

## SURFACE ROUGHNESS CHARACTERIZATION OF NIOBIUM SUBJECTED TO INCREMENTAL BCP AND EP PROCESSING STEPS\*

H. Tian<sup>+#</sup>, G. Ribeill<sup>\*</sup>, M. J. Kelley<sup>+#</sup>, C. E. Reece<sup>+</sup>

<sup>#</sup>Applied Science Dept., College of William and Mary, VA 23187, USA

<sup>\*</sup>Physics Dept., North Carolina State University, NC 27695, USA

<sup>+</sup>Thomas Jefferson National Accelerator Facility, VA 23606, USA

### *Abstract*

The surface of niobium samples polished under incremental Buffered Chemical Polish (BCP) and Electro-Polishing (EP) have been characterized through Atomic Force Microscopy (AFM) and stylus profilometry across a range of length of scales. The results were analyzed using Power Density Spectral (PSD) technique to determine roughness and characteristic dimensions. This study has shown that the PSD method is a valuable tool that provides quantitative information about surface roughness at different length scales.

### INTRODUCTION

The performance of Superconducting Radio Frequency (SRF) cavities used in modern accelerators is intimately related to the final surface treatments. In the preparation of Nb cavities, surface damage and contamination are removed by chemical etching and followed by post-etch treatments. The two widely practiced etch technologies are buffered chemical polish (BCP) and electropolish (EP).

Surface roughness has proven to be a critical factor in determining the efficiency and maximum accelerating gradient that SRF cavities can achieve. Undesirable effects such as: magnetic field enhancement, the creation of “hot spots” and electron multipacting have been shown to be directly affected by the quality of the niobium surfaces [1-2].

The surface morphology of niobium after etching has not been systematically characterized, so it is unknown at what scale roughness is important to SRF performance. Seeking such an analysis, the roughness of niobium surfaces have been measured at a range of sizes and resolutions, which can only be accomplished using several different instruments. A method of combining the measurements from different instruments is therefore needed. Studies have shown that Power Spectral Density (PSD) based description of surfaces are less dependent on instrumental effects when measuring parameters such as surface roughness and correlation length [3]. From PSD, one can directly obtain the characteristic length of surface features as well as the fractal dimension of the surface.

### EXPERIMENTAL STUDIES

#### *Sample preparation*

The samples were high purity polycrystalline Nb slabs with dimensions of 22mm x 22 mm x 4 mm. All were mechanically ground using 1  $\mu$ m grit prior to any chemical treatment, as is done in cavity production. The BCP solution was a 1:1:2 (by volume) mixture of HNO<sub>3</sub> (69%), HF (49%) and H<sub>3</sub>PO<sub>4</sub> (85%). The EP solution was a 1:9 mixture of HF (49%) and H<sub>2</sub>SO<sub>4</sub> (96%). The samples used as a reference for an “untreated” state were etched in a fresh BCP (1:1:2) bath for 5 minutes to remove any residual surface contamination from previous mechanical work. In this study, “Light BCP” refers to the samples which were etched in a fresh BCP (1:1:2) bath at room temperature for 20 minutes. As measured by weight loss, 30  $\mu$ m removal was achieved. Those samples referred to as “Heavy BCP” were placed in a fresh BCP (1:1:2) at room temperature for 90 minutes which resulted in a 150  $\mu$ m removal.

For electropolishing, the Nb sample and a high purity aluminum plate with an area ratio of 1: 2 were inserted into Teflon clamps with a separation of 98 mm. Samples having undergone no BCP treatment, as well as those having undergone light and heavy BCP treatment were subjected to 30 minutes of EP at 6V individually. (The polarization potential is measured relative to a MSE reference electrode, with electrolyte temperatures of 24°C and 30°C.)

