

AN EXPERIMENTAL ANALYSIS OF EFFECTIVE EP PARAMETERS FOR LOW-FREQUENCY CYLINDRICAL Nb CAVITIES*

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Abstract

While the basic process of electropolishing niobium with 1:9 HF:H₂SO₄ electrolyte has been well characterized, the specific process parameters used to electropolish different superconducting radio frequency (SRF) cavity geometries requires thoughtful attention. One seeks to realize confidently local diffusion-limited polishing at each point on the surface while maximizing uniformity of removal rate. Since the reaction rate is temperature dependent, this implies that one must manage the cavity surface temperature during polishing. Too-high applied voltage aggravates temperature and thus removal non-uniformity, but too-low applied voltage risks placing the large-diameter locations “off the current plateau,” yielding etching rather than polishing. The majority of recent experience has been with elliptical L-band SRF cavities and some half-wave cavities at ANL. Lower frequency cavities with increased surface area and larger cathode-to-equator distance require fresh analysis and optimization. In preparation for the SNS PPU project, JLab performed some EP process development runs with SNS high beta cavities to help identify viable parameter regimes for communication to cavity vendors. Results from this study are presented.

INTRODUCTION

Substantial experience has been accumulated employing an electropolishing (EP) finishing process for the RF surfaces of niobium superconducting RF (SRF) accelerating cavities. While much of the process development history was empirical, the analytical contributions of Tian and Éozénou yielded insights that now enable system design for reliable effect [1-3]. While the basic process of electropolishing niobium with 1:9 HF:H₂SO₄ electrolyte has been well characterized, the specific process parameters used to electropolish different superconducting radio frequency (SRF) cavity geometries requires thoughtful attention.

The majority of recent experience has been with elliptical L-band SRF cavities and some half-wave cavities at ANL. Lower frequency cavities with increased surface area and larger cathode-to-equator distance require fresh analysis and optimization. In preparation for the SNS PPU project, JLab performed some EP process development runs with SNS high beta cavities to help identify viable parameter regimes for communication to cavity vendors.

To obtain effective electropolishing of Nb with the standard H₂SO₄/HF electrolyte requires that the polishing

surface experience diffusion-limited chemistry. This sustains 25–35 mA/cm² in the temperature range 20–30 °C, based on controlled sample measurements with 1:10 volume ratio of HF(49%):H₂SO₄(96%). This diffusion-limited current density is directly proportional to the concentration of HF in the electrolyte, so actual current depends on solution mixing and prior use details.

To understand an EP system response, it is important to recognize that the applied power supply voltage is divided into three parts: cathode polarization potential (current density dependent), electrolyte potential drop (dependent on current distribution), and anode polarization potential (the balance of the total and locally varying on the cavity surface).

The process with cavities is significantly constrained by the internal geometry which typically drives the use of a concentric cylindrical rod as the cathode. The total current required on the cavity must also be driven through the cathode chemistry. The polarization current requirement for hydrolysis at the cathode is observed to be 1 V per 57 mA/cm² [4]. This property is dominated by the H₂SO₄ concentration. Masking portions of the cathode inherently results in higher cathodic polarization to push the same total current.

The finite conductivity of the electrolyte also reduces the potential available for anodization of the cavity surface, also increasing in significance with larger surface areas and distance from the cathode. This complication increases as the maximum/minimum ratio of diameters of cavity parts increases. Sustained anodization is required for effective polishing.

Lower temperatures slow the diffusion and reaction rates, which in turn lower the integrated current, which lowers the potential drop through the electrolyte and also at the cathode. Thus, a lower applied cell potential (power supply voltage setting) is desirable to realize the intended polishing conditions at the cavity surface. (But too low applied voltage will not be sufficient to create polishing conditions at the larger diameter equators.)

The basic diffusion-limited process is not dependent on the anodic potential once diffusion-limited conditions are established. (This is what produces the “plateau” in a constant-temperature I-V curve.) Higher anodic polarization simply sustains a thicker oxide layer in steady-state. The local heat generated by pushing even a uniform current through this resistive oxide layer does, however, increase with oxide thickness. Unless actively cooled, this heat deposition, which varies with distance from the cathode, results in temperature variation, which increases the local reaction rate and thus the local removal rate and the integrated current. This is what leads to faster removal on the smaller diameter beampipes and iris locations compared to equator regions. Thus we have the motivation

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to control the cavity wall temperature via external water cooling.

