PLASMA TREATMENT OF BULK NIOBIUM SURFACE FOR SRF CAVITIES - OPTIMIZATION OF THE EXPERIMENTAL CONDITIONS ON FLAT SAMPLES*

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Abstract

Accelerator performance, in particular the average accelerating field and the cavity quality factor, depends on the physical and chemical characteristics of the superconducting radio-frequency (SRF) cavity surface. Plasma based surface modification provides an excellent opportunity to eliminate non-superconductive pollutants in the penetration depth region and to remove the mechanically damaged surface layer, which improves the surface roughness. Here we show that the plasma treatment of bulk Nb presents an alternative surface preparation method to the commonly used BCP and EP methods. We have optimized the experimental conditions in the microwave glow discharge system and their influence on the Nb removal rate on the flat samples. We have achieved etching rate of 1.7 μm/min using only 3% Cl_2 in the reactive mixture. Combining a fast etching step with a moderate one, we have improved the surface roughness without exposing the fresh sample surface to the environment. We will apply the optimized experimental conditions to the preparation of some single cell cavities, in pursuing improvement of their RF performance.

INTRODUCTION

Plasma etching of Nb thin films has been readily used in production of Josephson tunnel junctions [1]. However, except for our publications [2-4] there are no other reports of plasma treatment of bulk Nb used for SRF cavities. In discharge plasmas containing chlorine or fluorine radicals, the deformed, contaminated or oxidized bulk Nb surface interacts with these radicals producing volatile Nb halides. The production of reactive species in the discharge and, consequently, the Nb removal rate is determined by the input power, the pressure, the temperature, and the radical concentration. The same plasma parameters are determining the thickness of plasma sheath. the plasma sheath is the region in the plasma right above the Nb surface characterized by a drop in potential [5]. Positive ions from the plasma, accelerated in the sheath, hit the sample surface where they deposit their energy, recombine, and react. If excessively accelerated, the ions could knock out atoms or atomic conglomerates from the sample surface, thereby increasing the surface roughness. Moreover they can be implanted into the Nb surface and increase the level of impurities in the penetration depth region. Therefore, plasma parameters during the plasma etching process have to be finely balanced to achieve high etching rates without increasing the surface roughness or introducing impurities due to the process.

Before a single cell cavity would be etched in plasma discharge, the optimization of experimental conditions was performed on flat Nb samples. They were exposed to a microwave glow discharge system using Cl_2 as a reactive gas. The results of the etching rate optimization were published elsewhere [2]. We also presented results of the detailed surface composition analysis performed by x-ray photoelectron spectroscopy (XPS) [3]. The surface characteristics of the Nb samples exposed in the d.c. pulsed repetitively pulsed DC diode system (PLAD) with BF_3 as reactive gas, were published elsewhere [5]. Therefore, in this paper we will focus on the influence of plasma parameters on surface roughness of samples exposed in the microwave glow discharge.

EXPERIMENT

Sample Preparation and Experimental Set-up

In order to determine the influence of the sample preparation history on surface roughness after exposure to a microwave glow discharge, samples surface was prepared in one of the following ways. Some samples were left unprocessed (NP) i.e. they were exposed to plasma as received after electron beam cutting, while other were mechanically polished (MP) down to 9 μm average roughness. After that, some samples were exposed to 1:1:2 buffered chemical polishing (BCP) mixture for removal of approximately 100 μm of surface. Electropolishing (EP) in 1:9 mixture was performed after BCP for 4 hours removing additional 100 μm of surface.

After surface preparation, samples were exposed to reactive gas in the microwave cavity discharge system described in Ref. [3]. The experimental set-up is connected to a spectrometer with the CCD camera for emission spectroscopy measurements. Emission spectroscopy is used as a process monitoring technique as well as a tool to determine the reaction mechanism of the plasma etching process.

Surface Roughness Diagnostic Techniques

We measured the surface roughness using a commercial tapping mode atomic force microscope (AFM) (Digital

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Instruments: Nanoscope IV). Silicon tips with tip size of 10 nm were used. The scan sizes were 20 μm x 20 μm, 50 μm x 50 μm, and 100 μm x 100 μm, when ever it was possible. The limiting factor for the Nanoscope IV was the maximum vertical range of 6.5 μm. The typical measurement of surface roughness is expressed as the root mean square (RMS) roughness, defined as:

$$RMS = \sqrt{\frac{1}{n}\sum_{i,j}(h_{i,j} - \bar{h})^2}$$

where n is total number of taping points, i is number of taping points in x direction, j is a number of taping points in y direction, $h_{i,j}$ is height reading for the point (i,j), and $\bar{h}$ is average height reading for whole scan. The RMS depends on the scan size and instrument resolution. Therefore it is not recommended to compare the RMS from measurements performed on different instruments and with different scan sizes. To go around this problem and to be able to see real influence of plasma etching process on surface roughness, we performed the same size scans of a referenced region before and after exposure to plasma. These results are compared in this paper.

