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Gas cluster ion beam surface treatments for reducing field emission and breakdown of electrodes and SRF cavities

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Abstract

Sub-micron-scale surface roughness and contamination cause field emission that can lead to high-voltage breakdown of electrodes, and these are limiting factors in the development of high gradient RF technology. We are studying various Gas Cluster Ion Beam (GCIB) treatments to smooth, clean, etch and/or chemically alter electrode surfaces to allow higher fields and accelerating gradients, and to reduce the time and cost of conditioning high-voltage electrodes. For this paper, we have processed Nb, stainless steel and Ti electrode materials using beams of Ar, O_2, or $NF_3 + O_2$ clusters with accelerating potentials up to 35 kV. Using a scanning field emission microscope (SFEM), we have repeatedly seen a dramatic reduction in the number of field emission sites on Nb coupons treated with GCIB. Smoothing effects on stainless steel and Ti substrates, evaluated using SEM and AFM imaging, show that 200-nm-wide polishing scratch marks are greatly attenuated. A 150-mm diameter GCIB-treated stainless steel electrode has shown virtually no DC field emission current at gradients over 20 MV/m.

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Keywords: Cluster-surface interactions; Gas cluster ion beam; RF breakdown; Superconducting RF cavity; Field emission

1. Introduction

It has been recognized since the earliest days of vacuum high-voltage electronics that electrode surfaces need to be clean and smooth to prevent field emission and breakdown. However, even using the best existing technology, field emission remains orders of magnitude greater than the limits set by the Fowler–Nordheim (FN) field emission theory. Historically, the discrepancy has been reconciled by adding the field enhancement factor β to account for the fact that imperfections on the surface greatly increase local field strength ($E_{\text{local}} = \beta E_{\text{applied}}$). When the measurements of field emitted current versus applied field are fit to FN theory, typical values of β range from $100 > \beta > 1000$, very far from the ideal of $\beta = 1$ [1]. When the fitted data are also used to determine A_e, the area of the field emitters, typical emitter dimensions are in the range of 10 nm to 1 μm [1]. It has been postulated that these nanoscale features and even smaller atomic-scale features are the reason that electrodes do not approach the limits set by the FN theory – because present surface finishing techniques do not remove or passivate them. We are investigating electrode preparation using Gas Cluster Ion Beams (GCIB), a new nano-technology for smoothing, cleaning, etching, or chemically altering surfaces that is capable of achieving atomic level smoothness on planar and non-planar surfaces [2]. With GCIB, the surface is bombarded with an energetic beam of nano-size cluster ions. The clusters are formed as a high pressure gas (≈15 bar) expands adiabatically into a region of high vacuum ($\approx 3 \times 10^{-6}$ bar). During the expansion, the gas cools, condenses, and coagulates into a jet of clusters. The jet passes through a differential pumping aperture that

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selects the center of the jet and rejects most of the uncondensed gas. The clusters are ionized by electron impacts and then electrostatically accelerated (using potentials as high as 35 kV in these experiments). A dipole magnet deflects monomers and dimers out of the beam. Typically the treated substrate is mechanically scanned in the beam to assure uniform irradiation. Mack [3] contains more information about the GCIB equipment. Average parameters for the clusters in an Ar GCIB beam accelerated with 30 kV are: mass = 10,000 atoms, charge state = +3.2, and velocity = 6.7 km/s with beam currents >200 μA [4]. GCIB treatments are used industrially in the production of thin-film optical coatings, fixed disk memory technology, EUV lithography masks, and for the manufacture of semiconductor devices [5]. Previous papers have reported results of GCIB treatments of OFE-Cu, stainless steel, Ti and Nb electrode surfaces [6,7]. This paper contains additional data for stainless steel electrodes and reports the direct measurements of the suppression of field emission by GCIB treatments of Nb SRF cavity material.

2. Stainless steel electrode material

Samples of highly polished stainless steel electrode material used for extraction electrodes in high-field photoelectron guns [8] were obtained from Cornell Wilson Laboratory. These samples were carefully hand-polished using diamond paste to an average roughness of <1 μm. These surfaces appeared mirror smooth to the eye but are not smooth at nanometer and atomic dimensions as can be seen in Fig. 1, a typical AFM image of these surfaces. The surface shows isolated asperities and sharp-edged, ≈200-nm-wide scratch marks produced by the polishing compound. The samples were treated with a combination of high and low energy Ar GCIB followed by high and low energy O_2 GCIB. Fig. 2 is an AFM image of the same surface after treatment showing that GCIB effectively removes the asperities and scratch marks. Fig. 3 shows the Power Spectral Density (PSD) analysis of these two images and other AFM measurements of these surfaces. The PSD's are obtained from the Fourier transforms of the AFM pixel height maps and show the relative effectiveness of GCIB processing for different size scales of roughness. GCIB processing is particularly effective for roughness with wavelengths up to 600 nm, and continues to be effective up to roughness of 2 μm wavelength in these measurements. As discussed, this is the scale of roughness that is expected to contribute to field emission, direct evidence that GCIB is effective at smoothing features that could not be smoothed even using the best of previous mechanical polishing techniques for non-planar electrodes. The effective size scale for GCIB polishing is determined by the properties of the surface, the average size of craters made by individual cluster impacts (typically in the range of 10–20 nm for high energy Ar clusters [9–13]), and by the applied GCIB dose. We have seen similar results when treating Ti surfaces with similar initial polish using Ar GCIB.

