Laser polishing for topography management of accelerator cavity surfaces

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Abstract

Improved energy efficiency and reduced cost are greatly desired for advanced particle accelerators. Progress toward both can be made by atomically-smoothing the interior surface of the niobium superconducting radiofrequency accelerator cavities at the machine's heart. Laser polishing offers a green alternative to the present aggressive chemical processes. We found parameters suitable for polishing niobium in all surface states expected for cavity production. Careful measurement of the resulting surface chemistry revealed a modest thinning of the surface oxide layer, but no contamination.

Introduction

Particle accelerators continue to move forward as tools for frontier science and technology. Perhaps the best known is the Large Hadron Collider (LHC), for making possible discovery at last of the Higgs. The high energy physics users at LHC are already thinking beyond it to the next generation machine, the International Linear Collider (ILC). In another arena, for more than thirty years storage ring light sources have provided photons for inquiry to thousands of users. Seeking to push the frontier, linac-based light sources have begun to operate (Flash, Linac Coherent Light Source- LCLS), delivering a new realm of brightness, wavelength and short pulse length. Their next generation (XFEL, LCLS-II) will do even better.

Accelerator construction and operating costs, their size and complexity necessitate that they are located at dedicated international facilities, desirable though it would be to have them at (e.g.) major research universities. Further, the cost issue looms ever larger, both for construction and operation. Construction must provide enough accelerator (length $\times$ accelerating gradient- E_{acc} , MeV/m) to achieve the required final beam energy. Gradient is the major requirement for high energy pulsed machines, like the ILC. Most other machines operate cw (continuous wave). Energy consumption dominates, expressed as the ratio of energy stored in the accelerator cavity to energy deposited in its walls, the cavity quality factor Q_o . Accelerator performance may be depicted as a plot of Q_o vs E_{acc} .

For more than sixty years, particle beams have been accelerated by microwave energy supplied to room-temperature resonant copper cavities. "Warm copper" continues to be the technology of choice for low duty cycle at modest energy, such as synchrotron storage rings. It is not viable, however, for today's frontier machines, let alone tomorrow's. Already in the 1970's accelerator scientists, notably at Siemens and Wuppertal, began to pursue development of a superior alternative: superconducting radiofrequency (SRF) niobium. The journey of the SRF community has been chronicled [1]; the current state and future path have been recently reviewed [2]. The world SRF conference occurs every two years; the proceedings papers are available a few months later [www.JACoW.org]. A consensus view of the present state of SRF

is that the 35 MeV/m gradient target for ILC has been attained and will be further validated by deployment in other machines (XFEL). Reduced energy consumption (higher Q_0) continues to be pursued.

Surface topography has long been viewed as a major culprit via magnetic field enhancement at sharp surface features [3]. The RF magnetic field is excluded from the surface in the superconducting state. It is oriented parallel to the surface and is most intense at the cavity equator. It must deflect to accommodate sharp features, enhancing the local field. Ultimately the critical field is exceeded locally. Though the resulting normal-conducting volume may be too small to trigger a general quench, the cumulative effect of many such small defects turning on with increasing gradient would be manifest as decreasing Q_0 [4] at higher gradient. Features produced on the niobium surface by chemical etching have been reported [5] with edge radius of curvature as low as 50 nm. Thermal feedback may make even apparently-modest defects more pernicious than once thought. A small amount of power deposited at the defect raises the local temperature slightly, increasing the population of normal-conducting electrons. The local surface resistance grows, which further increases power deposition until either heat transport out of the absorbing region limits further temperature rise or a quench occurs. Efforts to explicitly model the effect of topography on Q_0 are beginning to meet success [6, 7].

