ISBN: 978-3-95450-211-0 doi:10.18429/JACoW-SRF2019-THP002

METALLOGRAPHIC POLISHING PATHWAY TO THE FUTURE OF LARGE SCALE SRF FACILITIES*

O. Hryhorenko[†1], C.Z. Antoine[2], M. Chabot[1], D. Longuevergne[1]
[1]Institut de Physique Nucléaire d'Orsay, CNRS-IN2P3,
Université Paris-Sud, 91400 Orsay, France
[2]Service des Accélérateurs, de Cryogénie et de Magnétisme,
CEA-IRFU, 91191 Gif-sur-Yvette, France

Abstract

Optimization of SRF cavities mainly focuses on pushing the limits of bulk Niobium, cost reduction of cavity fabrication and development of new SRF materials for future accelerators (ILC, FCC). Nowadays chemical etching is the only surface treatment used to prepare SRF surface made of Nb. However the operational cost of chemical facilities is high and these present a very bad ecological footprint. The search of an alternative technique could make the construction of these future large scale facilities possible. Metallographic polishing (MP) is a candidate not only for bulk Nb treatment, but could also provide the mirror-finished substrate for alternative SRF thin films deposition. Recent R&D studies, conducted at IPNO & IRFU, focused on the development of 2-steps MP procedure of Nb flat samples. Roughness of polished surface has been proven better than standard EP/BCP treatment and less polluted than CBP. MP provides on flat surfaces a high removal rate (above 1 μm/min) and high reproducibility. The paper will describe the optimized method and present all the surface analysis performed. The first RF characterization of a polished disk will be presented.

INTRODUCTION

Standard production processes of SRF cavities (rolling of Nb ingots, forming of half-cells and electron beam welding) cause the appearance of damaged layer of the order of 100-200 μm into the inner surface of the material [1]. Damaged layer has to be removed in order to recover optimum superconducting properties and avoid limited performances. Moreover high electro-magnetic fields (up to 50 MV/m and 200 mT) penetrate typically through a thin layer of Niobium inducing a high current density (up to 10^{12} A/m^2) decaying exponentially over several hundreds of nanometres into Nb bulk [2]. Due to the reasons mentioned above, good superconducting properties of the material have to be preserved not just on the surface layer, but also over a few hundreds of nanometers. The standard Nb preparation for SRF applications are either buffered chemical polishing (BCP), electropolishing (EP), or centrifugal barrel polishing (CBP) in combination with light chemistry (10-20 μm of EP) [3, 4]. In standard pathway of cavities production the surface polishing is applied on the closed geometry after forming. However in order to find an alternative to the heavy chemistry and to possibly reduce the cost of cavity fabrication, mechanical polishing may be applied on flat sheets before forming steps. It has been shown that the observed damaged layer after forming is significantly thinner (~ 10 μm) than after rolling step of Nb sheets and would only require a flash BCP or EP [1].

Metallographic polishing (MP) is a sub-type of mechanical polishing (for example as CBP) used for metal, wafer, optical lenses and others material preparation [5]. C. Z. Antoine initiated the investigations of metallographic polishing for SRF purposes [6]. This article will describe the advances done at IPNO and CEA on this specific topic.

METALLOGRAPHIC POLISHING REQUIREMENTS FOR SRF

So as to ensure optimal superconducting properties and also to be competitive to standard polishing processes for SRF the following requirements have been defined:

- Remove polluted and damaged layer induced by Nb sheets fabrication (lamination of niobium ingots and rolling of sheets).
- Preserve superconducting properties of material over few hundreds of nanometers.
- Limit the number of steps to 2 or 3 (instead of 5-6 steps in metallographic polishing) to allow industrialization of process and to be competitive compared to standard techniques of the treatment (BCP - 5 hours, EP - 8 hours and CBP - weeks).

EXPERIMENT

Experimental Set-up and Specimen Preparation

Metallographic polishing has been performed on a machine MasterLAM 1.0, see Fig. 1, manufactured by a French company LAM PLAN [7]. Polishing disks may be charged with various type of abrasives (diamonds, silica carbide, aluminium oxide and etc.) with different size (from 100s microns down to nanometers scales. Abrasives could be embedded in the polishing disk or added as a liquid suspension between the disk and the specimen.