#### *Surface roughness measurement*

Profilometry measurements were obtained with a stylus profilometer (KLA-Tencor: P-15) with a 2  $\mu$ m diameter tip. The samples were scanned in three different regions with a scan size of 200  $\mu$ m x 200  $\mu$ m and 1000  $\mu$ m x 1000  $\mu$ m. The 200  $\mu$ m x 200  $\mu$ m scan was taken as an array of 101 traces with 401 points in each trace, and the 1000  $\mu$ m x 1000  $\mu$ m scan was taken as an array of 251 traces with 2501 points. AFM measurements were performed using a commercial AFM (Digital Instruments: Nanoscope IV) in a tapping mode using silicon tips with a diameter of 10 nm. The samples were scanned in three different regions with scan sizes of 20  $\mu$ m x 20  $\mu$ m, 50  $\mu$ m x 50  $\mu$ m, and 100  $\mu$ m x 100  $\mu$ m. The AFM images were captured as arrays of height values with 256 x 256 points. Figure 1 presents 50  $\mu$ m x 50  $\mu$ m scan data from a freshly ground surface with 5 minutes of BCP and the

\* This manuscript has been authored in part 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. It is also funded by DE-FG02-06ER41434. - huit02@jlab.org

same surface after 30 minutes of electropolishing. The former shows very significant peaks and valleys, as well as a very non-uniform surface, whereas the latter is much smoother. The typical measurement of surface roughness is expressed as the root mean square (RMS) roughness, defined by equation (1).

$$R_q = RMS = \sqrt{\frac{1}{n} \sum_{i,j} (z_{i,j} - \bar{z})^2} \quad (1)$$

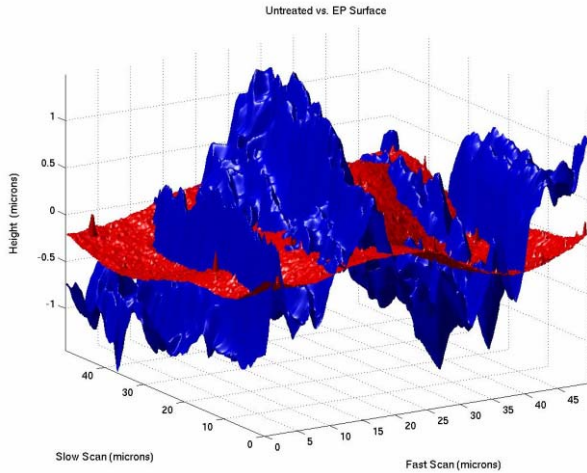


Figure 1: Superposition of typical scans from the Nb surface having received a 5 minute of BCP (in blue), and the surface having received 30 minute EP (in red).

Figure 2 shows a plot of different RMS values computed from the data at all of the scan sizes. While RMS roughness is a useful and widely used measurement for Nb surface roughness, it is dependent on the scan size and the particularities of the area being scanned. It is therefore difficult to compare roughness values taken using different instruments, especially when one considers that the range of frequencies which any instrument can resolve is necessarily limited and unique to that instrument itself [3-5]. Furthermore, this roughness measurement considers only vertical information; it does not give any information about the lateral morphology of surface features.

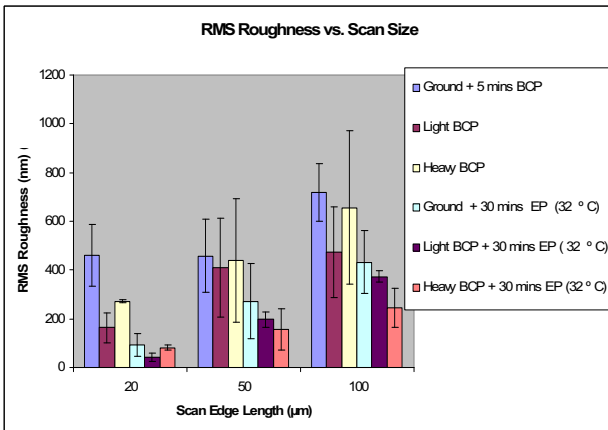


Figure 2: RMS roughness values computed from AFM data taken at different scan lengths.

