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Effect of self-bias on cylindrical capacitive discharge for processing of inner walls of tubular structures—Case of SRF cavities

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Cylindrical capacitive discharge is a convenient medium for generating reactive ions to process inner walls superconductive radio-frequency (SRF) cavities. These cavities, used in particle accelerators, presents a three-dimensional structure made of bulk Niobium, with axial cylindrical symmetry. Manufactured cavity walls are covered with Niobium oxides and scattered particulates, which must be removed for desired SRF performance. Cylindrical capacitive discharge in a mixture of Ar and Cl_2 is a sole and natural non-wet acid choice to purify the inner surfaces of SRF cavities by reactive ion etching. Coaxial cylindrical discharge is generated between a powered inner electrode and the grounded outer electrode, which is the cavity wall to be etched. Plasma sheath voltages were tailored to process the outer wall by providing an additional dc current to the inner electrode with the help of an external compensating dc power supply and corrugated design of the inner electrode. The dc bias potential difference is established between two electrodes to make the set-up favorable for SRF wall processing. To establish guidelines for reversing the asymmetry and establishing the optimal sheath voltage at the cavity wall, the dc self-bias potential and dc current dependence on process parameters, such as gas pressure, rf power and chlorine content in the Ar/Cl_2 gas mixture was measured. The process is potentially applicable to all concave metallic surfaces. https://doi.org/10.1063/1.5045692

I. INTRODUCTION

Hollow non-planar surfaces, found in superconductive RF (SRF) cavities, supersonic nozzles or other large-volume tubular devices, often contain particulates or chemical impurities that lead to electric losses or unfavorable hydrodynamic turbulences. These problems can be solved by purification and reduction of surface roughness by critical material removal from the surface either by various acid or discharge-plasma etching techniques. Cylindrical coaxial capacitive discharges are the natural choice for discharge-plasma etching of hollow surfaces, where a discharge is generated between a cylindrical convex powered electrodes, which is centered inside the grounded coaxial tube representing second, outer electrode with the concave surface to be processed. The processes for removal of impurities and particulates are reactive ion etching and field emission. This arrangement is geometrically asymmetric since the two cylindrical smooth surfaces have substantially different areas, leading to distinctively smaller sheath voltage at the outer electrode, which provides unfavorable reactive ion etching rates for processing the concave surface. However, there are geometrical and electric techniques to reverse the asymmetry and provide

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satisfactory etching conditions.[1] To apply correctly those techniques, one needs to know quantitatively the DC parameters arising from the electrode asymmetry – the self-bias voltage and the net DC current between the two electrodes. While this phenomenon has been extensively studied in planar, parallel plate discharges, the case of coaxial cylindrical electrodes was given less attention.

Due to extensive use in the semiconductor industry, planar asymmetric plasma reactors are relatively well understood. Early studies were performed in the context of developing a technique of bias sputtering, in which the substrates in a glow discharge sputtering system are intentionally subjected to positive-ion bombardment during film deposition.[2] This was essentially one of first studies of asymmetric discharge generated electrically in planar electrode geometry. The change in plasma potential and, in turn, the change in ion energy by applying a positive dc voltage was repeated.[3–6] The theoretical model for sheath voltage ratio between two electrodes for these discharges and its dependence on their surface area is also provided in Refs. 7 and 8 and the relation between the self-bias and the etching rate in Si and SiO_2 was studied.[9] The scaling model for sheath voltage ratio between two electrodes for these discharges and its dependence on their surface area is provided in Refs. 6–8 and the operation for dc and rf plasma is described in Ref. 10. Early work on the geometrically asymmetric rf capacitive discharges was focused on a planar excited electrode and a large area grounded enclosure that had sometimes the cylindrical form.[2,3,6,10–14] The enclosure was applied to fully confine the discharge. Plasma surface processing of a large concave metallic surface with the aim of decreasing its surface roughness and metal impurity has been scarcely addressed.