Previously we reported measurements showing a 10^6 reduction of field emission resulting from GCIB treatment for a 150-mm-diameter stainless steel electrode that was polished similar to those shown in Figs. 1–3 [7]. The field emission was measured at the Jefferson Laboratory Large Area Electrode Test Chamber. This same electrode was re-measured several months later on an identical test stand at Cornell Wilson Laboratory. Whereas the first measurement was swiftly made because of time constraints, the re-measurement allowed more time for high-voltage conditioning of the electrode (hi-potting). Not only did the electrode maintain its original performance but, with

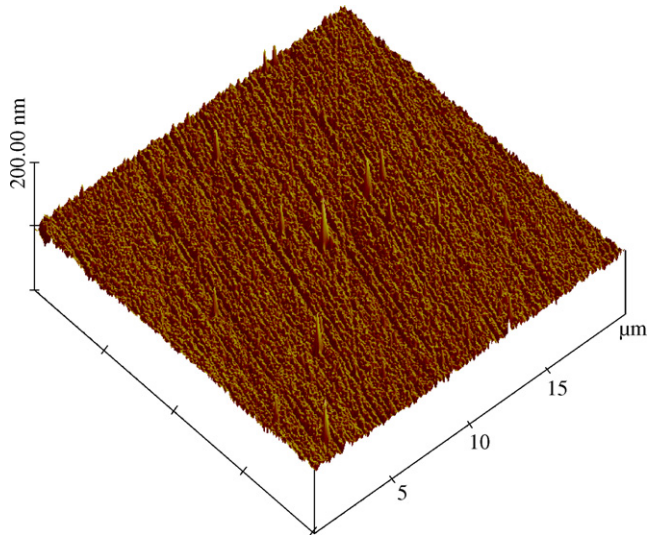

Fig. 1. AFM image (20 × 20 μm) of highly polished stainless steel electrode material before GCIB treatment showing asperities and scratch marks from polishing. The vertical scale is 120 nm/division.

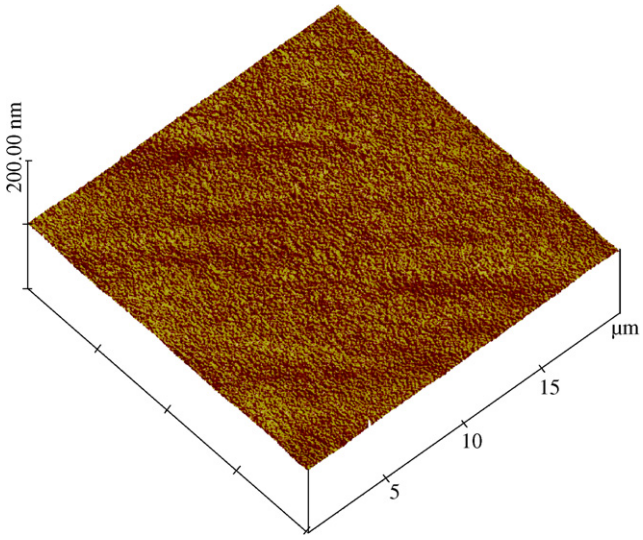

Fig. 2. AFM image (20 × 20 μm) of highly polished stainless steel electrode material after GCIB treatment. The vertical scale is 120 nm/division.

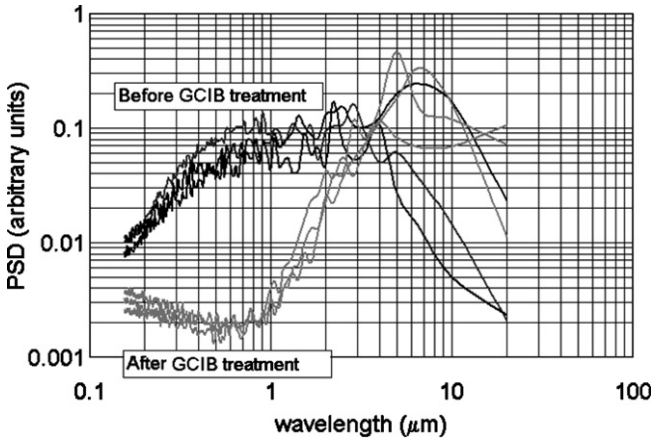

Fig. 3. Six plots of relative two-dimensional isotropic PSD's of 20×20 μm AFM images showing effect of GCIB processing on highly polished stainless steel electrode material. The three black lines are before processing and the three gray lines are after processing. The images were measured at different locations. The vertical scale units are arbitrary. PSD data from Figs. 1 and 2 are included in this plot.