In order to overcome these limitations, power spectral density (PSD) has been used as a tool to combine measurements from different scales and different instruments [7-8]. The PSD can easily be computed as the square of the Fourier transform as in equation (2).

$$PSD(f) = \frac{1}{L} \left| \int_0^L dx z(x) e^{i2\pi fx} \right|^2 \quad (2)$$

Or, in the case of discrete data consisting of N points separated by  $\Delta x$ , it can be computed as in equation (3).

$$PSD(m) = \frac{\Delta x}{N} \left| \sum_{n=0}^{N-1} z(n) \exp(-i2\pi m n / N) \right|^2 \quad (3)$$

A more complete mathematical treatment can be found in ref [3,5-7]. The PSD represents the squared amplitude of surface features plotted against the spatial frequency of those features. This provides information about both the lateral and vertical size of features. Data from different instruments can also be combined. The range of spatial frequencies over which the PSD is valid for a certain scan size is determined by the Nyquist limits [3, 6, 8]. To simplify the calculations, in this study the power spectral density for all traces in the fast scan direction were averaged to approximate the true 2D PSD. This assumption is valid in the case of a surface with isotropic roughness [8]. For all the samples, all the PSD profiles from a sample were averaged together, and the PSD profiles from the profilometer data were filtered using a 4-sample moving average in order to eliminate high-frequency noise [9].

## RESULTS AND DISCUSSION

### Power Spectral Density (PSD)

Figure 3 shows the combined AFM and profilometer data from the sample with 5 minutes of BCP treatment. The measurements using the two techniques agree very closely over the range of frequencies that are common to both instruments. This shows that the PSD is a method which can combine surface profile information from different instruments in a consistent and useful manner. At the high-frequency end of the spectra, the slope of all the graphs shows a remarkable decrease and begins to level off. This is likely due to the effects of limited resolution for the AFM, and the finite tip size for the profilometer [10]. Of particular interest in this figure is the change of slope that occurs at a spatial frequency approximate  $0.04 \mu\text{m}^{-1}$ . Changes in slope of the PSD are related to the inverse of the correlation length of the sample. The correlation length sets the scale at which surface features are similar [8-9]. For this data, the correlation length is approximately  $25 \mu\text{m}$ , which is on the order of the size of niobium grains in the polycrystalline material studied.

For clarity, the rest of the graphs in this study only show data from the  $1000 \mu\text{m} \times 1000 \mu\text{m}$  profilometer scans and the  $20 \mu\text{m} \times 20 \mu\text{m}$  AFM scans. A comparison of the ground samples after receiving different polishing

can be found in Figure 4. The smaller amplitude of the PSD for the electropolished sample shows that electropolishing produces the smoothest surface, consistent with the results of RMS measurement. For electropolished sample, the slope change at  $0.04 \mu\text{m}^{-1}$  is no longer visible, indicating that differences between grains have been greatly reduced. Furthermore, the greater slope of the PSD from the middle to high frequency region as compared to the other polishing techniques further indicates that EP provides the best polish at micro-range [11]. The PSD profile of the electropolished sample also shows a change in slope at the smallest frequency, corresponding to a surface with the longest correlation length. This also supports the conclusion that EP produces the smoothest surface, since surface features are self-similar on larger scales.

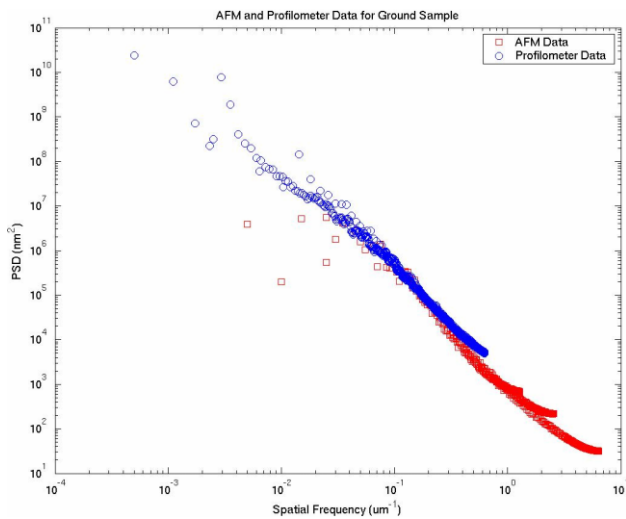


Figure 3: Combined AFM and profilometer power spectral densities from all scan sizes from the sample having undergone 5 minutes of BCP treatment.