In what follows we will be using an analogy between the planar and cylindrical electrode geometries. In the generic case of the cylindrical asymmetry (see Fig. 1), the two electrodes are two coaxial cylinders. The inner (powered) cylinder has smaller surface area. The outer cylinder with larger surface area is grounded electrode. Although it is not shown in the schematic, the outer electrode is approximately three times longer, and it is closed with two bases. This was found to be the optimal configuration for the outer electrode surface processing, but it becomes essentially a three-dimensional electrode configuration. To complete the whole setup for plasma processing additional components are attached to the outer cavity including the gas flow setup and vacuum pumps. This addition makes the grounded surface area much larger than the simple diagram shown in Fig. 1. In the case of smooth cylinder surfaces, the discharge is also asymmetric, because the surface area of the two electrodes is substantially different. The geometrical approach to reducing the asymmetry in cylindrical geometry may be treated as a qualitative analogue to planar asymmetry corrections, which were studied in detail by Schmidt *et. al.*[15] The self-bias dependence on gas pressure and rf power for planar asymmetric plasma is also reported in Ref. 16. However, its behavior in Ar/Cl_2 plasma with coaxial type asymmetry is not known. Measurement of the self-bias potential for different diameter electrodes and its variation dependence on process parameters is important for any kind of modifying the inner surface of three dimensional structures.

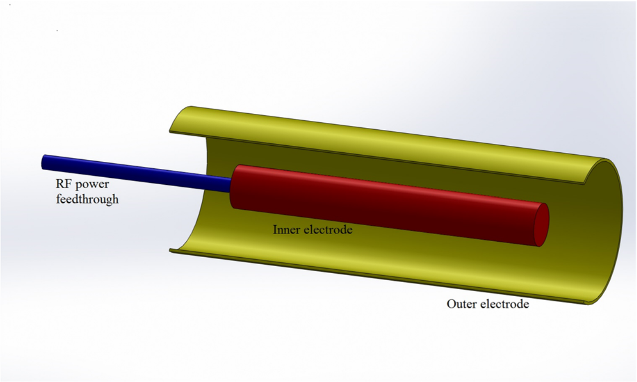

FIG. 1. Schematic layout of two coaxial cylindrical electrodes.

We have developed a method to modify the inner surface of superconducting radio frequency (SRF) cavities made of Niobium (Nb) by using coaxial cylindrical capacitive radio-frequency discharge in Argon/Chlorine (Ar/Cl_2) gas mixture.[17] Radiofrequency discharge can be used to uniformly clean, purify and polish concave metallic surfaces, which is impossible to perform with microwave discharge excitation at constant transverse conditions due to low wavelength compared to the characteristic surface dimension. While present work is focused on the treatment of inner surface of an accelerator SRF cavity the application may be more general. In all these cases, the generic setup is the coaxial cylindrical geometry where the inner electrode is powered, the outer electrode to be processed by ion assisted etching, is grounded, and the cylindrical radio-frequency discharge is generated. We are applying the cylindrical radio-frequency discharge in a mixture of Ar and Cl_2, which implies that the discharge is asymmetric and electronegative, with several consequences to its properties. We have partially corrected its asymmetry by geometrical means and by introducing a dc voltage to optimize the sheath voltage on the outer cylinder for ion-assisted reactive etching of its wall.[17] Additional dc current is needed to bring the negative self-bias potential at the inner electrode to zero a positive value. The main subject of this work is to inspect the effects of the Ar/Cl_2 discharge properties on the asymmetry, compared to the pure Ar plasma. The goal is to remove non-superconductive Nb oxides and metal impurities from the surface of the inner cavity wall. The removal of Nb oxides depends on the ion-assisted variant of reactive ion etching of the inner surface of the outer electrode.[1]

