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PROGRESS WITH Nb HiPIMS FILMS ON 1.3 GHz Cu CAVITIES*

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Abstract

In recent years, efforts have been invested to leverage the different processes involved in energetic condensation to tailor Nb film growth in sequential steps. The resulting Nb/Cu films display high quality material properties and show promise of high RF performance. The lessons learned are now applied to 1.3 GHz Nb on Cu cavity deposition via high power impulse magnetron sputtering (HiPIMS). RF performance is measured at different temperatures. Particular attention is given to the effect of cooldown and sensitivity to external applied magnetic fields. The results are evaluated in light of the Nb film material and superconducting properties measured with various microscopy and magnetometry techniques in order to better understand the contributing factors to the residual and flux induced surface resistances.

This contribution presents the insights gained in exploiting energetic condensation as a path towards RF Q-slope mitigation for Nb/Cu films, correlating film material characteristics with RF performance.

INTRODUCTION

Superconducting radio frequency (SRF) cavities based on bulk Nb have been used for decades in particle accelerators [1]. However, bulk Nb suffers from many drawbacks among which are included: fundamental accelerating gradient limitation, poor thermal conductivity, manufacturing difficulties and high material costs. One proposed possibility for alleviating some of these challenges is to take advantage of the surface nature of the SRF phenomenon.

In order to truly engineer the active SRF surface, one can exploit the shallow RF penetration depth by using alternative materials for the bulk resonant cavity structure with more favourable properties, such as Cu, and coating a ~1 µm Nb thin film on the interior as the active SRF surface. One can then optimize the RF response by tuning the various coating parameters available during film growth; theoretically allowing performances outside of the bulk material property bounds.

Thin film SRF cavities were first implemented in the 1980's at CERN for LEP II [2], with Nb/Cu thin film cavities deposited using DC magnetron sputtering (DCMS). While these films had good low field Q-values, they exhibited a strong decreasing trend of the Q-value on RF field, known as the "Q-slope", limiting their applications. To this day, the cause of the Q-slope has not been understood and has yet to be overcome.

Many causes proposed for the Q-slope [3-5] can be associated with the low energy film deposition methods utilized. DCMS has been shown to yield superconducting films with properties inferior to bulk. However, energetic condensation methods, such as high-power impulse magnetron sputtering (HiPIMS), promise to create films with enhanced SRF properties [6]. Among these methods, HiPIMS is of specific interest due to its ease of adaptability to coating tri-dimensional shapes and capability of implementation within existing DCMS cavity coating system designs.

In HiPIMS, the magnetron is pulsed to extremely high power densities thereby increasing the plasma density by several orders of magnitude over DCMS and resulting in ionization of a significantly higher fraction of the metal atoms [7, 8], thus, allowing control of the deposition energy by application of a substrate bias. HiPIMS has been shown to yield films with much improved microstructure, density, surface roughness, adhesion and overall quality compared to the low energy DC methods [9-12]. This opens the possibility to produce higher quality films for SRF application.

In order to explore HiPIMS for SRF application, a cavity deposition system was designed, built and commissioned at Thomas Jefferson National Accelerator Facility (JLab) [13]. This system offers many benefits such as the ability to mount and unmount cavities while leaving the larger system under vacuum which allows a higher turn-around cycle and reduced risk of contamination; as well as allowing deposition of small coupon samples under the same pump down cycle as the cavity through a load-lock and enables conditioning of the magnetron isolated from the cavity and samples before deposition. A custom built HiPIMS pulser powering a cylindrical Nb cathode in a Kr atmosphere is used to deposit Nb films. Here we present HiPIMS coated cavity RF results and data from small samples carried out in the same system.

EXPERIMENT

Within the last year, the cavity deposition system described above underwent its first full-scale maintenance cycle initiated by the Nb cathode reaching the end of its lifetime. These repairs included many pre-planned upgrades but focused mainly on cathode replacement. As previously reported, and recapped below in this paper, a study of Nb films on Cu substrates was performed within the last system iteration in order to investigate the effect of peak pulse power and applied substrate voltage bias on the resulting film properties. Since the system maintenance, calibration samples were created in order to verify continuity of film quality. Also, a new sample temperature series of Nb films on Cu substrates was done holding peak power and bias parameters constant at the "optimized" set determined from the previous peak power and bias series,

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while exploring the effect of varying deposition temperature.

The Cu substrates used were commercially bought, polished polycrystalline OFE Cu with a rated surface roughness of less than or equal to 5 nm. Substrates were ultrasonically cleaned in successive acetone and methanol baths then mounted in a clean environment prior to growth to limit particulate contamination. Depositions were performed with a target-to-substrate distance of 10cm, the approximate distance to the equator of a 1.3 GHz low surface field (LSF) cavity.

After growth, the structural, surface and superconducting properties of the samples (lattice parameter, grain size, crystal orientation, roughness, RRR, T_c…etc.) were determined via X-Ray diffraction (XRD) Electron Backscatter Diffraction (EBSD), atomic force microscopy (AFM) and RRR.