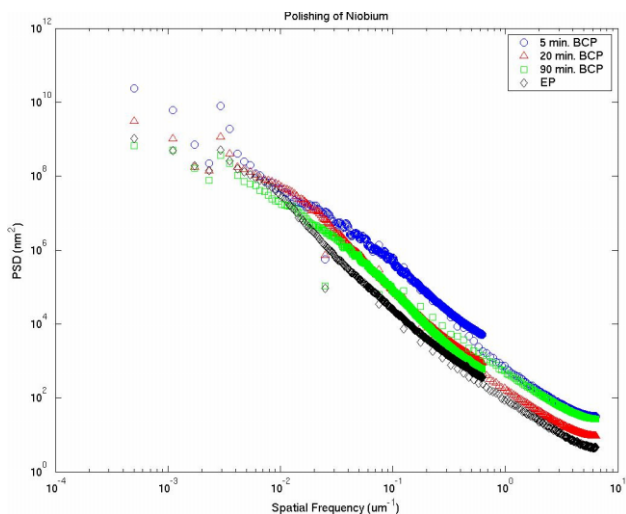


Figure 4: PSD spectra for the different polishing methods performed on a mechanically ground sample.

From this figure, we also found that, interestingly, the light BCP treatment seems to produce smaller PSD

amplitude at low frequencies than that of heavy BCP treatment, but the situation is reversed at high frequencies. This effect would be impossible to analyze using only RMS as a measure of surface roughness; it indicates that a longer BCP treatment produces a better micropolish, but a rougher macropolish. This effect may be due to the differential etching by BCP for grains with different crystallographic orientations [12]. As more material is etched, even though the damage caused by the mechanical polish is removed, material from different grains is removed at different rates, leaving a rougher surface.

Figure 5 as shown below compares the PSD of samples that have undergone different pre-treatments before receiving 30 minutes electropolishing. Light BCP again has the smallest amplitude, and the longest correlation length which corresponds to the smoothest surface. This result is not surprising in the context of the results previously discussed; a longer BCP treatment produces rougher surface at large scales which have not been smoothed out by EP.

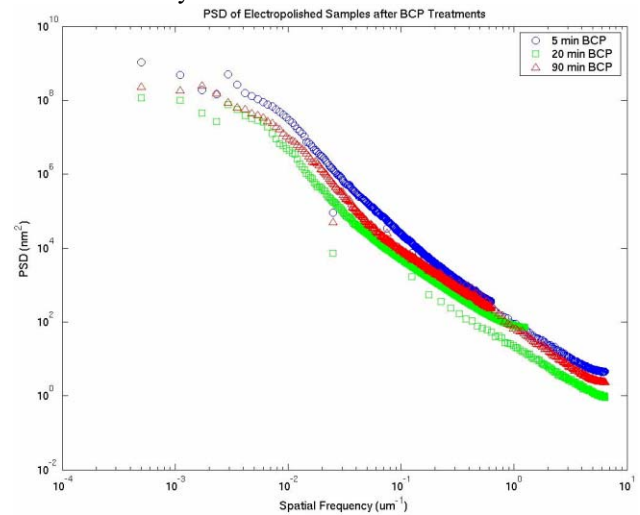


Figure 5: Power spectral density plots of niobium samples that have undergone different treatments.

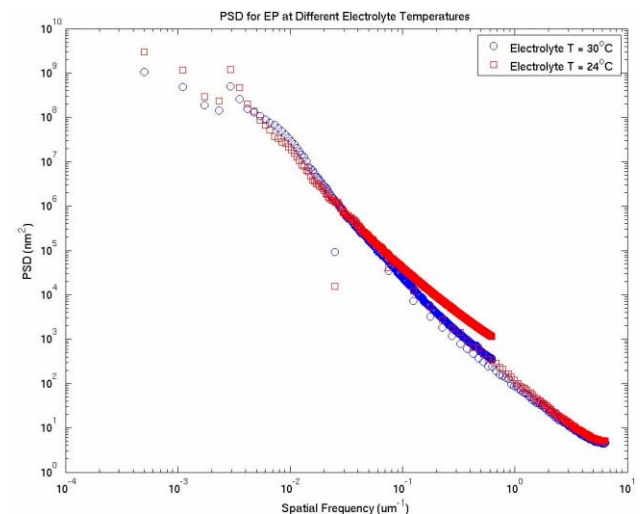


Figure 6: PSD graphs for ground samples electropolished at different temperatures.

