EFFECT OF SURFACE FLOW ON TOPOGRAPHY IN NIOBIUM ELECTROPOLISHING*

L. Zhao, M. Kelley, Applied Science Dept., the College of William and Mary, VA 23187, U.S.A.
C. Reece[#], Thomas Jefferson National Accelerator Facility, VA 23606, U.S.A.

Abstract

Electropolishing (EP) is reliably delivering improved performance of multi-celled niobium SRF accelerator cavities, attributed to the smoother surface obtained. This superior leveling is a consequence of an etchant concentration gradient layer that arises in the $HF-H_2SO_4$ electrolyte adjacent to the niobium surface during polishing. Electrolyte circulation raises the prospect that fluid flow adjacent to the surface might affect the diffusion layer and impair EP performance. In this study, preliminary bench-top experiments with a moving electrode apparatus were conducted. We find that flow conditions approximating cavity EP show no effects attributable to depletion layer disruption.

INTRODUCTION

Future accelerator project require improved performance from the SRF cavities at their heart [1]. Improved interior surface smoothness is understood to be important, motivating replacement of the established buffered chemical polish (BCP) by electropolish (EP) [2]. Levelling in EP relies on a surface-adjacent mass transfer layer (called δ in what follows) [3], measured as 8 – 20 μm thick with rotating disk electrode (RDE) [4]. It might be vulnerable to fluid flow from rotation of the cavity during polishing and circulation of the electrolyte for cooling. Computational modelling indicates their combined effect can produce surface flow rates up to about 4 cm/s [5].

Our purpose is to gain understanding of how the flow velocity will affect surface roughness after EP. Rotating speed of the samples is selected within the combined range of cavity motion and electrolyte flow.

EXPERIMENT

High purity fine grain Niobium squares, 1.9 cm by 1.9 cm, were buffered chemical polished (BCP) for 1 hour and inserted to a Teflon holder. It was mounted vertically on the edge of a rotating Teflon disk, with a distance of 3.5 cm from the center of the disk. A high purity Al rod with a diameter of 1.27 cm was used as the cathode, mounted vertically as the axis of the disk, with an active area of 10.88 cm^2. 800 ml fresh solution of HF:H_2SO_4=1:10 (vol. %) was used as the electrolyte. EP was conducted at 14V at 20~22°C for 90 minutes. Nb rotates around the vertical axis of Al rod at different speeds of 0 ~ 10 RPM, providing surface flow rate of 0 ~ 3.7 cm/s. Surface roughness and topography of Nb after EP were obtained with AFM, Profilometer and Hirox optical microscope.

RESULTS AND DISCUSSION

Influence of Flow Rate on Current-Time Curve

Figure 1 is the I-t curves collected at different flow rates and the steady state current mean. The first 500 seconds of the curve is unsteady, considering at the beginning of EP the reaction on Nb is complicated. The EP current reaches a plateau after 500~1000 seconds, which means the etching process has reached a steady state. The steady state current is around 0.125 A for static EP, and rises slightly to 0.14~0.16 A for EP at 0.7~3.7 cm/s flow. I-t curve plateau reflects the etching rate of Niobium, therefore Nb removing rate at this rotating speed range is has no significant change.

Influence of Flow Rate on Surface Roughness

Figure 2 shows the average roughness of the three spots on each Nb sample measured by AFM and the roughness at the center of each sample measured by Profilometer. For AFM, three spots were selected near the center of the sample, each with an area of 50 um by 50 um. The average roughness R_q decreased from 80 nm for static EP to around 35 nm for EP under 3.7 cm/s flow. It seems rotating electrode helps improve slightly surface roughness on this scale within this rotating speed range, noticing such a small area is comparable with grain size.