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The topography that impacts the performance of SRF cavities arises during the manufacturing process. Details differ somewhat among the organizations, but the main steps at JLab have much in common with those elsewhere. The standard niobium starting material is specially purified and processed sheet, about 3 mm thick with a $\sim 50\ \mu\text{m}$ grain size. Discs are cut, deep-drawn and trimmed to form cups (half-cells). If needed to remove major defects, the cavities are mechanically polished. They are etched to remove a few μm of surface ("light BCP") and e-beam welded to form the cavity. More than $100\ \mu\text{m}$ is etched from the interior surface ("heavy BCP") and the cavity is subjected to high pressure rinsing. BCP (buffered chemical polishing) is immersion in a 1:1:1 or 1:1:2 mixture of concentrated hydrofluoric, sulfuric and phosphoric acids [1]. For many years, cavities in this state were tested and assembled into cryomodules for installation in the accelerator. Such would be the surface state of cavities coming from cryomodules being routinely cycled out for maintenance. Currently new production cavities are electropolished to remove a further $30\ \mu\text{m}$ from the interior surface ("EP") before the final low temperature bake. Alternative schemes are being investigated, including polishing-chemistry free of hydrofluoric acid and more extensive mechanical polishing. For example, at JLab centrifugal barrel polishing ("CBP") is being tested as a substitute for the BCP heavy etch, leaving only the $30\ \mu\text{m}$ removal by EP [8-11]

All these approaches combine mechanical and chemical polishing of various sorts in varying degrees. They may bring improvements, but they are not likely to be game-changers. Moreover, the increasing standards for gradient and energy efficiency are forcing cavity builders toward 100% inspection of the interior surface. As an alternative, laser polishing offers some attractive benefits. Among the leaders: no chemistry, no surface contact, simultaneous 100% monitoring and option to re-do all or part of the surface. All that and more said, a lengthy journey lies ahead to the point where the accelerator community will build a laser processed machine. The small step presented below aims to demonstrate that laser polishing can attain the topography and chemistry needed, given the expected range of starting surface conditions.

Laser polishing relies on beam heating to partially melt the metal surface. Partial melting of the surface (or shallow surface melting, SSM) is preferred instead of formation of a full liquid surface layer (or surface over-melting, SOM) [12]. Usually pulsed lasers are used so that the melt duration is short and cooling happens quickly. During the short period of melting (a few nanoseconds), the surface tension works to smooth out sharp features. Recall that sharpness per se is harmful to cavity performance. The absence of a fully-formed surface melt layer avoids hydrodynamic effects such as the formation of ripples that can remain upon solidification, a harmful sharp roughness. The final smoothening is the consequence of the total melt duration, the sum of the effect of all pulses delivered to a location. Since the effect of any pulse is reduced by the smoothening resulting from previous pulses, it is expected that there will be an optimum total melt time (= number of laser pulses) for cost effectiveness.

Laser polishing could be inserted at a number of points in the process for new cavities, beginning with the completion of mechanical assembly. It could also be applied to existing cavities as the cryomodules in which they are housed are routinely cycled out for refurbishment. Several surface states need to be examined as starting points: mechanically polished only, centrifugal barrel polished (CBP), light BCP and heavy BCP (existing cavities). A state with essentially no initial topography is provided by coupons metallographically polished to $< 50 \text{ nm R}_a$, termed nanopolished ("NP") in our previous work. Such smoothness facilitates application of the characterization techniques discussed later.

Experiment

Material and preparation

Niobium squares $1 \text{ cm} \times 1 \text{ cm}$ were cut from 3 mm-thick sheet material used for cavity fabrication. For the "CBP" surface, a niobium tube was internally polished in the CBP equipment, operating as described in reference [11], and then squares were cut from the tube by electrical discharge machining (EDM).

For the welding preparation surface, the niobium squares were lapped (mechanical polishing with rough sanding material) and treated 1:1:1 BCP at ambient temperature for 1 minute ("light BCP"). A further sample set was subjected to 1:1:2 BCP for time sufficient to remove mass equivalent to $120 \mu\text{m}$ ("heavy BCP"). Finally, nanopolished samples (see later) were prepared for SIMS ("NP"). The quotes above indicate the name by which each item is called later. All samples were cleaned ultrasonically with detergent and de-ionized water after polishing. They were then dried and loaded onto the sample holders in the laser treatment apparatus.

