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RECENT RESULTS FROM Nb_3Sn SINGLE CELL CAVITIES COATED AT JEFFERSON LAB*

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Abstract

Because of superior superconducting properties (Tc ~ 18.3K, $H_{s\,h}$ ~ 425 mT and Δ ~ 3.1 meV) compared to niobium, Nb_3Sn promise better RF performance (Q_0 and E_{acc}) and/or higher operating temperature (2 K Vs 4.2 K) for SRF cavities. Nb_3Sn-coated SRF cavities are produced routinely by depositing a few micron-thick Nb_3Sn films on the interior surface of Nb cavities via tin vapor diffusion technique. Early results from Nb_3Sn cavities coated with this technique exhibited precipitous low field Q-slope, also known as *Wuppertal slope*. Several Nb_3Sn single cell cavities coated at JLab appeared to exhibit similar Q-slope. RF testing of cavities and materials study of witness samples were continuously used to modify the coating protocol. At best condition, we were able to produce Nb_3Sn cavity with Q_0 in excess of ≥ 5×10^{10} at 2 K and ≥ 2×10^{10} at 4 K up the accelerating gradient of ~15 MV/m, without any significant Q-slope. In this presentation, we will discuss recent results from several Nb_3Sn coated single-cell cavities linked with material studies of witness samples, coating process modifications and the possible causative factors to Wuppertal slope.

INTRODUCTION

Superconducting radio-frequency cavities (SRF) are the essential technology for modern particle accelerators. Niobium (T_c ~ 9.2 K, H_{sh} ~ 210 mT and Δ ~1.45 meV) is almost the only material of choice so far to build them. Several decades of research and development is bringing the performance of niobium cavities close to intrinsic material limit [1]. They often require to operate at ~ 2 K for optimal performance, which demands complicated cryogenic facilities, and it is one of the major cost drivers for SRF based accelerators. SRF cavities made of superior superconducting materials (higher T_c, H_{sh} and Δ) promise simplified small cryogenic facility and potentially enhance the performance. The intermetallic compound Nb_3Sn (T_c ~ 18.3 K, H_{sh} ~ 425 mT and Δ ~3.1 meV) is a potential alternate material [2] that could allow SRF cavities operation at 4.2 K for similar performance of Nb at 2 K. Several labs working on Nb_3Sn are aiming at high performance Nb_3Sn cavities applicable for economic and powerful accelerators [3-5].

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Due to restrictive material properties of Nb_3Sn, its application for SRF application is limited into a thin film/coating form, which should be deposited inside of built-in metallic cavity structures. Vapor diffusion process, a leading technique so far to fabricate promising Nb_3Sn coated Nb cavities has been adopted at JLab since 2012. The Nb_3Sn deposition system here allows to coat coupon samples, single-cell cavities, two-cell cavities, and recently multi-cell cavities. The coated cavities initially had quality factors (Q_0) as high as > 1×10^{10} at 4 K, but suffered strong Q-slope limiting the attainable maximum gradient. The Q-slope, very similar to the one seen in early cavities coated at Wuppertal University was consistently seen in several cavities [6]. Following the upgrade of the coating system in 2017, we were able to produce almost Q-slope free Nb_3Sn cavity for the first time, but Q was below 1×10^{10} at 2 K [7]. Since then we have prepared and tested several cavity coatings to understand and improve quality and performance limitations. Witness samples coated with cavities were studied as well for continual improvements in the coating process. In this paper, we will present recent results from several Nb_3Sn cavity coatings. We will mainly focus on two cavities, RDT10 and RDT7, which were coated several times with witness samples.

Nb_3Sn COATING

Cavities under discussion here were single-cell cavities made from high purity (RRR~300) fine grain Nb. Each cavity had low field Q_0 ~ 1.6×10^{10} and normally was limited to ~ 30 MV/m by a high field Q slope during the baseline test at 2 K. Each cavity normally received BCP or EP for 15-25 μm removal followed by HPR before the coating. Cavities were coated in *single-cavity* and *two-cavity* setup, discussed below.

