A STUDY OF THE EFFECTIVENESS OF PARTICULATE CLEANING PROTOCOLS ON INTENTIONALLY CONTAMINATED NIOBIUM SURFACES*

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Abstract

Particulate contamination on the surface of SRF cavities is known to limit their performance via the enhanced generation of field-emitted electrons. Considerable efforts are expended to actively clean and avoid such contamination on niobium surfaces. The protocols in active use have been developed via feedback from cavity testing. This approach has the risk of over-conservatively ratcheting an ever increasing complexity of methods which are found to result in adequate cavity performance in particular circumstances. A complementary and perhaps helpful approach is to quantitatively assess the effectiveness of candidate methods at removing intentional representative particulate contamination. Toward this end, we developed a standardized contamination protocol using water suspensions of Nb_2O_5 and SS 316 powders applied to BCP'd surfaces of standardized niobium samples which resulted in particle densities of order 100 particles/mm^2. From these common starting conditions, controlled application of high pressure water rinse, ultrasonic cleaning, or CO_2 snow jet cleaning were applied and the resulting surfaces examined via SEM/scanning EDS with particle recognition software. Preliminary results comparing these methods and selected parametric variations of each are reported.

INTRODUCTION

The role that particulate contamination can play in limiting the performance of SRF accelerating cavities has been well established. Laboratories have developed protocols for cleaning sensitive surfaces and controlling the introduction of contaminating particulates. The employed techniques include ultrasonic cleaning, ultra-filtered ultra-pure water, high pressure water rinse, ambient cleanroom environments, filtered gas purging, and CO_2 snow jet cleaning. Such are amply documented in the proceedings of previous SRF Workshops. The record shows that significant progress has been made in reducing the occurrence of particulate-induced field emission loading as the principal performance limitation of niobium SRF cavities. General characterization of particulate size association with potential for producing field emission as a function of surface electric field has recently been summarized by Dangwal Pandey et al.[1] Particulates source control and effective removal techniques remain serious quality assurance challenges. Because the protocols in use have been developed in largely an empirical manner, process parameterization that might provide a basis for process optimization is not yet available.

We would eventually like to establish a basis for selection and design of cleaning strategy, sequence, and duration process steps that confidently remove all identified particulate contamination sources that affect cavity performance. Toward this end, we have initiated an effort to build a quantitative characterization of the effectiveness of particular cleaning processes when applied to deliberately heavily contaminated samples. A variety of types of particulates have been found associated with SRF cavity performance degradation. We considered two candidate representative materials for use in the present investigation: Nb_2O_5 and stainless steel 316 (SS316). Particles of the former appear to be produced by some of the chemical treatment steps applied to cavities [2], while the latter is taken as a representative environmental contaminate generated by mechanical operations within the cavity-handling and assembly cleanrooms.

In order to be able to directly compare the effectiveness of each cleaning process, we established a reproducible high contamination state as the starting condition for each sample. We also established a systematic counting methodology able to discriminate the deliberately applied contaminate and applied it to each sample after each cleaning treatment. For the present work, the samples were all BCP etched high purity polycrystalline niobium disks.

EXPERIMENTAL STUDIES

Sample Preparations

The standardized particulates used in this study were Nb_2O_5 and SS316 powders obtained commercially. The Nb_2O_5 powder grade HPO 400 was provided by H.C. Stark Inc. The high purity of Nb_2O_5 powder was confirmed by XRD measurements. The particle size in this powder was found via SEM survey to range from 0.3 to 31 microns. The SS316 powder was obtained from

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Goodfellow Inc. and was found to be made up of particles ranging from 8 to 50 microns.

The circumstance of cavity contamination that we sought to model is that following active chemistry when the surface is wet and susceptible to fallout/precipitation of chemical by-products and capture of ambient particulates. We thus developed a standard suspension concentration in ultra pure water for each powder type and pipetted 0.5 ml of suspension onto standard niobium disks which were freshly etched by BCP, as illustrated in Figure 1. The samples were allowed to air dry overnight in a Class 1000 cleanroom environment.

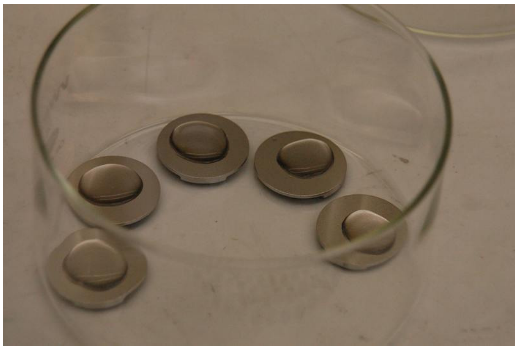

Figure 1. Five samples with 0.5 ml particulate/UPW suspension drying in cleanroom air.