CONSIDERATIONS FOR HB CAVITIES

As part of the SNS PPU Project R&D, JLab was asked to support the “Conceptual Development of Processing and Testing Recipe for PPU Cavities.” The SNS cavities HB-071, HB-072, and HB-073 were received at JLab and processed nominally according to the requested directions, with concurrent analysis and comment toward process improvement.

An initial attempt was made to qualify an EP procedure which did not involve external water spray cooling so as to provide a path for potential vendors to employ their existing capability. This attempt was not successful. The cold acid, 10 V process resulted in etching rather than polishing at the equators. The rf test showed significant Q -slope at 8–9 MV/m without radiation. Subsequent reprocessing of this cavity with 17 V applied with external waterflow cooling yielded clear polishing of the equator regions. A more structured analysis was applied to the situation.

Cathode Issues

A 1” diameter aluminum cathode used for SNS HB 6-cell cavity, if fully unmasked with 125 cm exposed length, has an active area of 997 cm². A cavity processing that draws 350 A (which results in 350 mA/cm² current density on the cathode), will then require 8.4 V of the applied potential just to push the current through the cathode [4]. The potential drop at the cathode would also increase the generation of sulfur [5]. For the HB cavity design, with a 14,000 cm² internal surface area, this condition corresponds to ~25 mA/cm² avg. The problem accounting for cathodic polarization is increasingly significant with larger surface area RF structures with the present typically used small diameter Al cathode design. (With lower HF concentration that results in 16 mA/cm² average, the total current is 225 A and requires 6.0 V cathode polarization, at ~20 °C.) Increasing the cathode surface area in these cavities with larger clear aperture would reduce this problem. Masking portions of the cathode increases this problem [6].

Electrolyte Conduction Issues

The resistivity of the typical Nb EP electrolyte has been measured to range from ~13 Ω-cm²/cm to ~8 Ω-cm²/cm between 20 °C and 32 °C [4]. In a cavity, the current distribution is very non-uniform, but we may approximate that the iris radius to equator radius of an HB cavity is 4 to 16 cm. So, it may not be unreasonable that a potential drop variation of several volts may exist through the electrolyte between iris and equator of a large (SNS HB) cavity. Such would create a condition of effective polishing only at the irises, but not at the equators. (Potential drop variation when the cell potential is not large enough may create the desired diffusion controlled process at irises, but not at the equator, which may be still under polarization control—*etching*. In the etching condition, there is no persistent

oxide layer and local reaction rates vary for different niobium grain faces presented at the surface [4]. This results in faceted topography with sharp corners at grain interfaces—bad for high-field SRF performance. In addition, in the “etching” condition, there is no sustained oxide on the niobium surface, so the natural barrier to hydrogen diffusion into the bulk is removed. This then also creates a mechanism for loading the cavity niobium with hydrogen, greatly increasing the vulnerability to “ Q -disease” from hydride formation during a slow transition through the vulnerable precipitation temperature range ~100 K.

Current vs. Temperature Issues

For a given cathode surface area with a large surface area cavity, too high a current draw may starve the equator regions of adequate polarization potential. Higher applied power supply voltage is required, but without temperature control via external cooling, the local current will increase via small-radius heating and aggravate removal uniformity and increase generation of sulfur precipitation (which becomes a source of field emission if not thoroughly removed). The S generation may not be avoidable with large cavity/cathode surface area ratios and practical polishing rates; increasing the surface area of the cathode as much as possible within constraints imposed by the cavity structure will help minimize sulfur production.

So, for SNS HB cavities, one should expect that currents of order 220–350 A will be needed to obtain the required current density for full-surface polishing (plateau current), and at least 14 V power supply potential should be needed. Experimental confirmation of the plateau condition is needed for each EP system configuration. High currents will present greater opportunity for sulfur generation than with smaller cavities. The large equator/iris aspect ratio makes temperature control of the irises and endgroups more of a challenge than with all smaller cavities. Unrestricted cooling water flow should be assured in these regions.

EXPERIMENTAL TEST RUNS WITH HB CAVITIES

Low Voltage Run

For the purpose of potentially identifying a suitable EP method for HB cavities which does not employ external cooling water, a test run was performed on HB-071. This run attempted to minimize heat production by using an applied voltage of 10 V and a supply sump temperature of the circulating electrolyte of 10 °C. Recent EPs of other cavities have been quite successful using an applied voltage of 10 V. Any voltage higher than that necessary is understood to only generate additional heat, complicating removal uniformity and cathodic potential drop from high currents. (Very successful EP of the SNS MB cavities was previously accomplished at JLab with 13 V and external water spray cooling.)