In the *single-cavity* setup, each cavity was coated individually according to a typical Nb_3Sn coating process at JLab [6]. 3 g of Sn (99.999% purity from Sigma Aldrich) loaded in a crucible and 3 g of $SnCl_2$ (99.99% purity from Sigma Aldrich) packaged inside two pieces of Nb foils were placed inside the cavity at the bottom flange. Both sides of the cavity were closed with Nb covers before installation into the furnace. A witness sample was also hung inside the cavity by attaching it to the top cover using a Nb wire. The temperature profile included nucleation step at ~500°C for an hour and coating step of three hours at ~1200°C. The temperature was monitored with sheathed type C thermocouples attached to the cavity at different locations. There was a temperature gradient of ~20 °C between the top and bottom of the cavity.

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In the *two-cavity* setup, cavities to be coated (RDT7 or RDT10) were coupled to another single-cell cavity, RDT2 as shown in Fig. 1. RDT2 had many macroscopic pits inside the cavity, and was used as a dummy cavity on top of another cavity to be coated. About ~ 3.4 g of tin and 3 g of $SnCl_2$ were placed at the bottom similar to the first set of experiments. Since larger coating volume increased the chance of non-uniform coating in the past [7], the coating setup also comprised of a secondary tin crucible, which was loaded with 1.4 g of Sn. It was attached to the top cover with a Nb rod, and hung inside the bottom beam pipe of RDT2. A witness sample was always suspended to secondary Sn crucible with Nb wire. The heat profile was similar to the first set of experiments except there was a temperature gradient of ~ 85 °C between the top and bottom of the whole setup, as shown in Fig. 2. The bottom tin container was mostly covered with a diffuser, consisting of a molybdenum disk with holes.

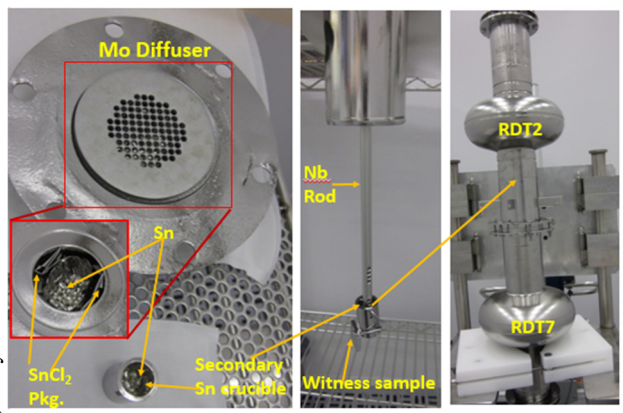

Figure 1: Two-cavity setup for RDT7 coating.

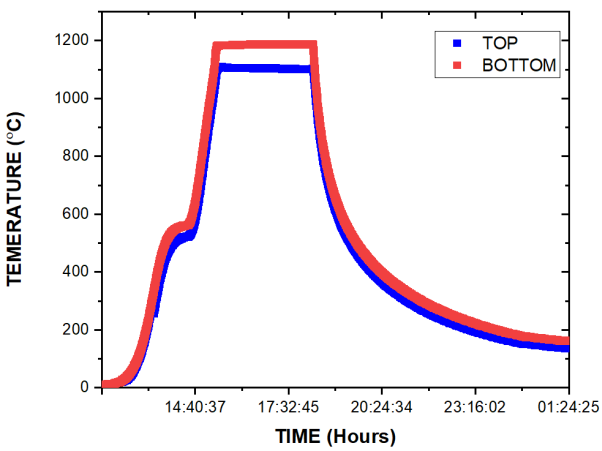

Figure 2: Temperature profile used to coat cavities in two-cavity setup.