In both cases material removal is due to the combined actions of normal and tangential applied forces. The normal force defines the depth of abrasives penetration into the bulk

* Work supported by the European Nuclear Science and Applications Research - 2 (ENSAR-2) under grant agreement N°654002.
† hryhorenko@ipno.in2p3.fr

ISBN: 978-3-95450-211-0 doi:10.18429/JACoW-SRF2019-THP002

Figure 1: Metallographic polishing (MP) device (LAM PLAN company).

Nb and is controlled by a pneumatic adjuster on the centre of sample holder. The tangential force creates a cutting effect and results in the material removal. The efficiency of material removal (removal rate) depends on: hardness of used abrasives, carrier of the abrasives (metal, resin or cloth), applied pressure, rotational speeds of holder and polishing disk.

The rotational speeds of the sample holder and the polishing disk were 150 and 300 rotations per minute (RPM). Working pressure is typically set to 305 g/cm^2 corresponding to a pressure of 30 kPa for 3 samples (10 x 40 mm). The polishing procedure was optimized on small flat specimens (10 mm x 40 mm) and than the recipe has been transferred to Nb disks of diameters of 50 and 126 mm.

In order to remove the influence of the specimen preparation history all specimens before MP were etched by BCP mixture to 1:1:2.4 (HF:HNO_3:H_3PO_4) to remove about of 200 μm of the surface layer. Fig. 2 shows topography of Nb surface after BCP treatment with $S_a = 1 \pm 0.25\mu$m and $S_z = 16.4 \pm 2.75\mu$m.

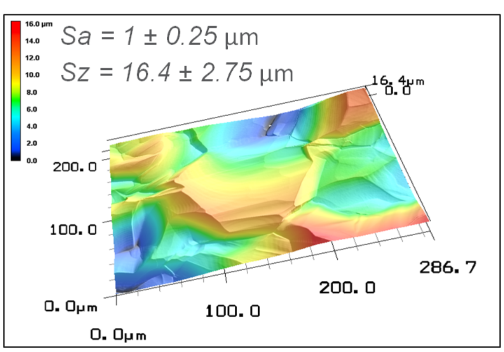

Figure 2: 3D reconstruction of Nb surface state after BCP treatment.

Two Steps Polishing Recipe

So as to be competitive the number of polishing steps has been significantly and successfully decreased down to 2 steps.

First step, called the abrasion step, has to planarize the surface and remove the polluted and the damaged layer created due to the rolling process. A compromise has to be done in order to have at the same time a high removal rate (μm/min), low surface roughness, low damaged and polluted layer. Abrasion step requires to remove few hundreds microns of the material. Second step (may consists from additional sub-step), named the polishing step, has to remove damages and depollute (embedded abrasives) the surface after abrasion step and achieve surface average roughness $S_a < 0.1$ μm.

Step 1. A suspension of polycrystalline diamonds of 3 microns in grain size in combination with rigid composite disk have been chosen for abrasion step. Approximately 100 μm layer has been removed within 90 minutes in order to simulate the removing of the damaged layer. Removal rate corresponds to 1.1 μm/min (constant during abrasion step) to be compared to BCP - 1 μm/min, EP - 0.5 μm/min and CBP - some μm/hour.

Step 2. Polishing step has been performed by a special solution composed of 50 nm grain sized particles of colloidal silica (SiO_2, pH = 9) diluted in the deionized water (20 %), carried on microporous polyurethane cloth. In this case, polishing of Nb material happens due to the combined action of the mechanical and the chemical action (chemical-mechanical polishing (CMP)). Removal rate of material corresponds to 0.01 μm/min (constant during polishing step), however this parameter could be increased with increased concentration of colloidal silica in solution.

As shown in Fig. 3, mirror finished surfaces have been obtained for all Nb samples by using 2 steps polishing recipe.