RESULTS

Influence of Plasma Parameters on Surface Roughness

Plasma etching of Nb in Ar/Cl_2 microwave discharge is chemical in nature what means that the etching rate is primarily determined by the concentration of reactive species in the plasma. The dependence of the etching rate on the initial concentration of molecular chlorine Cl_2 at constant power is presented in Fig. 1. The tching rate is increasing with initial Cl_2 concentration on power 3/2. Surface RMS of same samples is presented in the same diagram. Scan size was 20 μm x 20 μm. We can conclude that surface roughness does not depend significantly on concentration of Cl_2. This can be attributed to the kinetics of the chlorine dissociation process, which saturates for given power and at relatively low initial concentration of Cl_2.

The etching rate depends less dramatically on the input power density than on concentration of Cl_2, as shown in Fig. 2. Conversely, the RMS dependence on the input power density shows a peak for medium power densities. It also has to be noted that the scan size for the set of measurements presented in Fig. 2. was only 10 μm x 10 μm due to instrument limitations.

The Nb etching rate much stronger depends on the pressure in the reaction chamber than on the power density, as shown on Fig. 3. The surface roughness, measured by RMS is following that dependence. Reasons for this behavior may lay in the increase of plasma density with the increase of pressure and power. The plasma sheath is proportional to the plasma density and therefore if we increase the power or the pressure, we increase the energy of ions that are hitting the surface. This contributes to an increase of the surface roughness. After a peak, the chemical etching rate overcomes the sputtering rate and the chemical process contributes to the smoothening of the Nb surface.

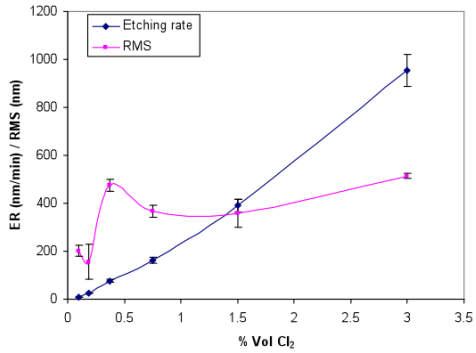

Figure 1: Etching rate and surface RMS dependence on concentration of Cl_2.

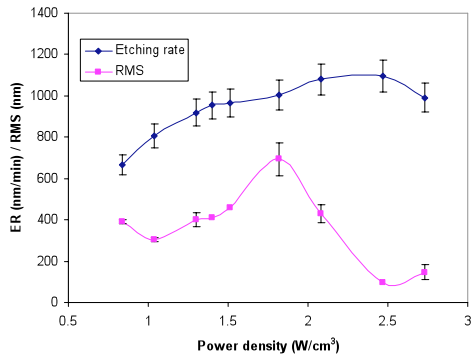

Figure 2: Etching rate and surface RMS dependence on input power density.

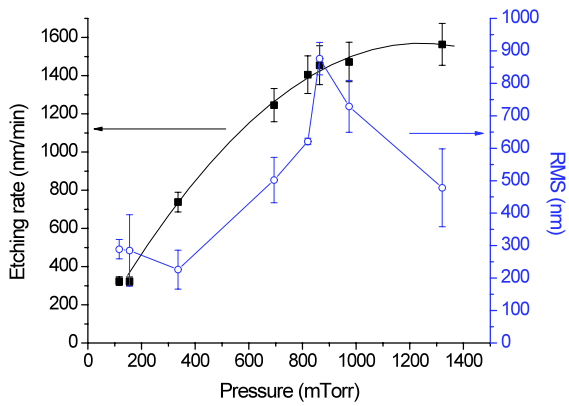

Figure 3: Etching rate and surface RMS dependence on pressure.

Tri-step Plasma Etching Process

Taking into account the influence of the plasma parameters on the surface roughness, a tri-step plasma etching process was designed. In the first step, pure argon (Ar) discharge was produced under a total pressure of 500

mTorr and a power density of 2.08 W/cm^3. During the 30 minutes of etching, no Nb was removed from the surface (etching rate is 0 nm/min as shown before [4]) but all physisorbed gasses and organic residues were removed. Starting with a clean surface, we are performing fast plasma etching in the second step. 3 Vol% Cl_2 in Ar is used as etching gas to remove the surface necessary for cavity production. The exposure time was 120 min under a total pressure of 550 mTorr and input power density of 2.08 W/cm^3. The etching rate in this step was 1 μm/min and approximately 120 μm of surface was removed during 2 hours of exposure. The third step was designed to remove Nb from the surface under conditions more favorable for surface smoothening. 1.5 Vol% Cl_2 in Ar was used under a total pressure of 1250 mTorr and input power density 1.4 W/cm^3. Etching rate was 0.5 μm/min. Approximately 100 μm of surface was removed during 4 hours of exposure.