For Profilometer, the roughness was taken from an area of 500 um by 500 um at the center of the sample. No significant difference was found between rotating EP and static EP samples on this large scale and the roughness of all the samples fluctuates slightly between 0.8 um and 1 um. On such a large area the contribution of grain boundaries to roughness is surely included.
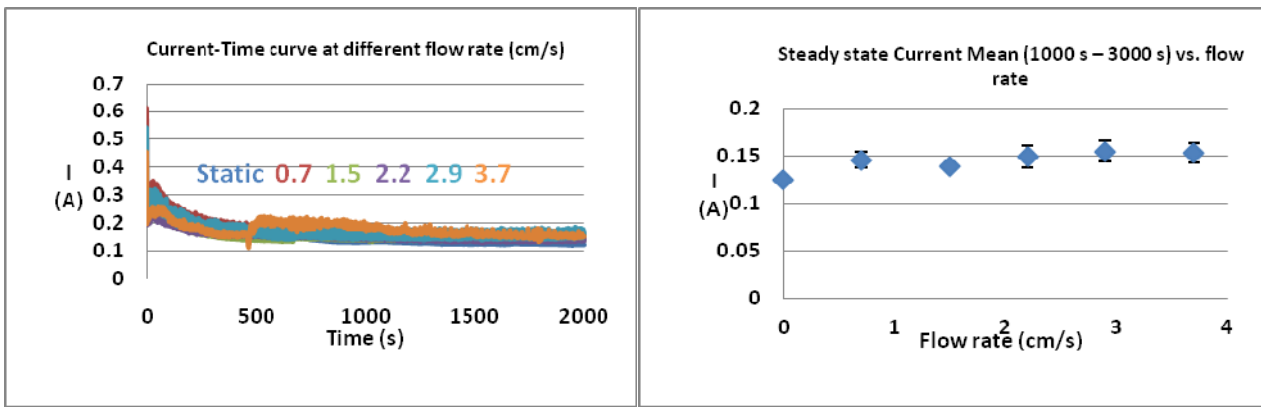
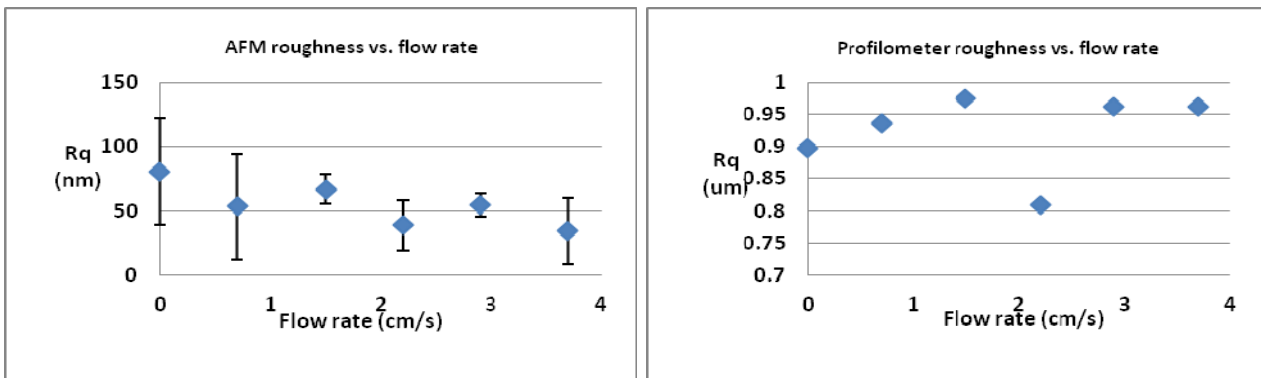Figure 1: Current-Time curves (left) and steady state current (right) at different flow rates from 0 to 3.7 cm/s.

Figure 2: Nb roughness (Rq) after EP at different flow rates, by AFM (50um×50um) (left) and Profilometer (500um×500um) (right).

Influence of Flow Rate on Surface Topography

Figure 3 shows the images of Nb samples after static EP and some typical images after rotating electrode EP, with an area about 320um×240um for Hirox and 50um×50um for AFM. Images were all taken at the center of Nb samples.
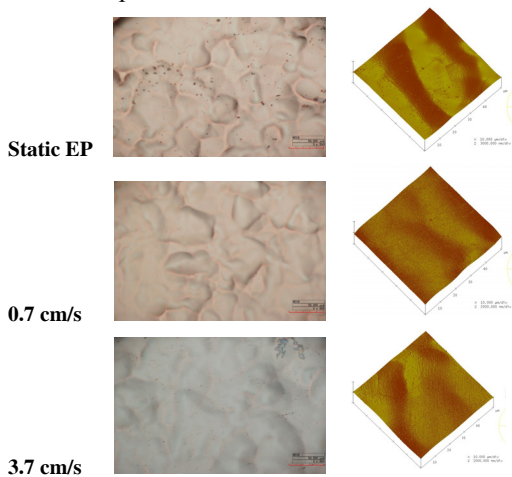

Figure 3: Topography at the center of Nb samples after EP at different flow rates, Hirox images 320um×240um, AFM images 50um×50 um.

On the larger scale Hirox images, the surface sharp edges are smoothed out by EP, and less surface unevenness is observed as flow rate increases, however, the changes were not significant. On the smaller scale AFM images, surface topography variation is not obvious either.