Laser treatment

A Spectra Physics High Intensity Peak Power Oscillator (HIPPO) micromachining laser operating at 1064 nm provided the beam. The repetition rate was 19 kHz, and the pulse length was about 8 ns. The beam was directed into the process chamber of a PLD 5000 system from PVD Products Inc. The angle between the incident beam and the metal surface normal was 60° . The beam incident onto the metal surface was p-polarized, with an absorption ratio of 33 %. The sample holder could accommodate three discs and each disk could rotate at a speed up to 60 rpm.

The square samples were fit into three aluminum discs, each having four 1 cm x 1 cm openings on each one. The focused spot size on the sample was 62 μm x 124 μm (2σ of Gaussian beam) (equivalent to a spot area of $6.04 \times 10^{-5} \text{ cm}^2$). The spot size was measured separately by monitoring the intensity reflected from a mirror at the sample position as a razor blade traversed the beam spot. The vacuum chamber was pumped down to 10^{-7} ~ 10^{-8} Torr before the polishing and never exceeded 10^{-7} . Polishing parameters were chosen according to previous study [13]. The fluence studied was from 0.67 J/cm² to 1.12 J/cm² with 34 pulses accumulated in one spot width.

Characterization

The samples were examined with optical microscopy (HIROX KH-7700 High Resolution Digital-Video Microscopy System) and scanning electron microscopy (SEM - Phenom Desktop SEM). Atomic force microscopy (AFM- Digital Instruments Nanoscope IV) was applied to selected smooth-appearing areas to determine if indeed they were smooth as it is relevant for SRF – free of sharp features. AFM data were collected as previously. The instrument was operated in "tapping" mode with a scan rate of 0.5 Hz. The scanned area was 25 μm x 25 μm , each scan comprising 512 equally-spaced points. Power spectral densities (PSD) were calculated from AFM data, as described in references [6, 7, 14].

Melt processing the surface brings attention to the possibilities of grain re-growth vs nucleation of new grains or perhaps other unanticipated outcomes. Electron back-scatter diffraction (EBSD) was used to visualize surface grain crystallographic orientation. The work was carried out at the Nanoscale Characterization and Fabrication Laboratory at Virginia Tech, using a FEI Helios 600 Nanolab dual-beam FIB equipped with a Pegasus XM-4 EBSD system. The e-beam used was 2.7 nA at 20 kV with a 15 mm working distance.

The shallow RF penetration in the superconducting state (~40 nm) lends great importance to the chemistry of this layer. It is reasonable to inquire as to what effect laser polishing might have. We have previously employed secondary ion mass spectrometry (SIMS) to determine composition as a function of depth below the surface (sputter profile) of niobium materials [15]. The present measurements were performed by the Analytical Instrumentation Facility at North Carolina State University.

TOF-SIMS [16] is a highly sensitive surface analytical technique for acquisition of elemental and molecular information from the surface of a material with high spatial and mass resolution. A finely focused, pulsed primary ion beam is rastered across the surface of the sample and the secondary ions emitted at each irradiated point or pixel are extracted into a time of flight mass spectrometer, mass filtered, and counted. With dual beam operation, meaning sputtering with Cs ion beam and analyzing with liquid metal ion beam Bi, a depth profile of ions can be obtained. TOF-SIMS analyses were conducted using a TOF-SIMS V (ION TOF, Inc. Chestnut Ridge, NY) instrument equipped with a Bi_n^{m+} ($n = 1 - 5$, $m = 1, 2$) liquid metal ion gun and Cs sputtering gun. The analysis chamber pressure is maintained below 5.0×10^{-9} mbar to avoid contamination of the surfaces to be analyzed. For the depth profiles acquired in this study, 1 keV low energy Cs^+ with 10 nA current was used to create a 150 μm by 150 μm crater, and the middle 10 μm by 10 μm area was analyzed using 25 keV Bi_3^+ primary ion beam at 0.35 pA current. The negative secondary ion mass spectra obtained were calibrated using H^- , C^- , O^- , AlO^- , Nb^- .