An objective of the study was to gain statistical advantage by deliberately maximizing the particulate density, while avoiding significant aggregation effects. This preliminary effort selected the following suspension concentrations for the powders used based on observed particulate densities using our scanning Amray 1830 SEM equipped with an EDX Genesis Particle Analyzer system: Nb_2O_5 – 8.2 mg/liter, and SS316 – 698 mg/liter. Use of the composition particle detection capability enabled us to discriminate the intentional particulates from others that may incidentally arrive from the environment.

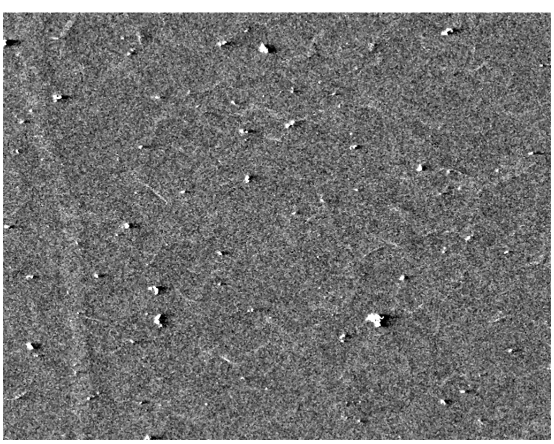

Figure 2. Nb_2O_5 particles scanned at 100x magnification (0.841 mm^2).

A standardized survey protocol was developed to sample five areas on each sample within the drying area. The above protocol was found to result in an observed particle density of ~110 particles/mm^2 for Nb_2O_5 and ~13 particles/mm^2 for SS316. This provided the initial state for all subsequent cleaning tests. The distribution of particle sizes found on the contaminated surfaces is shown in Figure 3. The Nb_2O_5 contaminated samples were scanned at two magnifications, 100x, covering an area of 0.814 mm^{2,} and 750x, covering an area of 0.01 mm^2. The stainless steel contaminated samples were scanned at 50x covering an area of 3.37mm^2.

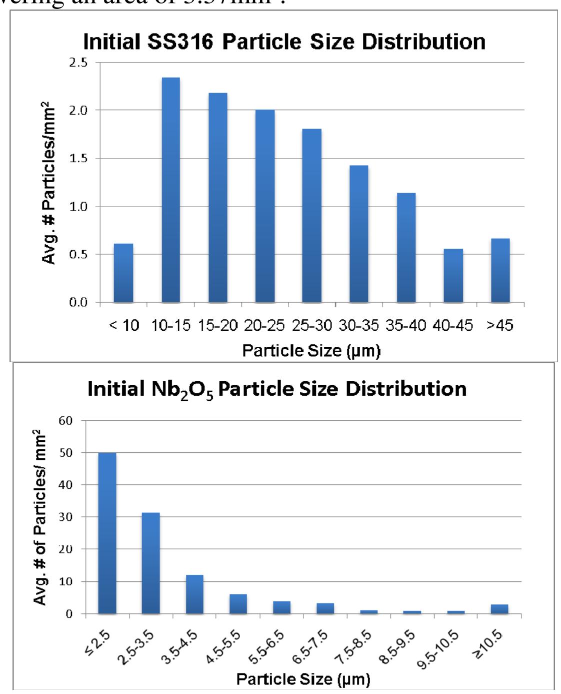

Figure 3. Distribution of particle sizes used in this project.

Ultrasonic Cleaning

The effectiveness of particle removal in a standard ultrasonic bath (Crest Ultrasonics Genesis unit) was investigated with respect to the concentration of Micro detergent and then with respect to time of ultrasonic exposure. The ultrasonic (US) cleaning applied in this series did not include flushing or filtering of the ultrasonic bath, so we recognize the potential for recontamination. Each sample was rinsed with a gentle 15-second flush with UPW at the conclusion of ultrasonic treatment.

The following summary results were obtained:

- US cleaning with only water removed 100% of Nb_2O_5 particles > 5 µm.
- US cleaning with 1% Micro solution was comparable to water only (95%) for removing Nb_2O_5 particles 0.5 - 5 µm.
- US cleaning with > 2% Micro solution was significantly worse at removing Nb_2O_5 than only water for all sizes sampled >0.5 µm.
- Removal efficiency of Nb_2O_5 particles with 2% Micro was independent of the amount of cleaning time in the range 5–60 minutes, and averaged 94–98% for all particle sizes 1–8 µm. Inadequate statistics were available for larger sizes.

- US cleaning with only water removed 100% of SS particles > 15 µm, 96% of all SS particles.
- US cleaning with 2% Micro solution removed 100% of SS particles.