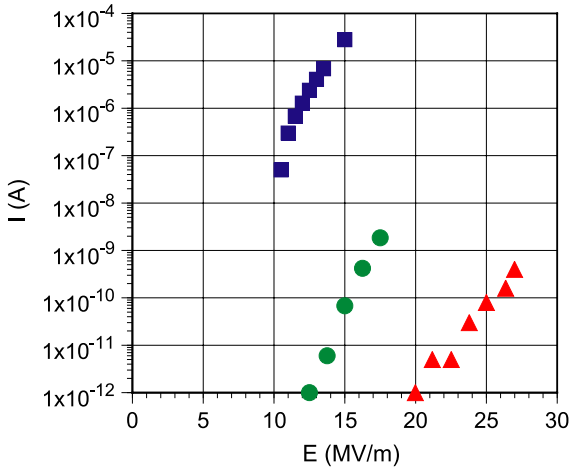

Fig. 4. Field emission current as a function of applied gradient for a 150-mm-diameter stainless steel electrodes: (squares) a typical untreated sample, (circles) first measurement of GCIB treated sample, (triangles) re-measurement of GCIB treated sample after high-voltage conditioning [14].

conditioning, a gradient of over 20 MV/m was reached with no measurable field emission, as plotted in Fig. 4 [14]. It should also be noted that the processing of this electrode included treatments with O_2 clusters which increased the thickness of surface oxide layer from 1.5 nm to >10 nm [7]. The treated surface was also measured to be twice as hard as the untreated surface [7]. The relative contributions of smoothing, oxide thickness and surface hardening to the reduction in field emission are receiving further study.

3. Nb material for SRF cavities

Samples of Nb SRF cavity material were supplied by Jefferson Laboratory, were GCIB treated at Epion, and then returned to Jefferson Laboratory for study. These 25-mm-diameter coupon samples were prepared using buffered chemical polishing (BCP), typically used for SRF cavities [15–17]. Field emission from the samples was studied using the Scanning Field Emission microscope (SFEM) located at the Surface Science Laboratory [18]. The SFEM automatically maps the locations of field emitters on the surface of a sample by raster scanning a biased micro-tip over the surface. At each point on the surface the threshold for field emission is determined by ramping the tip voltage until a specified current level is reached. The system also allows the samples to be transferred under high vacuum into an SEM for imaging, and for study of elemental composition using EDX. The samples are indexed so that field emitters found by the SFEM scan can be relocated and studied using the SEM and EDX. The samples were handled at Jefferson Laboratory and at Epion Corporation using particle-free glove-boxes to minimize contamination. A 25 μm thick stainless steel mask, located approximately 2 mm above the surface, was used to shield a portion of the sample from the GCIB processing. Fig. 4 is the first SFEM map of a GCIB treated Nb sample. The sample was masked into quadrants which received either: no processing, Ar processing, O_2 processing, or a combination of Ar then O_2 processing. All three treated quadrants showed fewer field emitters than the unprocessed quadrant. The untreated quadrant had 11 field emitters while the O_2 treated quadrant had only one field emitter. Comparing these results to a binomial distribution shows less than a 1 in 70 chance that this is a random distribution. The O_2 treatment was retested and again, as is apparent in Fig. 5, there was a dramatic reduction in the number of field emitters, confirming the previous result. On the second sample a total of 23 field emitters were found on the untreated half of the sample and only three on the treated

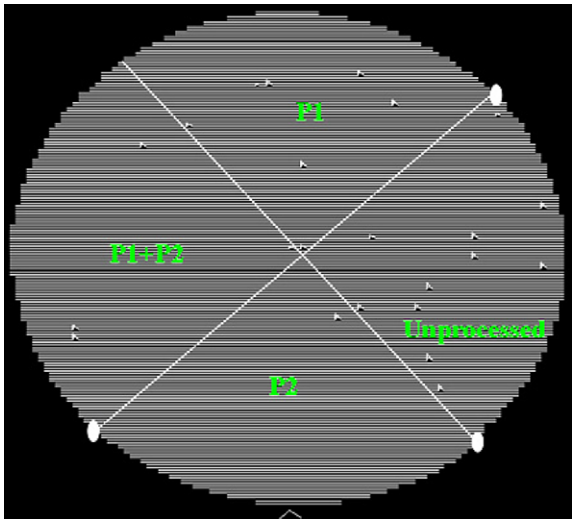

Fig. 5. SFEM Plot of field emitters on surface of a 25-mm-diameter coupon sample of BCP-polished Nb SRF cavity material. The sample was masked into equal quadrants for treatment as designated on the plot. The areas designated with P1 (P2) were treated with Ar GCIB (O_2 GCIB). One quadrant received no treatment and one quadrant received both treatments.