Plasma surface processing of a large concave metallic surface with the aim of decreasing its surface roughness and metal impurity has been scarcely addressed. Here, a generic discharge geometry would be a coaxial system of cylindrical electrodes as shown schematically in Fig. 1 and a radio frequency discharge with two coaxial sheaths generated between them. Although SRF cavities exhibit variable diameter and thus require a special processing procedure[18,19] for clarity, we have adopted as a first step the flat cylinder configuration, where the cavity is the outer cylinder and an auxiliary, powered electrode is the inner cylinder. The surface area asymmetry between inner and outer electrodes due to the coaxial geometry creates a negative self-bias potential on the plasma sheath at the inner electrode. While the self-bias formation in planar geometry can be advantageous for semiconductor wafer processing (as it is placed on the electrode having the self-bias potential) it is detrimental in the case of processing the outer surface of cylindrical structures. The negative dc self-bias potential provides much higher energy of ions in the inner (powered) electrode sheath compared to the ion energy to the outer (grounded) electrode. Since the ion energy gained in the outer electrode sheath is very low, it is not feasible to etch outer electrode without applying a positive dc bias to inner electrode by an external positive dc voltage. In this approach, a DC power supply drives an additional dc current through the inner electrode to bias it positively and change the plasma potential of the bulk plasma, whereby increasing the voltage drop in the outer electrode sheath, which leads to the energy increase of the ions impinging the outer wall.

Particle accelerators and accelerator based light sources in use, or proposed to be built in the future, require SRF cavities, which are the cylindrical structures but with varying diameter. To remove the mechanically contaminated or damaged layers from the inner surface of these structures, hydrogen fluoride (HF) based chemical methods like buffered chemical polishing (BCP) or electro polishing (EP) have been used.[20] Plasma based surface modification will not only reduce the cost and environmental hazards but also open the possibilities of tailoring the surface for better SRF properties. We previously established the dependence of the etch rate on process parameters for the plasma based method,[17] determined the etching mechanism of Nb,[21] and established corrective geometry and external bias for the reversal of asymmetry in cylindrical capacitive discharges.[1] The etch rate non-uniformity due to depletion of radicals along the gas flow direction on process parameters is reported in Ref. 21. We have determined that the etch rate was substantially higher when the negative self-bias at the inner electrode was reduced.

In this paper, we present the dependence of the self-bias potential and external dc current on gas pressure, rf power, and concentration of the chlorine in gas mixture for different diameter inner electrodes. In the following, we describe the experimental setup and discuss the results. The discussion section contains the definition and evaluation of the exponential asymmetry scaling parameter, based on the measured self-bias voltage and the plasma potential in the bulk. The result shows that the 3D effect in the finite-length coaxial capacitive discharge leads to the different type of the asymmetry parameter variation with power compared to the planar geometry.

II. EXPERIMENTAL SETUP

To evaluate the asymmetry in a coaxial type cylindrical rf plasma reactor, we are using, as an example, a 7.1 cm diameter and 15 cm long cylindrical tube as an outer electrode, which is the part of the vacuum vessel. The inner electrode is of varied diameter (2.5, 3.8 and 5.0 cm) and fixed length of 15 cm. An rf (13.56 MHz) power supply is used to generate the discharge. It is connected to an automatic impedance matching network, which also measures the self-bias potential developed on the inner electrode sheath. The matching network has an additional option to connect a dc power supply in order to vary the dc bias on the inner electrode. The setup includes a dc power supply to provide the current required for each condition to reduce the dc self-bias to 0 V. The gases used were pure argon or chlorine diluted to 15% by adding argon. More details of the experimental setup were described earlier.[1]

The inner electrode was powered, and the outer electrode was grounded. The ions gain energy in the sheath due to the potential difference between the time averaged potential of bulk plasma V_p and the surface. An increase of negative bias at the inner electrode leaves the plasma potential unchanged, but it can be changed significantly if a positive dc bias is applied.[3–6] The dc coupling allowed a dc current to flow to the powered electrode and to expand the plasma structure to the whole chamber. In the case of low rf power without dc bias, the plasma is confined to the inner electrode, as similarly observed for planar geometry.[6] The etch rate data for different diameter of the inner electrode indicate that the variation in plasma potential was smaller in the case of the smaller diameter electrode and the etch rate of Nb (inner wall of outer electrode) was reduced.

The negative self-bias potential developed across the inner electrode sheath for all three inner electrode diameters was measured at different gas pressure, rf power and two gas compositions. The required dc current to bring this potential to zero or to positive value at a certain value was also recorded.