High Pressure Water Rinsing

SRF cavities are now commonly cleaned with a high pressure rinse of ultrapure water (HPR). To seek to discern cleaning effectiveness dependence on detailed geometrical parameters, we exposed the standard contaminated samples to representative HPR configurations and then examined them for residual particles. The Jefferson Lab HPR cabinet was used in all cases with the vertical fan jet nozzles [3] and rotation/advance sequence as is currently applied to all cavities. A vertical stroke of 20 cm travel, centered on the samples, was used for the HPR spray wand. Three samples were treated simultaneously. Each run took approximately 45 minutes. The holding fixture is illustrated in Figure 5.

Sets of samples were cleaned for each of five different orientation angles with respect to the spray: 35°, 45°, 65°, 75°, and 90° (normal to the jet) with the nearest edge of the samples maintained 9 cm from the nozzles.

In addition, sets of samples were cleaned at six different distances from the nozzles: 2.5, 5.1, 9.0, 11.6, 16.0, and 20.3 cm, with the sample holder set at a fixed angle of 65°. Figure 4 shows the observed percentage of Nb_2O_5 particles removed by the HPR treatment. Within the sampling sensitivity used, the similar removal efficiency of the SS316 particles was 100% for all angles and distances tested. Whether this was due to the distinctly different size distribution or some particular material-dependent adhesion feature is unknown.

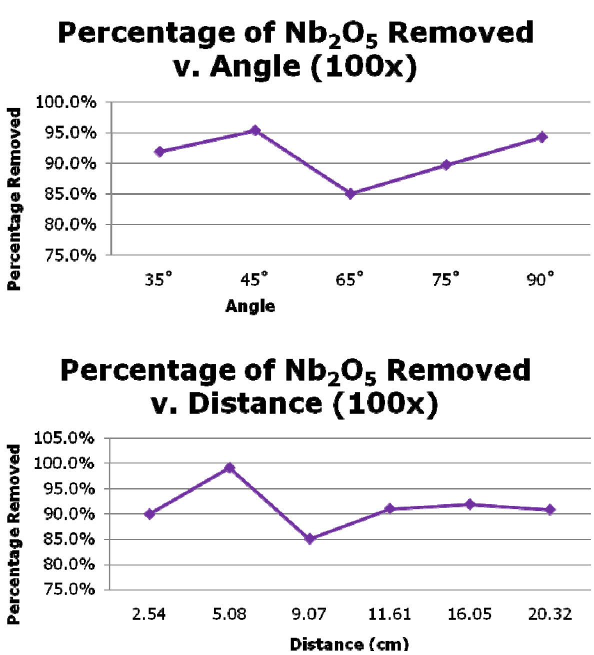

Figure 4. Percentage of Nb_2O_5 particles removed by the applied HPR treatment with angle and distance variation.

The following summary results were obtained:

- All SS316 particles were removed in every HPR run.
- HPR cleaning removed ~90% of Nb_2O_5 particles for all sizes sampled 0.5 – 10 µm.
- HPR cleaning was more effective at 45º and 90º angle than 65º and 75º at 9 cm spray distance.
- HPR cleaning at 65º was ineffective (only 60-90%) at removing Nb_2O_5 particles 0.5 – 10 µm over the sampled distances 2.5 – 20 cm.

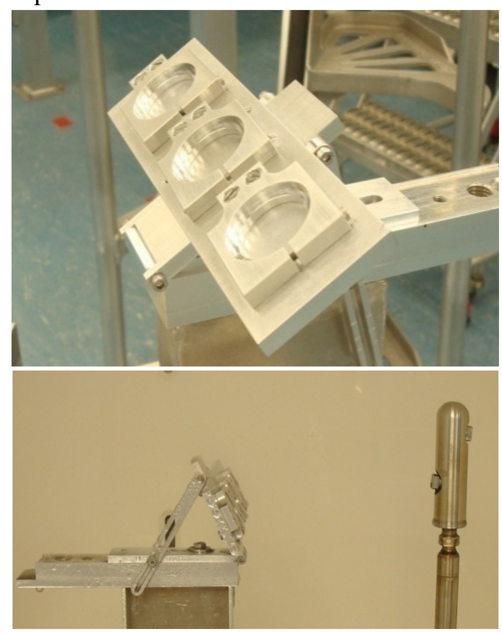

Figure 5. Fixture for holding three samples during HPR.

CO_2 Snow Jet Cleaning

Some development efforts have been applied to using CO_2 snow jet cleaning (also known as dry-ice cleaning) to single-cell niobium SRF cavities and the results have been positive [4]. To begin to establish a relative comparison of cleaning effectiveness with other techniques, we applied an initial CO_2 snow jet cleaning procedure to sets of contaminated samples prepared in the standard way.