For cavity depositions, 1.3 GHz LSF Cu cavities were manufactured at JLab and centrifugal barrel polished (CBP). Prior to deposition the cavities received an acid etch, typically 15 um, via SUBU [14], rinsed and dried via methanol sheeting. For all the assembly stages, deposition and RF testing, the cavities were high pressure rinsed, methanol sheet dried and assembled in an ISO-4 cleanroom following stringent quality procedures. Cavities were also mounted on the deposition system under a portable clean hood that creates a local ISO-5 quality environment in order to keep particulate contamination within the deposition system and on the deposition surfaces minimal. The coated cavities were RF tested in JLab's Vertical Test Area (VTA). After RF testing, the Nb film was stripped from the interior cavity surface via Nb BCP in preparation for the next coating cycle.

RESULTS

Small Samples

Previously, before system maintenance, a peak power and bias series were performed [15] to attempt to determine the effects of these parameters on film properties, and the results are summarized in Figs. 1 through 4. The power series samples were deposited at a constant Kr pressure of 3.9 mTorr, substrate temperature of 350 °C, bias of -100 V and constant pulse parameters, 83 Hz frequency and 110 µs pulse width, while the peak power (voltage) varied between samples. Power values were varied between 69 and 391 kW. The bias series samples were held at the same deposition parameters from the power series, except for the pressure at 4.2 mTorr, due to a pressure gauge change, and the "optimized" peak power setting of around 220 kW, as determined from the power series. In order to explore the effect of applied bias voltage, the bias was varied between 0 and -300 V for this series.

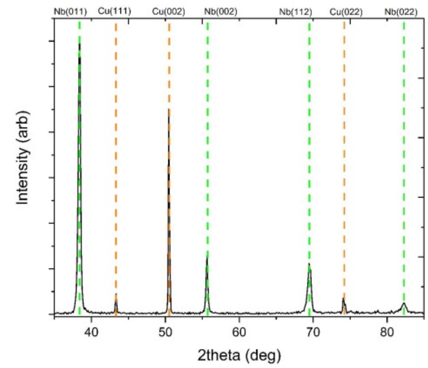

Figure 1: Representative XRD scan of Nb/Cu samples in power series

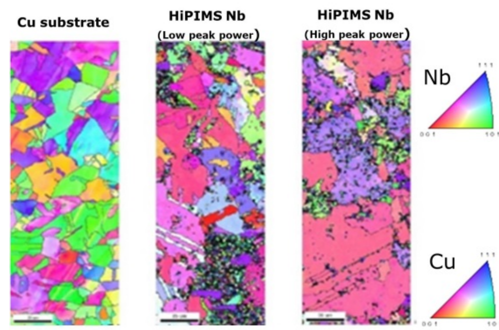

Figure 2: EBSD scans performed on Cu and Nb/Cu samples. From left to right: (a) an uncoated Cu substrate, (b) low peak-power Nb/Cu sample, and (c) high peak-power Nb/Cu sample. OIM colour maps included on right for reference of crystal phases.

To summarize the results from these two sample series, microstructurally, both series exhibited clear polycrystalline texturing dominated by the Nb (011) orientation, as evident from both the XRD and EBSD scans shown in Figs. 1 and 2. Figure 2 also shows the hetero-epitaxial nature of the film growth when images of both the Cu substrate and Nb/Cu films are analysed together.

With regard to surface morphology, AFM results, presented in Fig. 4, show a clear increasing trend of surface roughness with increasing power and bias independently. The samples exhibited very low surface roughness at the lower end of each parameter range, however both increased quickly with power and bias, with bias having a much stronger increasing trend than peak power. Applied bias is an extremely important parameter for energetic condensation methods due to the high sputtered metal ion fraction present in the plasma. In fact, many different deposition schemes proposed for overcoming Q-slope rely on this characteristic of energetic condensation, such as deposition at ion implantation energies to overcome adhesion issues. This result may hint at a possible roughness limitation to applied bias near the RF layer and require mitigation efforts to achieve high RF performance. It is noteworthy as well that no sharp features, detrimental to the RF performance, were observed on the surface of the samples by either SEM or AFM.

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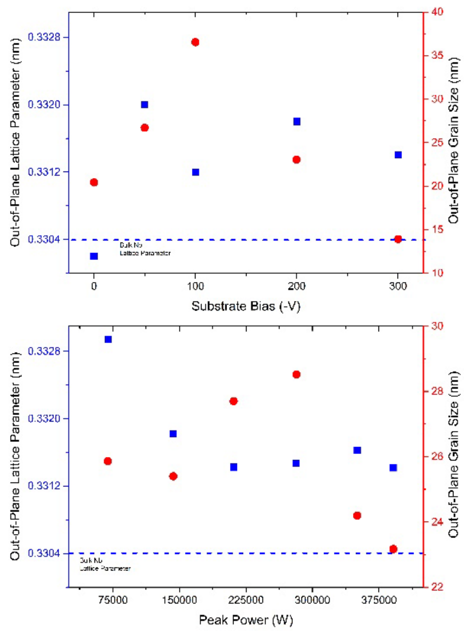

Figure 3: Plot of lattice parameter and grain size vs. (a) peak power in Nb/Cu power series and (b) applied bias in Nb/Cu bias series; horizontal dashed line included to represent the bulk Nb lattice parameter as reference.