Results and discussion

Figure 1 shows optical and SEM images of the CBP surface; the laser track is evident. The visual appearance is mirror-like, but the microscopy reveals scratches and pits. Figure 2 shows the AFM images of CBP surface before and after laser polishing. Small waviness was introduced by the laser polishing but the scratches and pits due to the CBP process were eliminated. The pre- and post-treatment roughness values (R_q) were 36 nm and 85 nm, respectively. These values are presented for comparison; they provide little insight into topography as it is relevant to SRF applications. The PSD analysis in Figure 3 presents another view of the topography change. After laser polishing, the contribution from the lower frequency region (below $3 \times 10^{-4} \text{ nm}^{-1}$) increased, while contribution from the higher frequency region (above $3 \times 10^{-4} \text{ nm}^{-1}$) decreased. This accounts for the higher roughness value after treatment. The relatively gentle waviness is understood to be without adverse impact on SRF performance, in contrast with the small, sharp features. We note that application of PSD analysis to CBP topography studies appears not to have occurred within the SRF community previously. Further study is needed to gain deeper understanding.

Figure 1: SEM image of CBP surface with laser polished area. Laser fluence was 0.71 J/cm^2 pulse and pulse accumulation at 34 pulses per spot width.

Figure 2 AFM image of CBP surface before (above) and after laser polishing, scanning area 25 μm x 25 μm .

Figure 3: PSD plot of CBP surface before and after laser polishing, corresponding to the AFM images in Figure 2.

Figure 4: SEM image of light BCP surface with laser polished area. Laser fluence was 0.71 J/cm^2 pulse and pulse accumulation at 34 pulses per spot width. Figure 4 shows the SEM images of the light BCP surface with the laser polishing region at the center. It should be noted that the typical small, sharp-edged features are not grain structure; the grains are much larger ($\sim 50 \mu\text{m}$ vs $10 \mu\text{m}$). The AFM images and PSD plots display the same patterns that we have previously reported.

Figure 5: AFM image of light BCP surface before and after laser polishing, scanning area 25 μm x 25 μm .

Figure 6: PSD plot of light BCP surface before and after laser polishing, corresponding to the AFM images in Figure 5.

Fig. 7 A: SEM image of the previous Light BCP sample acquired at 70° tilt in the EBSD instrument. The area viewed by the EBSD camera (green outline) is foreshortened accordingly. Note the laser track extending from bottom left to upper right.

Fig. 7 B. Orientation image map of area pictured in the previous figure. Area is 400 x 400 microns. Data acquired at 1.75 μm step size.

Fig. 8 Heavy BCP irradiated with 0.89 J/cm^2 pulse at 34 pulses per spot width

Fig. 8 shows the effect subjecting “Heavy BCP” material to laser irradiation of 34 pulses per spot width at 0.89 J/cm^2 per pulse, the fluence found earlier to give best smoothening for other materials. Significant rounding is evident

Figure 9. AFM image of heavy BCP niobium surface before (upper) and after laser polishing, Scanning area 25 μm x 25 μm . Laser polishing fluence was 0.89 J/cm^2 and 34 pulses were accumulated within one spot width.

Figure 10. PSD plot of heavy BCP niobium surface before and after laser polishing, calculated from the AFM data in Figure 9

Figure 11. SEM image of nano-polished niobium surface with laser polished track. Laser polishing fluence was 1.0 J/cm^2 and 34 pulses were accumulated within one spot width. Grain boundaries are barely evident. No features were visible in the optical images.

Figure 12. AFM image of nano-polished niobium surface before (above) and after laser polishing. Scanning area 25 μm x 25 μm . Laser polishing fluence was 1.0 J/cm² and 34 pulses were accumulated within one spot width.

Figure 13. PSD plot of nano-polished niobium surface before and after laser polishing, calculated from the AFM data in Figure 12. Note the increased intensity at long wavelengths, in agreement with the waviness seen in the images.

Figure 14. Optical image of multi-track laser polishing surface. Fluence 1.13 J/cm^2 , 34 pulses accumulated, 0.6 overlapping ratio.