The cleaning system used was a hand-held K6-10DG the Dual Gas CO_2 snow system from Applied Surface Technologies. The supply CO_2 pressure was 850 psi. An integral nitrogen gas shroud, supplied at 85 psi, served to reduce or eliminate water condensation on the cleaning surface. The unit included sintered stainless steel filters rated for 0.003 microns on both gas lines. The cleaning processes were applied in an exhausted clean workstation in a class 1000 cleanroom. For the cleaning of each sample, an attempt was made to maintain constant nozzle-sample work distance, angle of incidence, and total sweep time on the sample. A near-to-far rastered sweep pattern was used consistently.

Samples with Nb_2O_5 and SS316 particles were treated. Measurements were made for five different angles of incidence: 35°, 45°, 65°, 75°, and 90°. Samples were also cleaned with constant angle and varied working distance from 1.0 – 3.5 cm, in increments of 0.5 cm.

Cleaning the standard samples with CO_2 snow proved to be more effective than either US or HPR, so much so that the SEM scans produced no statistically significant results within the limited scanned surface area. Surveys with sensitivity as those summarized in Figure 4 all indicated 100% removal. A supplemental assessment method was then used to evaluate the CO_2 snow cleaned samples. Cleaned samples were subjected to N_2 gun blow-off directly into an airborne particle laser particle counter. While this procedure was less controlled than the SEM scanning, it allowed a sampling mechanism that covered the whole sample surface. Even then, the counts were low considering the highly contaminated initial state. No particles larger than 3 µm were detected by this procedure. The particle counts detected in this way are depicted in Figure 6.

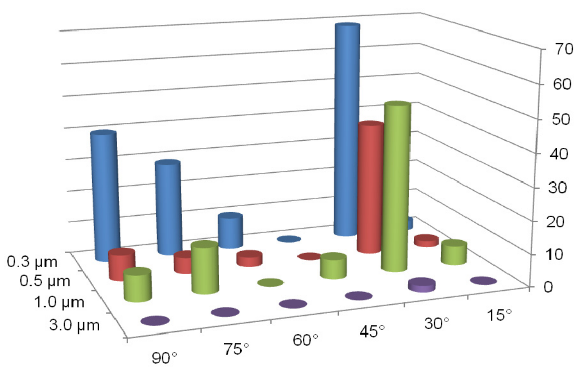

Figure 6. Integrated blow-off particle counts from samples cleaned by CO_2 jet with various incident angles.

From the preliminary CO_2 snow cleaning tests conducted here the following summary results were obtained:

- CO_2 snow jet cleaning removed 100% of deposited SS316 particles under all angles and distances tested.
- CO_2 snow jet cleaning of Nb_2O_5 was significantly more effective with an incident angle of 45º than 30º, 60º, or 90º for particles < 3 µm.
- CO_2 snow jet cleaning of Nb_2O_5 particles was equally effective with working distance 1.0 – 2.5 cm, but degraded quickly with further increase in distance. (The visible extent of the CO_2 crystal jet emerging from the nozzle used is ~3 cm.)
- Significant redeposition of Nb_2O_5 particles from a contaminated sample to a clean witness sample 3 cm distant via CO_2 snow jet cleaning of the former was easily demonstrated.

DISCUSSION

Comparisons

- Particles of Nb_2O_5 and SS316 > 5 µm were thoroughly cleaned by all techniques except HPR, which left some Nb_2O_5 particles on the surface.
- Particles of SS316 < 5 µm were not tested.
- No clear dependence on nozzle-sample distance was found for HPR, although tests at the most effective angles, 45º and 90º, have not yet been performed.
- CO_2 snow jet cleaning of these Nb_2O_5 particles appeared to be significantly more effective than the other two techniques tested.

Lessons for Future Work

- Consider revisiting the starting suspension concentrations with a view to increasing by factor of >10 to improve the counting statistics.
- Continue the HPR study of effectiveness with distance using 45° and 90° angles.
- Repeat the Nb_2O_5 study with optimal EP-treated Nb samples for comparison with BCP surfaces
- As redeposition is a distinct possibility with both US and CO_2 snow jet cleaning, investigate the effectiveness of a sequence of fresh cleaning solutions, to approximate use of a flowing, filtered solution for cavity applications, and a purge gas flow in the CO_2 snow jet cleaning application to carry away removed particles.

The work reported here should be considered a preliminary investigation into appropriate and useful methods for evaluating the effectiveness of various cleaning techniques either used or considered for use on niobium cavities. It would be quite interesting to conduct a similar study with electropolished niobium samples in order to establish the extent to which particle adhesion is dependent on microscopic surface topography.

The niobium samples used in this study were of the design used for direct field emitter scanning in the JLab Scanning Field Emission Microscope and Field Emission Viewer. Future work will have opportunity to extend the correlation of remaining deliberate particulates to DC field emission behavior.

ACKNOWLEDGEMENTS

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