RESULTS

RDT7 and RDT10 were initially coated in single-cavity setup, described above. Coating parameters used for the coating were identical. Figure 3 [right] show post-coating pictures from RDT7 and RDT10. Post-coating inspection of both cavities indicated uniform coatings inside both of these cavities.

Witness samples obtained from each cavity coating were examined with secondary electron microscope (SEM) and energy dispersive X-ray spectroscopy [EDS]. SEM images captured from witness samples, Figure 3 [left] showed uniform coating in both samples. EDS analysis of both samples revealed (24.5±0.5) at. % Sn, close to the nominal composition of Nb_3Sn. However, high resolution SEM images from each sample revealed residues on the surface. These residues were few tens of nanometer in diameter as shown in Fig. 3 [left]. Precise probing of those feature was not always possible with EDS resolution, but some of the larger residues showed ~ 30 at. % Sn compared to neighbouring area. It indicated them to be Sn-rich particles.

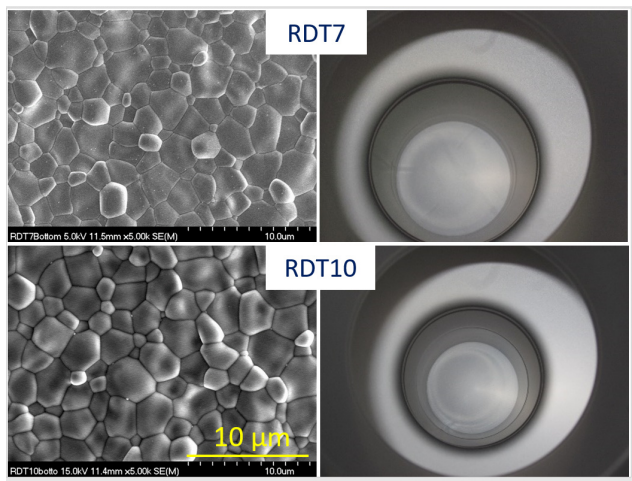

Figure 3: Top left and bottom left images are SEM images captured from witness samples coated with RDT7 and RDT10 respectively. Images to the right show the interior appearance of the cavities after coating.

RF test both at 4 K and 2 K showed precipitous Q-slope in each cavity as shown in Fig. 4. Low field Q_0 at 4 K was $\geq 1\times10^{10}$, but dropped sharply before quenching at ~ 11 MV/m. The coating experiments were repeated a couple more times on RDT10 with some variation in coating temperature, which consistently produced similar performance. Since Sn-residues appeared in the witness sample from each coating, they were considered as a potential causative of observed Q-slope.

Several potential solutions were proposed to obtain a residue free cavity. The first approach was to remove Sn residues from each cavity. We attempted different acid-treatments in an attempt to remove them. These features were mostly removed after one-hour soak in 5 % HCl at room temperature or 10 minute soak in 22% HNO_3 at 110 °C with a magnetic stirrer, see SEM images in Fig. 5. RDT10 was given 30 minutes of HNO_3 soak at room temperature to remove possible indium contamination after disassembly from the first test. It was then soaked in 5 % HCl for an hour, similar to witness sample, HPRed and tested again at 4 K and 2 K. Comparison of RDT10 performance before

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and after acid soak is plotted in Fig. 3, and does not show any improvement. It is also not clear yet how such acid soak affects Nb_3Sn surface.