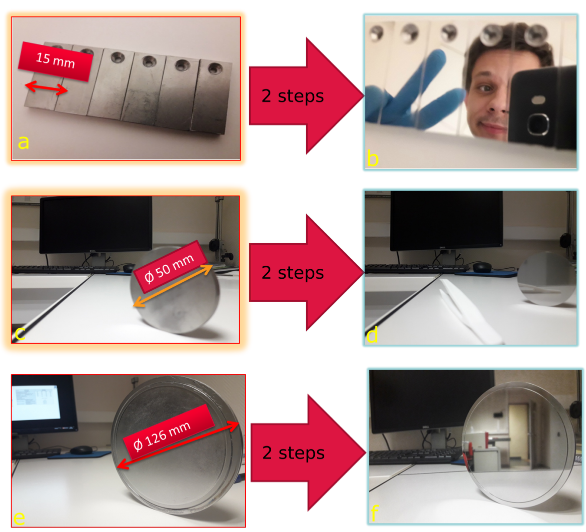

Figure 3: Raw Nb specimens before metallographic polishing (a-rectangle sample, c-disk of 50 mm diameter, e-disk of 126 mm diameter). Mirror finished specimens after 2 steps polishing recipe (b-rectangle sample, d-disk of 50 mm diameter, f-disk of 126 mm diameter).

ISBN: 978-3-95450-211-0 doi:10.18429/JACoW-SRF2019-THP002

SURFACE ANALYSIS

Roughness Characterization

The surface roughness has been measured by the laser confocal microscope (Keyence VKX-200). To reduce impact of local defects and to ensure sufficient statistics a set of 10 measurements at different positions of each sample have been performed. The average roughness (S_a) and the maximal height difference (highest peak and deepest valley) S_z are measured. S_z is an important local parameter giving in our case the deepest scratch caused by the abrasion step and thus, the minimal thickness to be removed by polishing step.

Figure 4 depicts the evolution of the average surface roughness after each abrasion and polishing step. The abrasion step (first 90 minutes in Fig. 4) removes the polycrystalline structure created by BCP, planarizes the surface, creates amorphous structure that will increase the efficiency of the following polishing step. Unfortunately this step leaves embedded particles all over the surface.

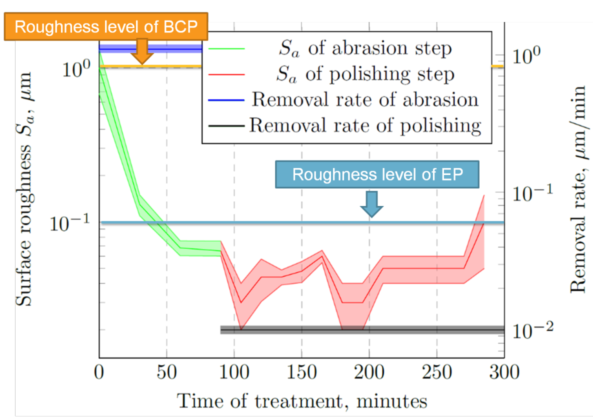

Figure 4: Evolution of the average surface roughness as a function of time.

The best average surface roughness is achieved after 1 hour of abrasion and remains constant if processed for a longer time. Average surface roughness has been saturated after 1 hour of abrasion. Fig. 5 shows final topography of Nb surface after the abrasion step with $S_a = 68 \pm 11$ *nm* and $S_z = 3.27 \pm 1.09$ μm.

The polishing step reduces the roughness down to 30 nm after 20 minutes, but tends to reveal some roughness of the surface with time due to reappearance of grains because of the combined chemical-mechanical (CMP) action. However polishing step can't be stopped because of embedded abrasives remaining in the material. For this reason polishing step has to be extended at least up to 200 minutes. Fig. 6 shows the final topography of Nb surface after 200 minutes polishing step with $S_a = 100 \pm 50$ *nm* and $S_z = 2.58 \pm 0.426$ μm. As seen in Figure 7, all surface damages and embedded abrasives are removed.

The control of surface roughness is important, however, not sufficient as chemical composition and microstructure

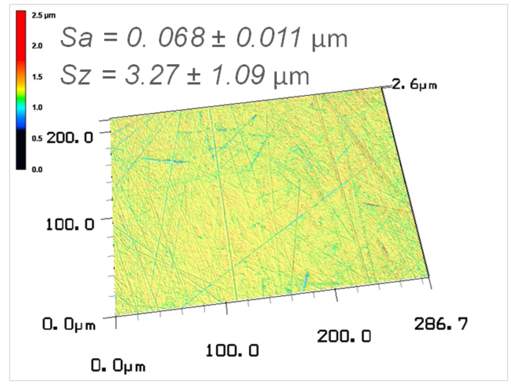

Figure 5: 3D reconstruction of Nb surface state after abrasion step of MP.