We found that after EP under flow condition, the leading edge (the left edge in our study) of Nb has smaller waviness on macro scope (e.g. eyes and Hirox), but has larger microscope roughness under AFM, as shown in Figure 4. R_q obtained with AFM is 163nm within 1mm from the left edge and 48 nm at the center. This can be a combined effect of electric field and flow that changed the polishing effectiveness at the edge.
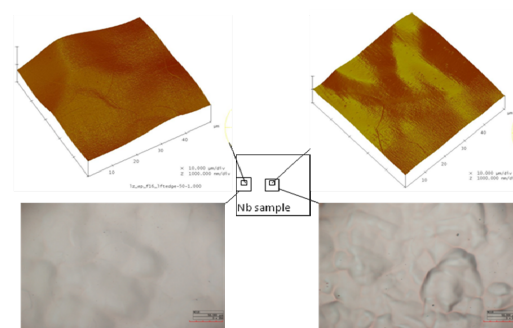

Figure 4: Topography at the leading edge comparing at the center of Nb sample after EP under 2.2 cm/s flow, Hirox images 320um×240um, AFM images 50um×50 um.

Influence of Flow Rate on Diffusion Layer

In static EP the convection is natural movement of the electrolyte due to density and temperature difference (not considered in our case), which means the gradient layer of fluorine (the etchant) is not affected by the electrolyte flow. However, in the rotating electrode EP experiments, convection introduced by flow cannot be ignored. A velocity boundary layer is formed due to the flow. The thickness of this velocity boundary layer on Nb surface due to the flow is [6]

$$\delta_B = 5\sqrt{vy/u_0} \qquad (1)$$

Where: v, coefficient of kinematic viscosity, 29 cP/(1.76 g/cm^3)=1.65×10^{-1} cm^2/s for EP solution at 19 °C[4]; u_0, bulk velocity of electrolyte, here we select the tangential velocity at the center of the Nb samples, which is 0~3.7 cm/s; y, distance from impact point (the leading edge). The flow rate u=0 at the surface of Nb, and u=u$_0$ outside the velocity boundary layer.

Figure 5 shows the calculated thickness of velocity boundary layer on Nb surface during EP at different flow rates. δ_B at the center of Nb sample reduced from 3 cm under 0.7 cm/s flow to 1.3 cm under 3.7 cm/s flow. It turns out to be comparable to the sample thickness, and this is what will happen in low and moderate Reynolds number situation. In our situation Reynolds number is about 70 and a considerable region of the electrolyte can "feel" the movement of the sample.
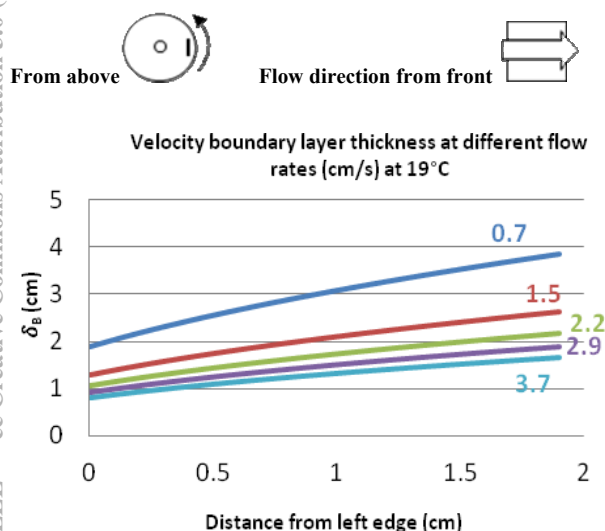
Figure 5: Flow direction sketch and calculated boundary layer thickness on Nb surface during EP at different flow rates.

Inside the velocity boundary layer very close to the Nb surface is the concentration boundary layer, whose thickness is δ. Within this concentration boundary layer mass transport is mainly accomplished by diffusion, and a gradient of fluorine is formed during EP process. The concept of effective thickness of the diffusion layer is

used when decide this concentration boundary layer thickness. According to fluid dynamics and convective mass transfer theories [6], the relation between the thickness of the effective diffusion layer δ and the boundary layer δ_B can be expressed as

$$\frac{\delta}{\delta_B} \approx \left(\frac{D_i}{v}\right)^{1/3} \qquad (2)$$

Where: D_i, diffusion constant of fluorine, about 8.8×10^{-8} cm^2/s at 19 °C [4].

Therefore, δ is about 0.08 δ_B in our study. At the center of Nb samples, δ is 0.24 cm under 0.7 cm/s flow and 0.104 cm under 3.7 cm/s flow. Considering the roughness (R_q) of Nb samples are at the scale of tens of nanometers for AFM and less than 1 micrometer for Profilometer, while the change of diffusion layer effective thickness at the scale of millimetre, which is much larger than R_q, should not have significant influence on the surface roughness, as was observed in our experiment.

CONCLUSION

No significant difference was found in current-time curves obtained at different flow rate within this range. AFM and Profilometer analysis indicate only slight difference on surface roughness of Nb electro-polished at these flow rates. Estimation of boundary layer and concentration boundary layer is coherent with experimental results. The primary EP process is not affected in this flow rate range.

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