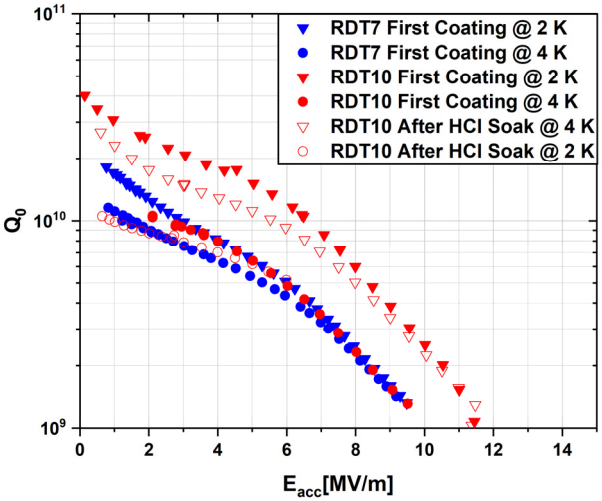

Figure 4: RF test results from RDT7 and RDT10 after first Nb_3Sn coating. Note that RF results following 5 % HCl soak are also shown.

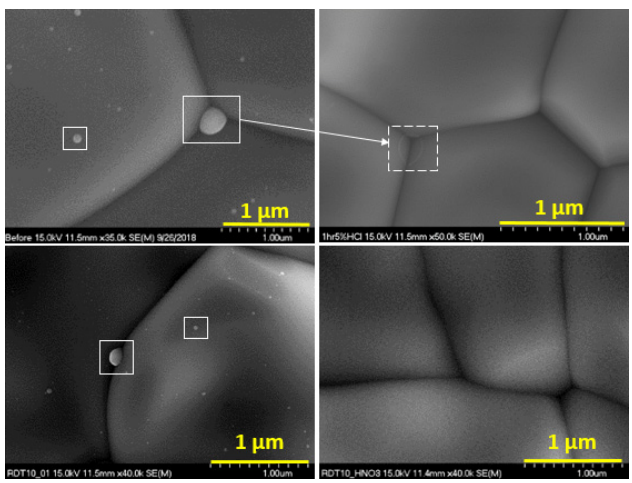

Figure 5: SEM images from RDT7 witness sample before [left] and after [right] 5 % HCl soak for one hour is shown on top. Images at the bottom compare RDT10 witness sample before [left] and after [right] 22% HNO3 soak at 110 °C. Bright features are Sn-rich residues.

Next, we annealed one of the coated cavities that has Q-slope for 2 hours at 1100 °C in the attempt to evaporate Sn residues away from the surface. Both sides of the cavity were closed with Nb covers which were not coated with Nb_3Sn. Witness samples, one previously coated and another a new Nb sample were also placed inside the cavity before annealing. Post-annealing inspection showed some discoloration in Nb covers exposed inside the cavity, which is linked to tin deposition on these covers. RF test showed significant degradation of Q_0 following the annealing. Almost constant Q_0 was measured up to ~ 5 MV/m unlike as-coated cavity, but followed by sharp Q-slope. SEM/EDS analysis showed no tin residues on previously coated witness sample indicating disappearance of Sn residues, and the Sn content appeared ~ 1 at. % less following annealing, which is within the instrumental error limit. However, another niobium sample, not coated before appeared coated in SEM image, see Fig. 6. EDS analysis showed ~ 10 at. % Sn, which indicates Sn transfer during the annealing. We have observed Sn loss from vapor-diffused Nb_3Sn following longer annealing of 12 hours at 1200 °C before, and also after annealing of sequentially sputtered Nb_3Sn sample at ≥1000 °C [8].

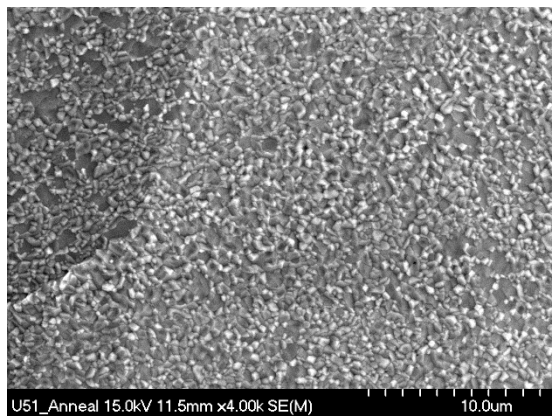

Figure 6: SEM image from Nb sample after annealing at 1100 °C for 2 hours.