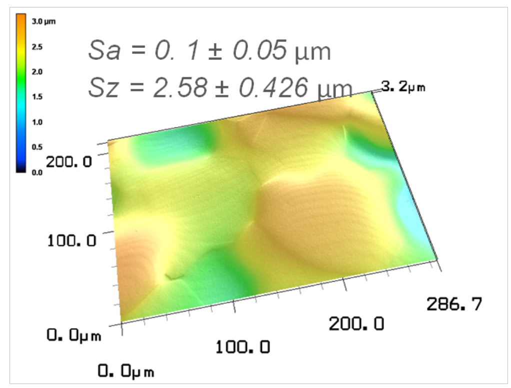

Figure 6: 3D reconstruction of Nb surface state after polishing step of MP.

of the surface layer are required to be comparable with SRF standards.

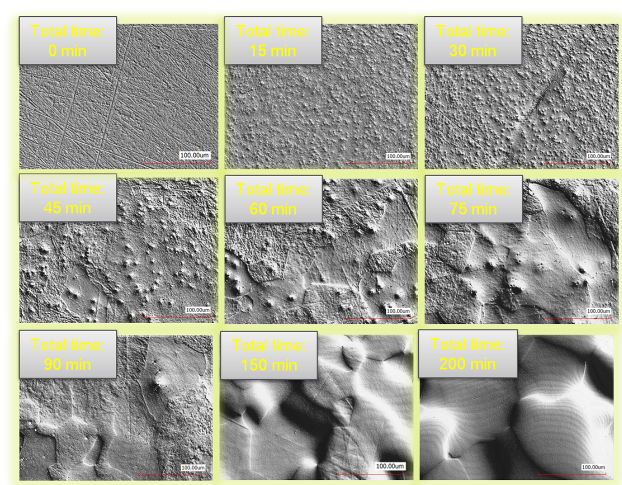

Figure 7: DIC images of Nb surface state during MP polishing in different periods of time. Note: figure shows also the de-pollution effect, grain formation and grain growing with time.

ISBN: 978-3-95450-211-0 doi:10.18429/JACoW-SRF2019-THP002

Damaged Layer Characterization

In order to characterize the damaged layer created by MP procedure, investigations of the polished specimens have been performed with two methods: multi-step BCP followed with optical analysis and electron backscattered diffraction (EBSD) on SEM microscope.

Etching process during the BCP treatment tends to reveal not only the grain structure, but also subgrain damages caused by mechanical processes. In order to estimate the depth of affected layer (high density of dislocations), the surface has been observed after multiple BCP etching of 5 μm (confocal imaging in DIC mode). Sub-grain structures are visible and presented in Fig. 8. Sub-grain patterns present in the affected layer, disappear approximately after 85 μm of BCP.

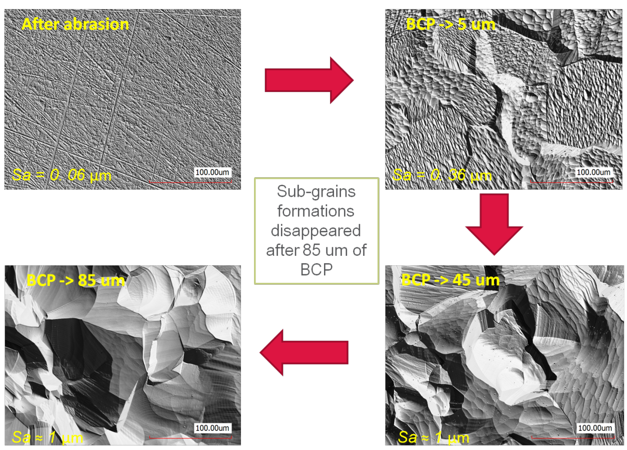

Figure 8: Etching figures of Nb surface after abrasion step.

Evaluation of affected layer has been done thanks to another method. This one involves EBSD analysis performed on the cross-section of the sample. In that sense this requires some additional surface preparation (cutting, mounting in resin and cross-section polishing). Figure 9 shows kernel average misorientation (KAM) and inverse pole figures (IPF) of polished face after 2 steps polishing procedure (right) as described before and secondly cross-section after abrasion step (left).

