Center, and the ODU Applied Research Center for the use of various diagnostics equipments.

REFERENCES

- [1] A. Lieberman and A. J. Lichtenberg, *Principles of Plasma Discharges and Materials Processing* (John Wiley & Sons, New Jersey, 2005).
- [2] X. Yang *et al.*, J. Nuclear Materials, 324, 134 (2004).
- [3] M. M. Chen and R. H. Wang, J. Vac. Sci. Technol. A 1, 708 (1983)..
- [4] M. M. Chen and Y. H. Lee, J. Electrochem. Soc. 131, 2118 (1984).
- [5] J. N. Sasserath and J. Vivalda, J. Vac. Sci. Technol. An 8, 3914 (1990).
- [6] J. N. Sasserath, J. Appl. Phys. 68, 5324 (1990).
- [7] T. Harada, K. Gamo, and S. Namba, Jpn. J. Appl. Phys. 20, 259 (1981).
- [8] T. T. Piotrowski, A. Pitowska, E. Kaminska, Z. Szopniewski, S. Kolesnik, J. Wrobel, P. Gierlowski, and S. Lewandowski, Mater. Sci. Eng. C 15, 171 (2001).
- [9] A. J. Tugwell, D. Hutson, C. M. Pergum, and G. B. Donaldson, Vacuum 37, 331 (1987).
- [10] A. Reible, IEEE Trans. Magn. 17, 303 (1981).
- [11] S. Franssila, J. Vac. Sci. Technol. B 12, 2963 (1994).
- [12] J. M. Martinis and R. H. Ono, Appl. Phys. Lett. 57, 629 (1990).
- [13] B. J. Curtis and H. Mantle, J. Vac. Sci. Technol. A 11, 2846 (1993).
- [14] A. C. Seabra, P. Verdonck, W. L. Xavier, and V. Baranauskas, SPIE 1185, 80 (1989).
- [15] M. R. Boucher, U. Hubner, W. Morgenroth, H. Roth, H.–G. Meyer, M. Schmidt, and M. Eich, Microelectron. Eng. 73-74, 330 (2004).
- [16] T. T. Foxe, B. D. Hunt, C. Rogers, A.W. Kleinsasser, and R. A. Buhrman, J. Vac. Sci. Technol. 19, 1394 (1981).
- [17] A. Pitowska, T. T. Piotrowski, E. Kaminska, Z. Szopniewski, P. Gierlowski, and S. Lewandowski, Electron Technol. 33, 387 (2000).
- [18] C.-K. Hu, N. Mazzeo, S. J. Wind, D. J. Pearson, and M. B. Ketchen, Thin Solid Films 206, 151 (1991).
- [19] D. Schelle and H. J. Tiller, Cryst. Res. Technol. 21, 477 (1986).
- [20] O. Kubashevski, *Niobium: Physico-chemical properties of its compounds and alloys* (International atomic energy agency, Vienna, 1968).
- [21] P. Kneisel, Nucl. Instrum. Methods Phys. Res. A 557, 250 (2006).
- [22] M. Rašković, S. Popović, J. Upadhyay, L. Vušković, L. Phillips, and A-M. Valente-Feliciano, J. Vac. Sci. Technol. A 27(2), 301 (2009).
- [23] M. Rašković, *et al.*, Nuclear Instruments and Methods in Physics Research A 569 663 (2006).
- [24] M. V. Malyshev and V. M. Donnelly, J. Appl. Phys., 88, 6207 (2000).