III. RESULTS AND DISCUSSIONS

Presented results are a part of the data base that is used to develop a model, which describes the ion-assisted etching process on the inner surface of the cylindrical structure. The results are presented in three sections. In first section the self-bias potential is plotted for pure Ar at variable gas pressure and rf power using electrodes with different diameters. In second section the self-bias potential variation for pure Ar and Ar/Cl_2 mixture is given for different gas pressure and rf power. The dc current required to increase the self-bias potential to zero is plotted for Ar and Ar/Cl_2 plasma for the same electrode diameter with different gas pressure and rf power in the third section. Though the self-bias potential is negative, it is plotted on positive axis for convenience. The error in measurements of Cl_2 concentration was 2%, in rf power 3 W, in pressure 4 mTorr, in dc bias 2 V and in dc current 5 mA. The cylindrical discharge was not confined and the plasma expanded longitudinally with power. This expansion is the main factor in inducing field effect and burn of particulates from the most critical areas of the cavity wall to possibly eliminate one of the main perturbing factors in the electron accelerator operation. It was shown in Ref. 19 where the expanded plasma region was used to induce field emission on the outer electrode to clean out the residual particulates. This resulted in the absence of field emission in the single-cell SRF cavity.[19] In addition, the measurements of the self-bias proved useful since the results helped to determine conditions for high etch rates of Nb in the reversed asymmetric cylindrical configuration.[1]

A. Self-bias voltage dependence on pressure and power

Dependence of self-bias voltage between the two electrodes on pressure and rf power for different electrode diameters in Ar discharge is an important preliminary indicator for reactive ion etching. It shows the bulk discharge properties, and its deviation helps overall discharge correction. All

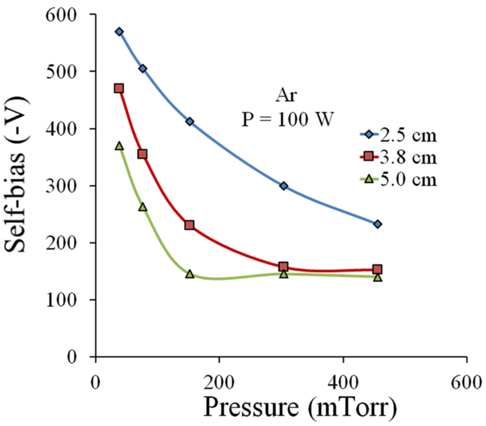

FIG. 2. Self-bias voltage dependence in the Ar plasma on the pressure for different electrode diameters. Solid lines are visual guidelines.

measurements of the self-bias were made using a blocking capacitor, which excludes the effective dc ion current in the circuit.

The self-bias dependence on pressure was measured at rf power of 25, 50, 100 and 200 W. The variation of the absolute value of the self-bias voltage with pressure for Ar plasma using the three diameter electrodes at rf power of 100 W is shown in Fig. 2. The trends of the curves for the other measured rf powers are similar. Although the self-bias potential is negative, its absolute value was used.

The self-bias voltage displayed in Fig. 2 shows two distinct pressure regimes, one below 150 mTorr and other above 150 mTorr. The increase of the self-bias voltage at low pressures could be explained by the expansion of the plasma volume toward the grounded area.[16] Note that the absolute value of the self-bias voltage increases with the ratio of inner and outer electrode radiuses.

Self-bias dependence on the rf power for different diameter electrodes in the Ar plasma is shown in Fig. 3. Presented data are taken at pressure of 38 mTorr.

The increase in the self-bias with the absorbed rf power is not only due to the expansion of the plasma but also the rf voltage increase. The fit to these curves shows almost square root dependence on the rf power indicating that all the rf voltage is dropping on the inner electrode sheath as a dc bias.

B. Self-bias voltage dependence on gas composition

The self-bias voltage varies for pure Argon and Ar/Cl_2 mixture. Figure 4 shows the variation of self-bias voltage with the effective rf voltage for the inner electrode diameters of 2.5 and 5 cm, that

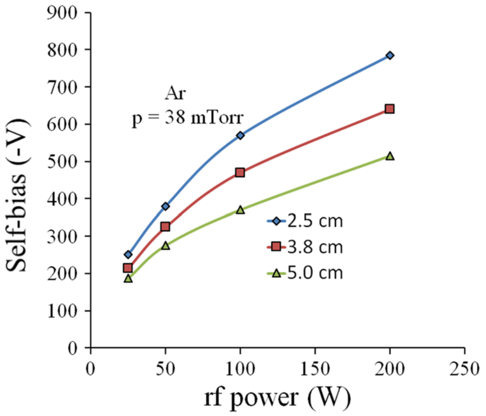

FIG. 3. Self-bias dependence in the Ar plasma on the rf power for different diameter electrodes. Solid line is a visual guideline.