The cathode was fully masked outboard of the endcell irises, otherwise unmasked. This kept the cavity equators

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~27 °C and beampipes ~28 °C. The process parameters are illustrated in Fig. 1. The process appeared well behaved, with very attractive temperature uniformity and moderated current ~ 180 A.

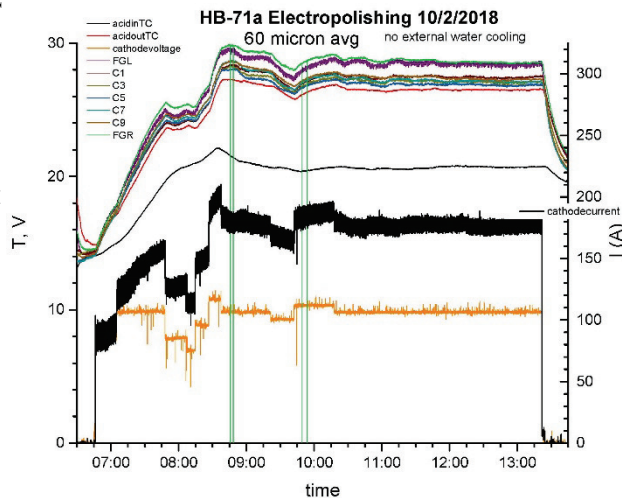


Figure 1: Low-voltage processing run.

The cavity cryogenic test, however, was disappointing (Fig. 2). While the low-field Q was excellent, the cavity showed evidence of Q -slope at very low field (beginning 8-9 MV/m) in the accelerating mode and also in several other passband modes. An exponential Q decline beginning ~ 40 mT B_{pk} . This indicates a phenomenology of limitation consistent in all cells. It was interpreted as failure of polishing in all cells.

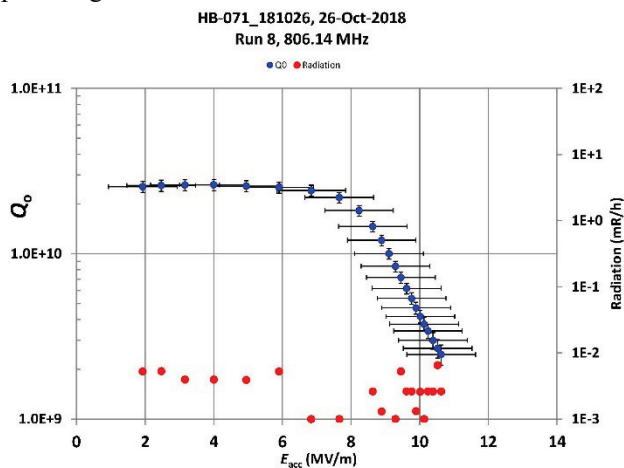


Figure 2: Cold RF test after low-voltage processing.

Subsequent internal inspection of HB-71 with the JLab Kyoto camera system confirmed the above interpretation. Good polishing was observed at the irises and radially outward to at least the position of the stiffening ring welds. At the radii of the equators, however, the surface was found to be very strongly etched, showing crystallographic structure in high relief—definitely not good for high-field SRF performance. The prompt step changes in current at changes in the applied voltage in Fig. 1 are also direct evidence that the desired “plateau” polishing conditions are not being met.

Table 1: Process Parameters for HB-71b

Acid sump temperature	10 °C
Acid circulation rate	~6 l/min
Applied cell voltage	17 V
Cathode condition	unmasked
External water cooling	aggressive
Cavity wall temperature	24–28 °C
Current	210 A
Average surface removal	60 μ m

High Voltage Run

The second pass of JLab EP on HB-071 used the parameters listed in Table 1 and demonstrated excellent polishing of the equator regions while actively managing the heat generation via cooling water in order to keep the current well controlled for uniform polishing. The desire to assure that a vendor’s EP setup would necessarily yield electropolishing in the “plateau” regime at the high magnetic field equators, led to the selection of 17 V applied voltage with fully unmasked cathode used for the second-round EP applied to HB-71. The unmasking of the cathode was done to increase assurance that the HOM coupler cans would be polished rather than etched. This will be likely unnecessary for the PPU HB cavities, since they do not have the HOM couplers. The process parameters are illustrated in Fig. 3.