As EBSD analysis is typically sensitive to the first hundred of nanometeres under the surface, we can conclude that 2 steps polishing procedure is causing very limited residual crystal damages. Indeed this IPF image (right) is showing crystallized surface comparable to bulk unaltered crystal. On the other side, the cross-section analysis tends to show some surface damages (higher dislocation density at the edge of sample). However, these damages have been more likely created during the preparation of the cross-section (cutting procedure) rather than from 2 step polishing procedure of the face. Additional analysis will be carried out to confirm this statement.

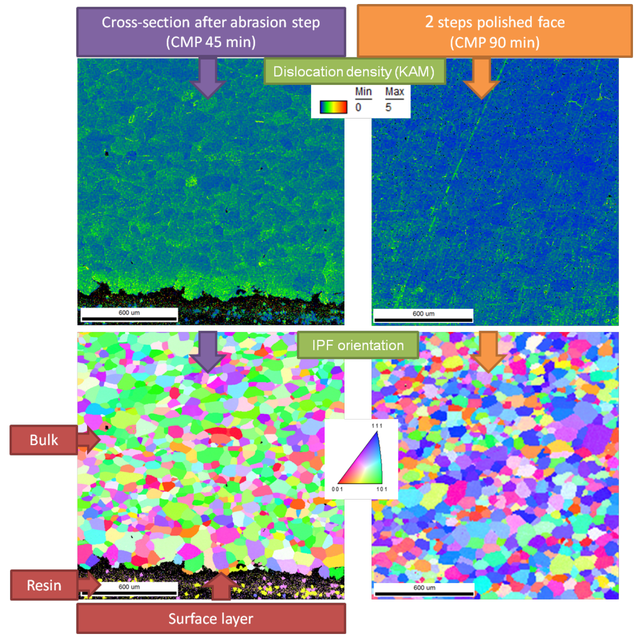

Figure 9: Kernel Average Misorientation (KAM) and Inverse Pole Figures (IPF) of cross-section after abrasion step (left, CMP 45 minutes) and polished face after 2 steps polishing (right, CMP 90 minutes).

RF Characterization

RF measurements have been done at SLAC (Stanford Linear Accelerator Center) in a hemispherical cavity which operates in pulse mode at high frequency (11.4 GHz) and at cryogenic temperatures between 12 K and 3.8 K [8]. The test bench gives the possibility to mount flat disks with diameter of 50.8 ± 0.3 mm. Two samples were prepared at IPNO, one with 2 steps CMP polishing recipe and the other with standard BCP used as a reference (samples with thicknesses of 3 ± 0.1 mm).

The quality factors of both samples (CMP and BCP) are shown in Fig. 10. At 9.2 K both samples show a sharp increase of quality factor as a consequence of the transition of Nb material from normal to the superconducting state. However, a lower quality factor is observed below 4 K after CMP process, indicating the presence of surface pollution or damages.

Fig. 11 shows the behaviour of extracted surface resistance versus T_c/T ratio. Measured surface resistance consists of the sum of the BCS surface resistance (ideal Nb) and the residual resistance (presence of the impurities in Nb). As shown in Fig. 11, the BCS model predicts that the surface resistance should be well below measured values (at 4 K and at frequency 11.4 GHz the BCS resistance would be typically of the order of 80 $\mu\Omega$). However the tested samples, even the BCP treated Nb shows a very early saturation of the surface resistance caused by a very high residual resistance (2 mΩ). These early stabilizations of the surface resistance for both samples may be the sign of the Q-disease (as samples were

ISBN: 978-3-95450-211-0 doi:10.18429/JACoW-SRF2019-THP002

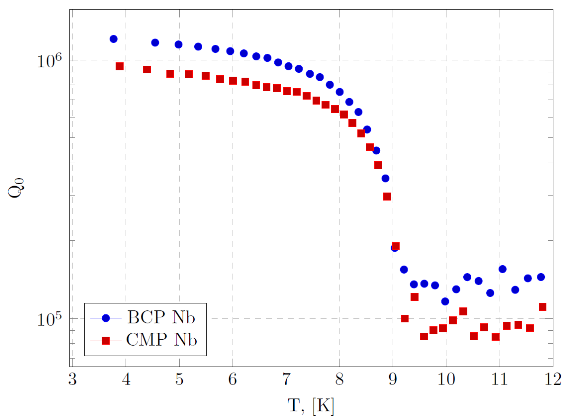

Figure 10: The quality factors of the BCP (blue curve) and the CMP (red curve) polished disks as a function of temperature.

not degassed in a furnace before RF test) or the existence of anomalous dissipation (RF contacts, lossy contacts...).