Figure 6 presents the power spectra of ground samples treated with EP at two different temperatures. The overlap of the two plots in the high-frequency region indicates that there is no significant difference in the quality of the electropolishing at the small scales. However, there is some evidence for a difference at low frequencies, since the graphs take on a different shape. The longer correlation length of the 24°C sample suggests that the lower temperature may cause a smoother finishing of the sample, as conclusion also supported by the RMS roughness values. They were 84 nm for the 24°C niobium sample and 94 for the 30°C sample, at a scan size of 20  $\mu\text{m}$  x 20  $\mu\text{m}$ . This is consistent with increased etching rate with temperature due to increasing diffusion of F ion [13].

## CONCLUSION

We have demonstrated the use power spectral density in quantitatively characterizing the surface roughness of niobium. PSD measurements allow for the combination of data taken from different instruments and at different length scales. It also allows for a detailed analysis of the surface properties of niobium in both the vertical and lateral dimensions, information otherwise inaccessible to classical methods of quantifying roughness. The power spectral density provides the most information about the surface from the most sources, and so should be the preferred diagnostic when analyzing surface roughness. This tool will therefore allow for improved characterization and analysis in the production of SRF cavities.

We have directly compared the PSD of BCP and EP treated surfaces and noted the scale-dependent difference. Our data also suggests that a lower temperature electrolyte promotes a smoother surface at larger scales.

The implication of Electrochemical Impedance Spectroscopy (EIS) for monitoring Nb surface during electropolishing shows this technology could be potentially used an on-line feedback reported in reference [13]. Correlating PSD and EIS with the expectation that such may be useful as a real-time process monitor. The most important future work, however, will be to determine at what length scales roughness is most important to cavity performance. This information will

allow for the development of a polishing process that deliberately optimizes the performance of SRF cavities and thereby accelerator systems.

## ACKNOWLEDGEMENT

This research was conducted at Thomas Jefferson National Accelerator Facility for the Department of Energy under grants DE-AC05-06OR23177 and DE-FG02-06ER41434 to the College of William and Mary, and also the DOE SULI program. Thanks to Olga Trofimova, Amy Wilkerson and Andy Wu for their support.

## REFERENCES

- [1] H. Padamsee et al., "RF Superconductivity for Accelerators", 1998, John Wiley & Sons
- [2] H. Padamsee, *Supercond. Sci. Technol.* **14**, 2001, p. 28-51
- [3] C. Walsh et al., *Appl. Optics.* **38**(22), 1999, p. 4790-4801
- [4] A. Duparré et al. *Appl. Optics.* **41**(1), 2002, p.154-171
- [5] J. M. Elson, J. Bennett. *Appl. Optics.* **34**(1) 1995, p. 201-208
- [6] "Optics and Optical Instruments-indication in optical drawing", in Draft International Standard ISO 10110 Part 8: Surface Texture, ISO/TC 173/SC 1/WG2 (Inter-national Organisation for Standardisation, Geneva, Switzerland)
- [7] J. Ferré-Borrull et al. *Appl. Optics.* **40**(13), 2001, p 2190-2195
- [8] O. Vatel et al. *Jpn. J. Appl. Phys.* Vol. 23, 1993, p. 5671-5674
- [9] F. Biscarini et al. *Phys. Rev. Lett.* **78**(12), 1997, p. 2389-2392
- [10] K. Westra, D. Thomson. *J. Vac. Sci. Technol. B.* **13**(2), 1995, p.344-349.
- [11] F. Eozénou, A. Aspart, C. Antoine, B. Maliki. CARE Report 06-10-SRF. EU contract number RII3-CT-2003-506395. (2006)
- [12] H. Tian et al. *Appl. Surf. Sci.* **253**(3), 2006, p.1236-1242
- [13] H. Tian et al. WE104, This proceeding.