Another approach was to avoid residue formation on cavity surface during the coating. We speculated that the Sn-rich residues were the result of Sn condensation from residual Sn vapor, which is present inside the cavity at the end of the coating. In an attempt to reduce Sn condensation in the cavity of interest, RDT2, was added on top as a dummy cavity, and maintained at a temperature lower than RDT7/RDT10 at the bottom to act as tin vapor getter. Coating temperature was set in such a way that there existed a ~ 85 °C gradient between the top and bottom of the paired structure as described as *two-cavity* setup in previous section.

RDT7 and RDT2 were paired first, and coated as described in the previous section. Coating appeared uniform during visual inspection. SEM examination of witness sample exhibited Sn-residues again. Note that the witness sample here was inside RDT2. Since it was suspended to secondary Sn-crucible, which was attached with Nb rod to the top cover, it could have lower temperature than the cavity, and more likely to have Sn-residues. Since there was no witness sample inside the cavity at the bottom, it is not clear if tin residue was present on RDT7 coating or not. RF test results obtained from RDT7 are shown in Fig. 7. The measured value of low field Q_0 was 3×10^{10} at 4 K and 1×10^{11} at 2 K without any significant Q-slope. The cavity maintained a Q_0 of ~ 2×10^{10} at 4 K and > 3×10^{10} at 2 K before quench at >15 MV/m. The cavity performance of RDT7 showed noteworthy improvement in the recurrent Q-slope, compared to data previously reported from Wuppertal and Jefferson Lab.

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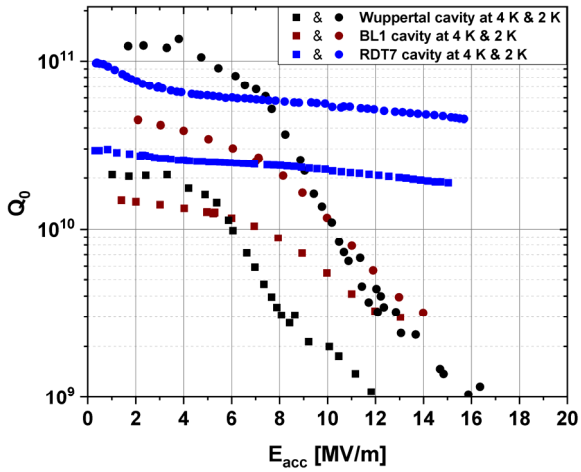

Figure 7: Comparison of RF results from RDT7 with previous data with Q-slopes.

Next, RDT10 was coated together with RDT2 with similar setup and parameters used for RDT7. Another witness sample was installed next to RDT10, which more likely represents the cavity coating. Some non-uniformity was visible first time in the bottom half-cell of the cavity, see Fig. 8 [right]. SEM images from witness sample, next to RDT10, revealed some patchy regions, known to have thinner coating as shown in Fig. 8 [left] and to be harmful to RF performance. The RF test result of this cavity (not shown here) was very similar to the previous test result of RDT10, shown in Fig. 4. Despite having a similar temperature profile compared to previous coating of RDT7, it is found that the consumed amount of tin was almost half (1.7 g vs 3.3 g) during RDT10 coating compared to the tin consumed during the previous RDT7 coating. It is known that low flux of tin produces patchy Nb_3Sn coating with non-uniformity [4, 8]. Reasons behind the lower tin evaporation are not understood completely. We speculate that the reduction in the effective surface area of tin molten pool reduced the evaporation rate. Note that the Mo diffuser used in RDT7 coating broke and was replaced with a new one, which had smaller holes. It was also suspected that the new diffuser has shifted from its original position during the installation into the furnace. Another attempt was made to coat RDT10 again without diffuser to allow maximum tin evaporation. The cavity was coated uniformly this time, but it was found that Sn was splattered and carried over to the cavity. SEM/EDS analysis of witness sample confirmed the splattering of tin. The diffuser made from Nb foil, which almost replicated molybdenum diffuser used in RDT7 coating was used in the third attempt to coat RDT10. Supplied tin was reduced based on RDT7 coating experiment, to limit tin vapor at the end.