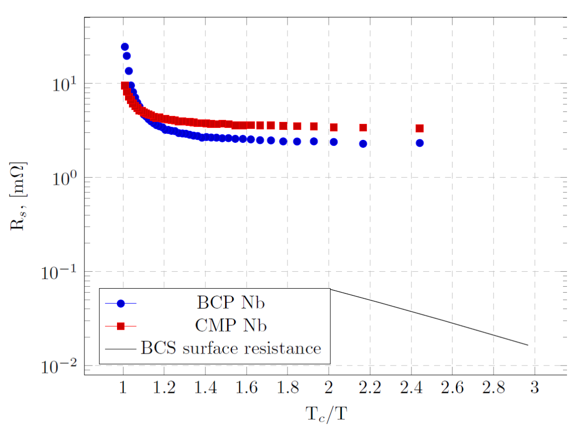

Figure 11: The surface resistance of the BCP and CMP samples versus T_c/T.

CONCLUSION

A two-steps polishing procedure was successfully developed at IPNO and applied on RF disk. Optical characterizations have been performed showing a non-polluted and non-damaged surface. Not only the quality of the surface is important, but also the material quality underneath over a depth of several hundreds of nanometers. Hence, optical quality is necessary but not sufficient. Thus, the ultimate surface characterization testifying of the quality of this polishing procedure is the RF test at cryogenic temperature.

This test performed at SLAC showed very promising results. Unfortunately, because of technical limitations inducing a very early saturation of the surface resistance, the decay of superconducting BCS resistance with T_c/T ratio couldn't be measured properly. A way to mitigate these "high frequency" limitations would be to test a sample at a lower frequency, as on IPNO test bench as soon as this one would be available (the sample disk is ready to be tested).

ACKNOWLEDGEMENTS

The author would like to thank the great work of SUPRATECH team for the high quality work performed for surface preparation of reference BCP sample (in the standards for spoke cavities treatment). Moreover we would like to thank P. Welander (from SLAC laboratory) for the performed RF test and J.M. Jeoffroy (from LAM PLAN company) for the his helpful advices on various technical issues connected with the metallographic tasks. This work was supported by the European Nuclear Science and Applications Research - 2 (ENSAR-2) under grant agreement N°654002.

REFERENCES

[1] Antoine, Claire. Materials and surface aspects in the development of SRF Niobium cavities. No. EuCARD-BOO-2012-001. 2012.

[2] R. Carter, *Reviews of Accelerator Science and Technology: Volume 5–Applications of Superconducting Technology to Accelerators*: Contemporary Physics, 2015, pp. 392–393. doi:10.1080/00107514.2015.1005681

[3] V. Palmieri, F. Stivanello, S. Y. Stark, C. Roncolato, and M. Valentino, "Besides the Standard Niobium Bath Chemical Polishing", in *Proc. SRF'01*, Tsukuba, Japan, Sep. 2001, paper PR016.

[4] C. A. Cooper, B. Bullock, S. C. Joshi, A. D. Palczewski, and K. Saito, "Centrifugal Barrel Polishing (CBP) of SRF Cavities Worldwide", in *Proc. SRF'11*, Chicago, IL, USA, Jul. 2011, paper WEIOA02, pp. 571–575.

[5] Geels, Kay, *et al.*, "Metallographic and Materialographic Specimen Preparation, Light Microscopy", in *Image Analysis, and Hardness Testing*: ASTM International (2007).

[6] C. Z. Antoine and R. Crooks, "Reducing Electropolishing Time with Chemical-Mechanical Polishing", in *Proc. SRF'09*, Berlin, Germany, Sep. 2009, paper TUPPO071, pp. 405–405.

[7] https://www.lamplan.com/fr/

[8] P. B. Welander, M. A. Franzi, and S. G. Tantawi, "Cryogenic RF Characterization of Superconducting Materials at SLAC With Hemispherical Cavities", in *Proc. SRF'15*, Whistler, Canada, Sep. 2015, paper TUPB065, pp. 735–738.