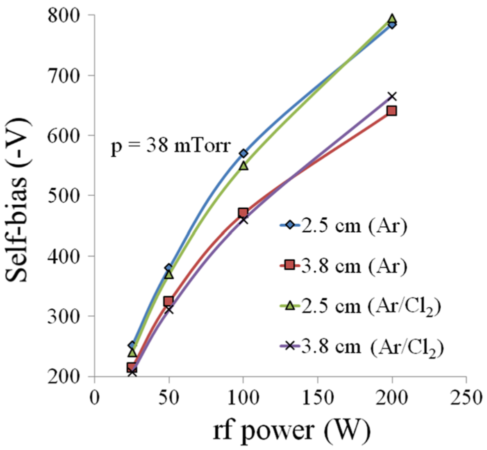

FIG. 4. Self-bias dependence on the rf power for different diameter electrodes in Ar and Ar/Cl$_2$ plasma. Solid lines are visual guidelines.

is with the radius ratio of 0.7 and 0.35. The absolute value of self-bias voltage for Ar/Cl$_2$ increases with the ratio of the electrode radiuses, but is practically the same for pure Argon and for the Ar/Cl$_2$ mixture. This behavior could be partially explained with the relative electron density decrease at lower power in Ar/Cl$_2$ plasma compared to the positive ion density due to large plasma electronegativity. The electron density is almost equal to the positive ion density at higher rf power density as reported in Ref. 22. This property of chlorine plasma is reflected in the self-bias voltage variation at higher rf power.

In the case of the inner electrode diameter of 5.0 cm, the plasma volume is smaller and equal density of electrons and positive ions is reached earlier, similarly to Ref. 22. However, the electron temperature is higher in the Ar/Cl$_2$ plasma compared to the Ar plasma, consequently the self-bias voltage in the Ar/Cl$_2$ plasma is higher than the pure Ar plasma, as shown in Fig. 5.

The variation of the self-bias with pressure at fixed rf power for pure Ar and Ar/Cl$_2$ plasma for inner electrode diameter of 5.0 cm is shown in Fig. 5.

Figures 4 and 5 also show that, due to lower electron density and higher electron temperature in Ar/Cl$_2$ plasma, the self-bias voltage is higher compared to pure Ar plasma. This higher self-bias voltage shows that the addition of electronegative gases such as Cl$_2$ increases the asymmetry in rf plasma reactors.

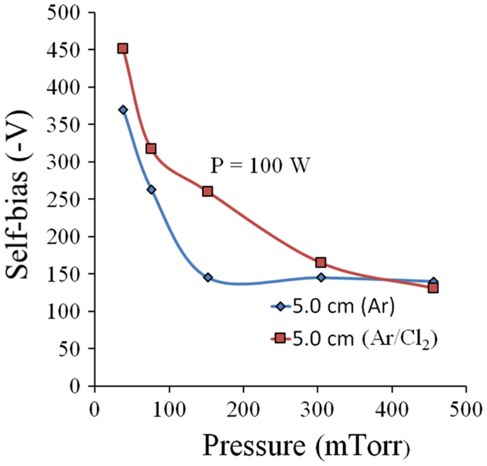

FIG. 5. Self-bias dependence on the pressure for pure Ar and Ar/Cl$_2$ mixture plasma for the same diameter electrode. Solid line is a visual guideline.