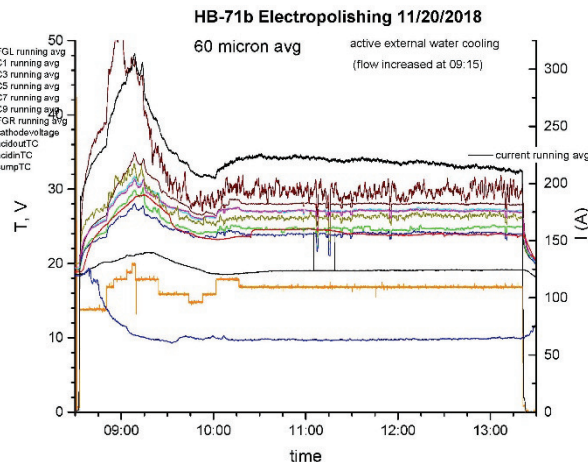


Figure 3: High-voltage electropolishing run.

Figure 4 compares same-scale views of HB-71 equator welds after the initial low voltage, no cooling water EP run (etching) and the 2nd run (good EP) taken with the JLab Kyoto camera. The finished surface would be smoother yet had the etching not taken place in the earlier run. Sharp corners are, of course, bad for RF due to local magnetic field enhancement which create local field $> H_{c,sh}$.

The left side beampipe (FPC end) was not well cooled, as indicated by the thermocouple FGL. (After ~9:15 am, the attachment of this sensor to the cavity was compromised reducing the integrity of subsequent data.) The measured removal at in this area was also more than double the rest of the cavity. By total charge passed, the net average removal was 60 μ m over the entire surface.

Removal thickness measurements were made at the same locations as the temperature sensors. These locations and measured removal amounts are depicted in Fig. 5.

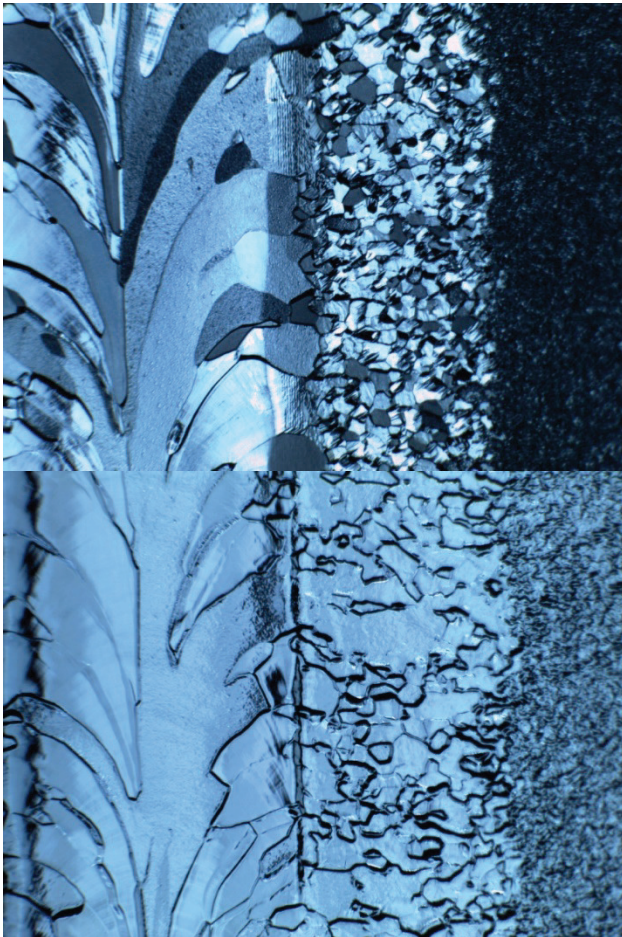


Figure 4: Equator weld region of HB-71 after the 10 V/ no cooling and 17 V/ water cooling EP runs.

The indicator that desired polishing is occurring over the entire surface is a “plateau” in the IV curve, i.e. increased voltage yields no increased current. This is strictly valid only if constant temperature is assured. This is what drives the need for independent external cooling. For the configuration used for HB-071b, the power supply applied voltage was varied between 14 and 18 V in an attempt to probe for this “plateau” condition. Fig. 6 shows the logged average temperature of the monitoring thermocouples and the smoothed current plotted against the applied voltage. The data are consistent with an interpretation that 14 V and higher indeed satisfied the “plateau” condition.

Analysis of the data from the HB-071b EP run indicated that for the conditions used, 15 V applied voltage would have satisfied the desirable “plateau” condition, while 10 V clearly did not.