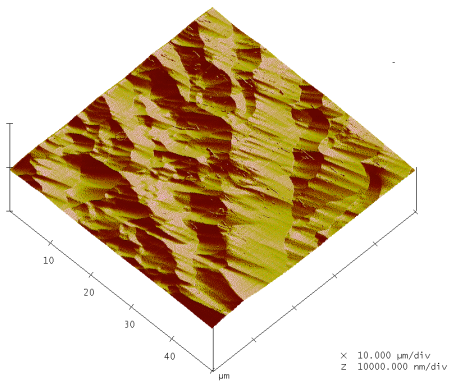

Figure 4: AFM scan (50 μm x 50 μm) of BCP prepared surface.

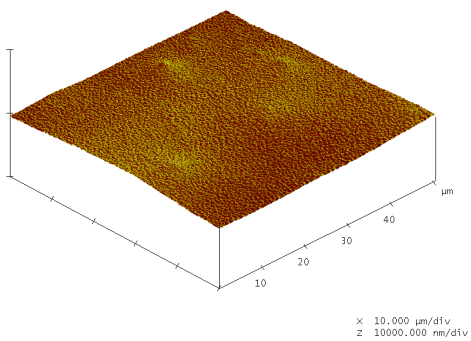

Figure 5: AFM scan (50 μm x 50 μm) of surface prepared by plasma etching.

Figures 4 and 5 present the same surface before and after exposure to plasma etching. The surface presented on the Fig. 4. is prepared by BCP so the polycrystalline structure is prominent. The plasma etching has eliminated the surface features (Fig. 5.) Scanning electron microscope (SEM) was used to enlarge and understand nature of peaks present on surface after plasma etching. Energy dispersive x-ray (EDS) analysis has shown presence of Si particle on the surface that could not be removed by plasma etching. The most probable source of Si is from residues of the mechanical polishing on the surface before exposure to theplasma discharge. Since etching rate of SiO_2 in Cl_2 plasma is much smaller than etching rate of Nb, SiO_2 acts as shield for this part of the surface, forming a feature present on Fig. 6.

Due to the element sensitivity of the plasma etching process, the surface preparation history plays an important role in determining the final surface quality. The comparison of RMS measurements before and after the plasma etching process depending on the surface preparation history is presented in Table 1. Note that the mechanically polished surface was too rough for a 50 μm x 50 μm scan before plasma processing, therefore 20 μm x 20 μm scan results were presented in table. From Table 1, one can see that plasma etching improves the surface roughness if the starting surface was prepared by mechanical polishing or BCP. The RMS does not change if the surface was prepared by EP. Therefore we can say that plasma etching produces surfaces of lower or comparable roughness with respect to other used Nb surface preparation techniques.

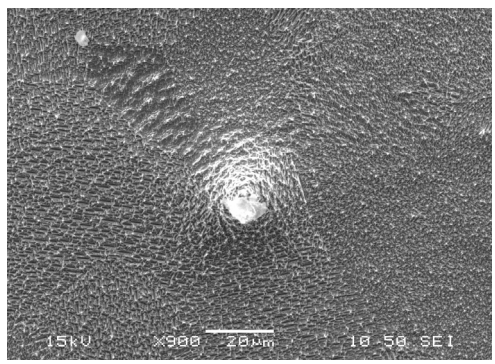

Figure 6: SEM scan of surface prepared by plasma etching.

Table 1: RMS Measurements Dependence on Surface Preparation History

Surface history	Scan size (μm x μm)	Plasma etching	RMS (nm)
NP	50 x 50	Before	254
		After	231
MP	20 x 20	Before	758
		After	637
BCP	50 x 50	Before	286
		After	215
EP	50 x 50	Before	133
		After	134

CONCLUSIONS

We have shown that the etching rates of bulk Nb as high as 1.7 ± 0.2 μm/min can be achieved in a microwave glow discharge using Cl_2 as the reactive gas. The Nb etching rate depends on the Cl_2 reactive gas concentration and the discharge parameters: input power density and pressure in reaction chamber. The surface composition analyses show that no impurities have been introduced into Nb during microwave discharge treatment.

To optimize the surface roughness, we have developed a three-step process that gives results comparable to wet processes, producing surfaces of satisfying roughness, which is close to optimal.

Emission spectroscopy results combined with measured etching rates, suggest that the Nb etching mechanism in Ar/Cl_2 MW glow discharge is more a chemical etching process than a physical sputtering one.

A modified experimental set-up that includes a single cell cavity has been built. It will be used to determine the influence of the plasma treatment on RF performance of tcavities. The microwave discharge treatment of Nb surfaces is a convenient, low-cost and less hazardous alternative to the presently used liquid acid etching techniques.

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