half. Preliminary SEM and SIMS studies show evidence that particles on the surface may be disrupted by the O_2 treatment and that the oxygen stoichiometry of the oxide is increased. It is to be expected that a hypervelocity collision of an oxygen cluster containing several thousand molecules will cause a powerful exothermic chemical reaction on metallic, hydrocarbon and other reactive surfaces in addition to the kinetic collision energy. These "nano-explosions" should be disruptive to particulates because they are structurally weak and thermally isolated. The effect of these reactions on the Nb surface is discussed in another paper [19].

An earlier study concluded that $NF_3 + O_2$ clusters can significantly etch Nb and blunt the angles of the grains that protrude from the surface [7]. The conclusion was based on multiple AFM measurements of the grain edges and SEM images. We treated half of a masked coupon sample with a $NF_3 + O_2$ GCIB and measured the number of field emitters with the SFEM (Fig. 6). Only 10 field emitters were detected on the treated half, while the untreated half had 38 emitters (the results are even more statistically significant than in the case of O_2; the result is four standard deviations from the mean for a binomial distribution). The sample was also studied using a scanning two-dimensional profilometer; however, this instrument did not show significant change in the average roughness measured over a 1500×1500 μm scan area. The fact that the range of heights did not decrease disagrees with our previous findings of possible grain removal [7]. The resolution of this measurement was insufficient to test blunting or rounding of the angles of the exposed grains. We have also measured the etch rate of $NF_3 + O_2$ GCIB using a Quartz Crystal Microbalance. Using 35-kV acceleration voltage, a maximum etch rate was of 17 nm cm^2 s^{-1} has been reached recently. Thus, assuming a skin depth of 50 nm, the entire active surface of an SRF cavity might be removed in a few hours using this processing. The measurements were made on a test stand that has less than half the capability of a production machine so the result is a conservative estimate.

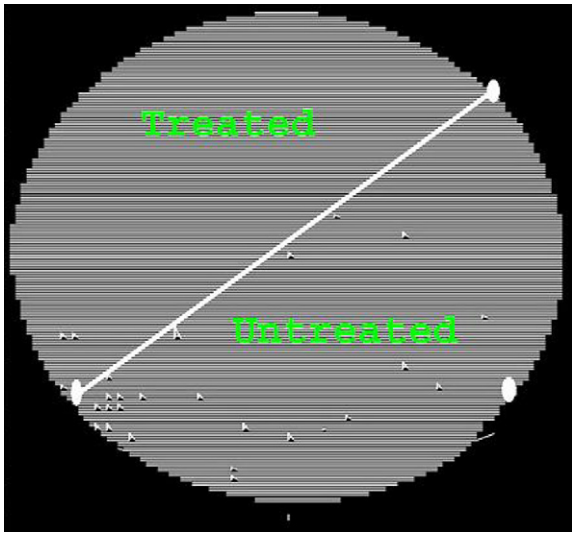

Fig. 6. SFEM plot of field emitters on surface of a 25-mm-diameter coupon sample of BCP-polished Nb SRF cavity material. Half of the sample was treated with O_2 GCIB beams.

4. Conclusions

It is evident that GCIB processing is particularly effective for smoothing roughness of size scales <1 μm. We have previously shown accelerated etching of isolated asperities 300-nm diameter and 35 nm height [6]. Thus GCIB processing is uniquely capable of smoothing roughness of the scale of the field emitters that have been identified on electrodes by fitting to the FN theory. There is also strong evidence that GCIB can dramatically suppress field emission on Nb SRF cavity surfaces. With this compelling evidence, Epion Corporation has begun the design of equipment for in situ treatment of Nb SRF cavities. Vertical RF tests of cavity Q will be used to evaluate the effectiveness of various GCIB treatments for reducing field emission, breakdown, "high-field Q slope" and for increasing the critical field for quenching. Finally, while achieving a 20 MV/m gradient on stainless steel without appreciable field emission is exceptional, it should be noted that a calculation using FN theory for an ideal electrode (assuming a workfunction $\phi = 4.5$ eV, $A_e = 115$ cm^2 and $\beta = 1$) yields a field emission threshold of over 1 GV/m. Thus, we are far from the ideal electrode and large improvements should be possible.

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