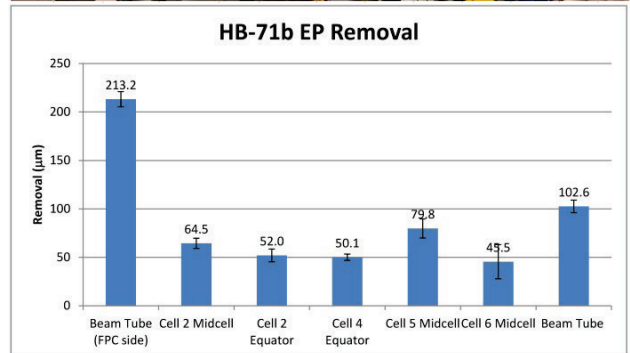
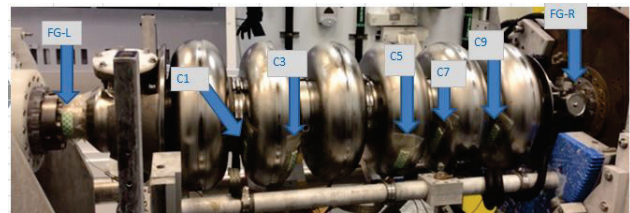


Figure 5: Temperature monitoring and thickness measurement locations.

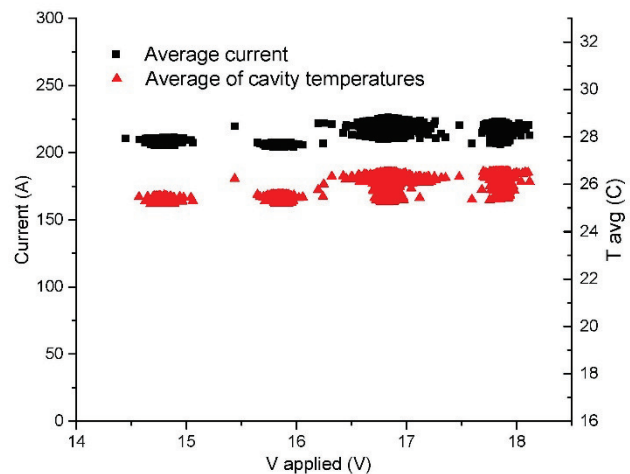


Figure 6: Current and average temperature versus applied voltage for HB-71b run, demonstrating “plateau”.

Hydrogen Loading

Due to happenstance, during the cooldown for RF test of HB-71 after the second EP, the cavity spent over 1 hr in the window of 80-120 K. The subsequent 2 K RF performance clearly reveals significant “Q-disease.” See Fig. 7. We interpret this to indicate that the “off-plateau” etching, not only significantly roughened the surface, but also enabled significant hydrogen loading into the bulk due to the absence of a sustained surface oxide. This cavity would require vacuum heat treatment for hydrogen degassing prior to any application use.

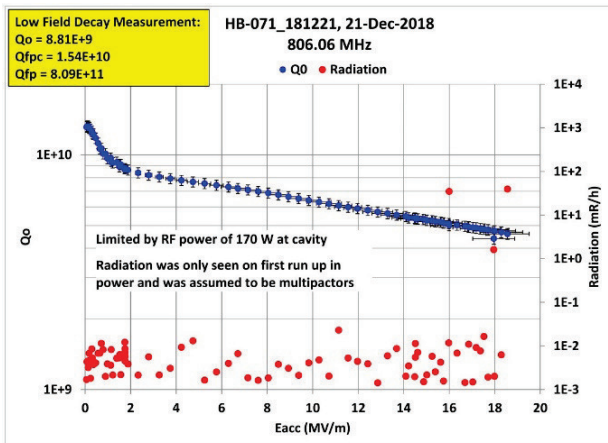


Figure 7: Q-disease performance of HB-71 with hydrogen loading from defective polish process.

FURTHER IMPROVEMENT OPPORTUNITIES

A few specific opportunities for improvement not yet implemented on HB cavities have been identified:

1. Relocate cavity electrical contacts from the stiffening rings to equators. The straps around stiffening rings block cooling water flow to the iris—where it is most needed.
2. Ensure generous cooling water flow to both endgroups to avoid excessive removal due to heating. (In this run, the cooling water to the left (FPC side) beamtube was weakly supplied.)
3. Increase cathode surface area via the use of thicker-walled Al pipe with added grooving. This could easily be done with the HB cavities while preserving existing mechanical interfaces.

CONCLUSION

It is quite clear that the translation of “successful” niobium cavity electropolishing procedures from one setup to another and one cavity geometry to another must be done with careful analysis. The underlying electrochemical processes are both non-linear and scale dependent.

The general advice, which also aligns with common commercial electropolishing experience, is to maximize the usable cathode working surface area and stabilize the cavity temperature to maximize uniform polishing and removal rate.

ACKNOWLEDGEMENTS

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