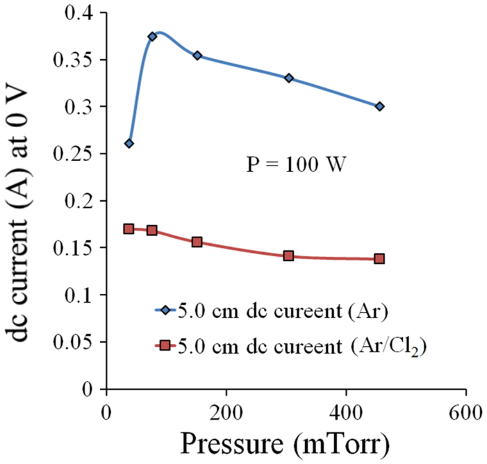

FIG. 6. The dc current dependence on the pressure for 5.0 cm diameter electrode for Ar and Ar/Cl$_2$ plasma. Solid lines are visual guidelines.

C. dc current dependence on the gas composition

To reverse asymmetry of cylindrical capacitive discharge, additional dc current must be provided with the help of an external positive bias voltage with the adequate DC power supply and in the absence of blocking capacitor. Addition of positive voltage source brings the overall dc bias voltage to zero or the positive value to increase the plasma potential for all three inner electrodes. The current, needed to increase the negative dc self-bias to 0 V, could be treated as an indicator of electron density in the plasma. The dc current required to lift up the self-bias voltage to zero for pure Ar and Ar/Cl$_2$ plasmas, is plotted in Fig. 6.

Figure 6 shows that much less current is required in the case of Ar/Cl$_2$ discharge, as the electron density is approximately an order of magnitude lower in Cl$_2$ plasmas compared to Ar plasma, similarly to Ref. 23. The same trend is recorded for the other two diameter electrodes for all the pressure and power conditions.

The addition of Cl$_2$ decreases the electron density in the plasma. The increase in dc bias voltage increases the current provided by the dc power supply. The dc current variation with the dc bias

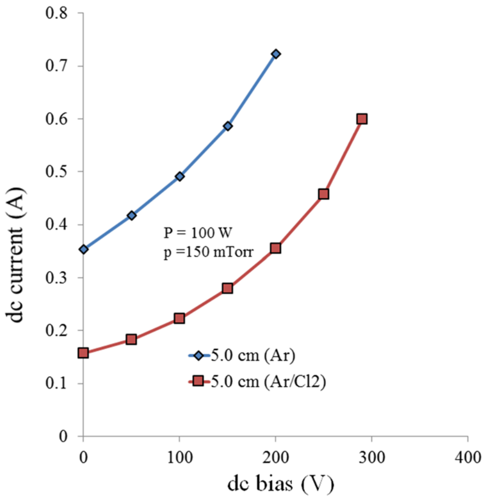

FIG. 7. The dc current variation with the dc bias voltage at fixed pressure and fixed rf power for Ar and Ar/Cl$_2$ plasma. Solid lines are visual guidelines.

voltage at fixed pressure of 150 mTorr and fixed rf power of 100 W for Ar and Ar/Cl_2 plasma is shown in Fig. 7.

The increase in dc current with biased voltage indicates that positive dc biasing not only increases the plasma potential but also the plasma density, as also shown in case of planar geometry.[16]

IV. CONCLUSION

This study presents the analysis of asymmetry scaling in cylindrical coaxial capacitive coupled discharge and the self-bias dependence on gas pressure and rf power using different diameters of the inner electrode. The self-bias data show a difference between Ar and Ar/Cl_2 plasma and an explanation for this difference is offered since the number of electrons is highly reduced in the electronegative Ar/Cl_2 discharge. Enclosed cylindrical geometry has been rarely studied in its generic form despite its simplicity and adaptability to practical surface processing of concave structures. Exponential scaling asymmetry factor depends on the discharge power. These experimental results will serve as a part of the data base that will be used to develop a processing model for the coaxial cylindrical configuration. The sheath potential asymmetry is a natural property of the coaxial cylindrical discharges and the results show that the dc bias approach can be used for reduction, elimination, or reversal of the asymmetry. This study also presents the variation in dc current required for bringing the self-bias potential to zero or positive for Ar and Ar/Cl_2 plasma and the role of plasma density in this variation. Results also lead to the observation that positive dc bias at the inner electrode increases the plasma density together with sheath potential at the outer electrode, which is beneficial in etching the inner surface of the cavity outer wall, as was demonstrated in Ref. 19. This does not exclude other effects, such as the temporal and spatial variations of the self-bias potential.

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