Figure 15. AFM image of multi-track laser polishing surface. Fluence $1.13 \text{ J}/\text{cm}^2$, 34 pulses accumulated. 0.6 overlapping ratio

Figure 16. PSD analysis of AFM image in Figure 15 and other overlap ratios.

SIMS Results

Fig. 17 B: SIMS sputter profiles of major components of laser polished NP niobium

Top: 1.04 J/cm^2

Bottom: 1.12 J/cm^2

Fig. 17 C : SIMS sputter profiles of major components of laser polished NP niobium in untreated material

The SIMS results from the other NP samples were closely similar. Consider the green (NbH) and red (H) traces from the untreated sample in the major components spectra. Both show a depressed value for the first 20-25 seconds of sputtering, corresponding to the surface oxide layer. It was found earlier that niobium oxide and niobium metal sputter at similar rates under the present conditions. Our previous work [17] reported the oxide layer thickness as 4-6 nm, depending on details, implying that the total sputter depth is of order 20-30 nm, well within the RF penetration depth. The only evident impact of laser polishing is that the oxide layer becomes slightly thinner, the more so as fluence increases. No evidence of contamination is apparent.

Future Work

The work on laying down multiple adjacent tracks to cover an area needs to be refined and extended, but it is clearly within reach to eliminate sharp topography by this means. The most important question is then: does it help ? Until recently answering this question required making a high quality coating on the interior of a single cell cavity, a major undertaking. Indeed a similar accomplishment is needed for most surface treatments, such as thin film deposition. Certainly some risk reduction is to be desired. To this end, a surface impedance characterization (SIC) instrument has been developed. Briefly, it provides that a material of interest in the form of a 50 mm flat disc serves as one end of an RF cavity that can be cooled to below 2 K. Energy deposition with the experimental material is compared to a disc of reference material [18].

Success with flat discs will motivate scale-up to cavities. It is needed to accurately deliver a suitable sequence of laser pulses to every location on the interior surface of a cavity. Fortunately, future inspection requirements create much of the same need. It has been determined that it is needed to view every location as an image and to collect white light interferometry data to describe the topography. Further, it is needed to be able to deliver a laser

beam able to repair damage spots that might be found. The machine (“CYCLOPS”) is in place and is undergoing commissioning [19]. It is not a far step to use it for the treatment purpose.

Conclusion

Laser polishing with optimized parameters was performed on niobium surfaces prepared in different states that might be expected or desired in SRF cavity production. They were examined by multiple characterization techniques. Especially in materials prepared by BCP, there was strong evidence of sharp edges, known to be associated with impaired performance. Laser polishing rounded them significantly in all instances. Grain re-growth offers the best description of the chief effect of laser polishing on surface grain structure. Careful examination of the RF-active surface layer uncovered no evidence of contamination.

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References

- [1] H.Padamsee, J.Knobloch, T.Hayes; “RF Superconductivity for accelerators” 2nd edn” 2008 Wiley VCH
- [2] Charles E. Reece and Gianluigi Ciovati, “Superconducting radio-frequency technology R&D for future accelerator applications,” *Reviews of Accelerator Science and Technology*, vol. 5, pp. 285–312, 2012.
- [3] J. Knobloch, R. L. Geng, M. Liepe, and H. Padamsee, “High-field Q slope in superconducting cavities due to magnetic field enhancement at grain boundaries” TUA 004 in *Proc. Intl. Workshop on Radiofrequency Superconductivity*, Santa Fe, NM, 1999, USA. p.77-79.
- [4] A. Dzyuba, A. Romanenko, and L. D. Cooley, “Model for quality factor degradation at high accelerating fields in superconducting radiofrequency cavities” *Supercond. Sci. Technol.* 23 125011 (2010).
- [5] X.Zhao, G. Ciovati, and T.R.Bieler, “Characterization of etch pits found on a large-grain bulk niobium superconducting radio-frequency resonant cavity” *Phys.Rev.ST Accel. Beams.* 13 124701 (2010).
- [6] Chen Xu, Hui Tian, Charles E. Reece, and Michael J. Kelley, “Enhanced characterization of niobium surface topography” *Physical Review Special Topics - Accelerators and Beams*, vol.14 (2011) 123501
- [7] Chen Xu, Hui Tian, Charles E. Reece, and Michael J. Kelley, “Topographic power spectral density study of the effect of surface treatment processes on niobium for superconducting radio