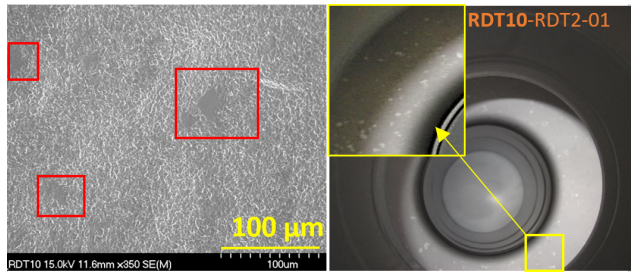

Figure 8: RDT10-RDT2 coating. SEM image [left] shows patchy regions observed in witness sample. Non uniformity in the coating can be seen in the right picture. Note that the top half cell had less non uniformity than the bottom.

Post-coating inspection showed uniform coating inside the cavity. The Sn consumption was very similar to RDT7 coating. Examination of witness samples, one from the bottom and another next to the bottom beam pipe of RDT2 showed uniform coating without any tin residue or patches, shown in Fig. 9. EDS examination showed usual Nb_3Sn composition. RF test results from RDT10 now appeared similar to RDT7 except for the quench field, which was lower in RDT10. The latest test results from RDT10 and RDT7 are compared to the ones after their first coating in Fig. 10.

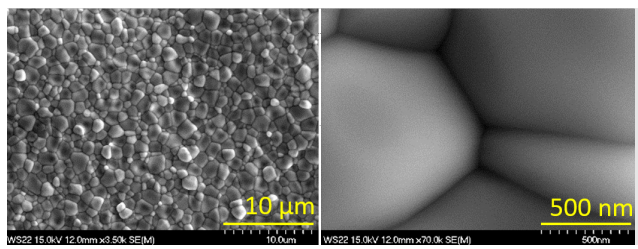

Figure 9: SEM images from RDT10-RDT2 coating. Note that there is Sn residue in the surface [right].

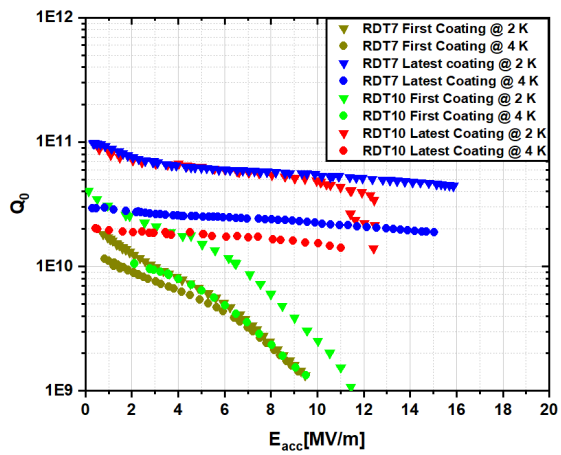

Figure 10: Comparison of latest RF test results from RDT10 and RDT7 with those after their first coating. RDT10 is expected to have higher Q_0 at 4 K than presented here as we expect losses on the flanges because of shorter beam pipes.