- frequency accelerator cavities,” *Physical Review Special Topics - Accelerators and Beams*, vol. 15, no. 4, p. 043502, 2012.
- [8] C. Cooper, Kenji Saito, B. Bullock, S. Joshi, and A. Palczewski, “Centrifugal barrel polishing of cavities worldwide,” in *Proceedings of SRF2011*, Chicago, IL USA, 2011, pp. WEIOA02, 571–575.
- [9] A. D. Palczewski, H. Tian, O. Trofimova, and C. E. Reece, “Detailed surface analysis of incremental centrifugal barrel polishing (CBP) of single-crystal niobium samples,” in *Proceedings of SRF2011*, Chicago, IL USA, 2011, pp. THPO071, 908–911.
- [10] A. D. Palczewski, H. Tian, and R. L. Geng, “Optimizing centrifugal barrel polishing for mirror finish SRF cavity and RF tests at Jefferson Lab,” in *Proceedings of IPAC2012*, New Orleans, Louisiana, USA, 2012, pp. WEPPC094, 2435–2437.
- [11] A. D. Palczewski, G.V Eremeev, R.L.Geng, I.Jezelow, W.Singer, X.Singer, “Exploration of material removal rate of SRF elliptical cavities as a function of media type and cavity shape on niobium and copper using centrifugal barrel polishing (CBP),” TUP063, Proc. 16th Intl.Conf. RF Superconductivity – SRF 2013
- [12] J. A. Ramos, D. L. Bourell, and J. J. Beaman, “Surface over-melt during laser polishing of indirect-SLS metal parts,” *Materials Research Society Symposium Proceedings*, vol. 758, pp. 53–61, 2002.
- [13] Liang Zhao, J. Michael Klopff, Charles E. Reece, Michael J. Kelley; “Laser polishing of niobium for SRF applications” Proc. 16th Intl.Conf. RF Superconductivity – SRF 2013 TUP068 p.593-595. 2013
- [14] Hui Tian, Guilhem Ribeill, Chen Xu, Charles E. Reece, Michael J. Kelley ; “A Novel approach to characterizing the surface topography of niobium superconducting radio frequency (SRF) accelerator cavities” *Appl. Surf. Sci.* 257 (2011) 4781-4786.
- [15] P.Maheshwari, H.Tian, C.E.Reece, M.J.Kelley, G.R.Myneni, F.A.Stevie, J.M.Rigsbee, A.D.Batchelor, D.P.Griffis; “Surface analysis of Nb materials for SRF cavities ”*Surf.Interface Anal.* (2011) 43 151-153
- [16] A. M. Belu, D. J. Graham, D. G. Castner, “Time-of-Flight secondary ion mass spectrometry: Techniques and applications for the characterization of biomaterial surfaces”, *Biomaterials*, 2003, 24, 3635 – 3653.
- [17] Hui Tian, Charles E. Reece, Michael J. Kelley, Shancai Wang, Lukasz Pulczynski, Kevin E. Smith, Matthew Nowell; “Surface studies of niobium chemically polished under conditions for superconducting radio frequency (SRF) cavity production.”*Appl. Surf. Sci.* 253 (2006) 1236-42
- [18] B.P.Xiao, C.E.Reece, H.L.Phillips, M.J.Kelley; “Calorimeters for precise power dissipation measurements on controlled-temperature superconducting radiofrequency samples” *Rev.Sci.Instr.* 83 (2012) 124905
- [19] C.E.Reece, A.D.Palczewski, H.Tian, J.Bearden, L. Jacobs; “A new internal optical profilometry system for characterization of RF cavity surfaces - CYCLOPS” MOPB062. Proc. LINAC 2012 318-320