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DISCUSSION

Despite promising quality factor and accelerating gradient, Nb_3Sn cavities are vulnerable to precipitous Q-slope. Reported first by Wuppertal University in 1980's [9], the origin of such slope, also known as "*Wuppertal slope",* has not yet been established. JLab coating followed Siemens configuration with single heater and active pumping during the coating, which is different from Wuppertal or Cornell setup, also resulted in similar Q-slopes as shown in Figure 11. Early studies of control samples as well as cutouts from coated cavity with Q-slope indicated mostly uniform coatings with composition close to nominal Nb_3Sn. There were no Sn residues on the surface. However, SIMS analysis of coated sample showed significantly higher presence of Ti in Nb_3Sn layer [10-11], when compared to Nb_3Sn coated sample obtained from Cornell University, where Q-slope free cavities were measured [12]. In case of JLAB samples, Ti contamination likely originated from TIG welds, which were present in the sample chamber. It hinted at the possibility of Ti migration from NbTi flanges of cavities during cavity coatings. EDS examination of alumina hardware, used during coatings also showed significant amount of Ti confirming Ti loss from the NbTi flanges. Ti was measured to affect the field dependence of Q_0 in Nb cavities [13-14]. At Wuppertal the potential source of Ti could be Ti foils, which were used as getter material on the outside of coated cavities to maintain the purity of niobium during the Nb_3Sn coating. During coating system upgrade in 2017, Ti-free hygiene was adopted for Nb_3Sn coating, which we believe produced first Q-slope free cavity. Several cavities coated later resulted in Q-slope, but as discussed above, they were linked with presence of Sn-residue, non-uniformity and also with macroscopic defects in the starting Nb substrate. Figure 11 shows a comparison of Q-slope from several cavities with that with Wuppertal result. The best results from RDT7 is also shown for comparison.

Despite steepness, Q-slope onset, quality factor and attainable gradient appears to vary in different coatings. It indicates several factors could contribute to Q-slope, and requires much more statistics to determine how each factor could contribute. Absence of Q-slope in RD7 and RDT10 indicates that the Q-slope-free cavities can be coated in Siemens coating configuration. As it seems defect-free substrate, defect-free coating and contamination free process are essential for Q-slope free cavity.

SUMMARY AND OUTLOOK

Several single cell cavities coated during the first years of R&D at Jefferson Lab exhibited a strong Q-slope. We were able to produce both Q-slope free cavities and cavities with Q-slope following the cavity system upgrade. Analysis of witness samples coated with single-cell cavities revealed a distribution of Sn-rich residues on the surface. These features mostly disappeared following HCl/HNO_3 acid soak or annealing in SEM images, but similar treatment did not improve the cavity performance. In an attempt to reduce Sn residues formation during the coating process, changes were made in the coating process, which resulted in nearly Q-slope free cavities. The best coated cavity had a Q_0 of $\sim \geq 2\times10^{10}$ at 4 K and $> 3\times10^{10}$ at 2 K before quench at ≥15 MV/m. Q-slope free results were reproduced in both experimental cavities, which had Q-slope before. Nb_3Sn cavities are vulnerable to precipitous Q-slope, which appeared to be caused by several factors in the coating process. Evaporation, consumption and distribution of Sn during the coating is shown to affect the cavity performance remarkably. More experiments with recently added several new cavities are in place to refine the coating process.

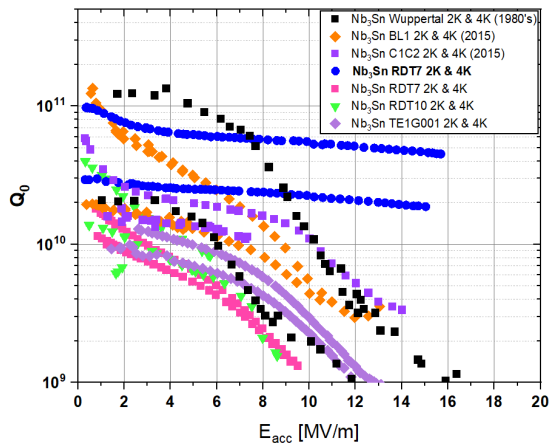

Figure 11: Results from several cavity coatings, which exhibited Q-slope. Note that Wuppertal data [black square] and the best cavity result from JLab [blue circle] are also shown for comparison.

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