After the system was shut down and had intensive maintenance performed in the spring of 2018, efforts were restarted to perform another sample series investigating the effect of substrate temperature on film properties. This effort also served a dual purpose of providing a quality check on the deposition system performance post-maintenance and gave feedback to confirm performance in-line with those previously observed in the power and bias series summarized above.

Temperature series samples were deposited at the same parameter set as the power and bias series with both the power and bias held constant at the "optimized" values inferred from the respective series analysis; "optimized" referring to attempting to achieve bulk-like microstructure and maintain good surface roughness and crystallinity. A list of temperature series deposition parameters and resulting sample properties is shown in Table 1.

After deposition, the samples were analysed using XRD to determine the resulting out-of-plane lattice parameter, grain size and mosaicity. The XRD scans exhibited similar behaviour to the power and bias series showcasing a polycrystalline texture dominated by the Nb (011) phase. Average grain sizes were obtained using the Scherrer equation and the FWHM of the Nb (011) peak.

Figure 5 shows the results from the XRD analysis of the samples; plotting both out-of-plane lattice parameter and grain size against the substrate deposition temperature. As can be seen, the samples exhibited no clear trend of lattice parameter with deposition temperature. However, the grain size shows a clear direct increasing trend with respect to temperature. This result is fully expected from the volume of research done on the relation between these two values.

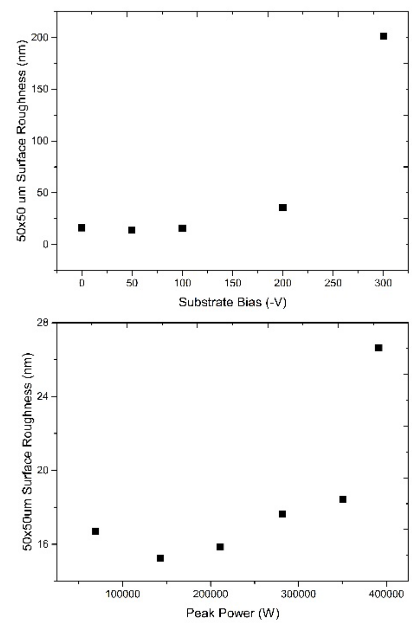

Figure 4: Plot of RMS roughness, on 50×50 μm scale, vs (a) peak power for Nb/Cu power series samples, and (b) applied bias for Nb/Cu bias series samples. Note the roughness scale difference compared to the power series results above.

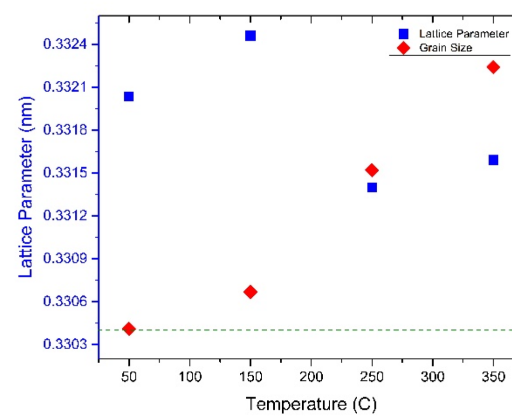

Figure 5: Plot of lattice parameter and grain size vs substrate deposition temperature. Dashed line included to represent bulk Nb lattice parameter value as reference.

One of the main takeaways from the temperature series is found in the analysis of the samples with respect to the previous power and bias series samples. The temperature series samples do not vary wildly from the previous sample properties. The lattice parameters and grain sizes are still well within the bounds of the previously measured values for each series. This result confirms the consistent and reproducible quality, from a microstructural perspective, of the films pre and post system maintenance.

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Table 1: Deposition Parameters and Film Properties of Interest for Temperature Series Samples

Temperature (C)	Peak Power (kW)	Applied Substrate Bias Voltage (V)	Out-of-Plane Lattice Parameter (nm)	Out-of-Plane Grain Size (nm)
350	332	25	0.3316	43.77
250	360	35	0.3314	36.24
150	377	25	0.3325	27.38
50	400	25	0.3320	24.69

Cavity RF Results

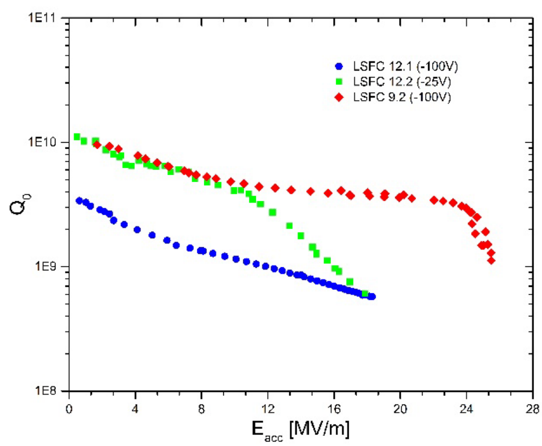

Figure 6: Quality Factor of 1.3 GHz HiPIMS Nb/Cu LSF cavities (B_{peak}/E_{acc}=3.68) at 2 K. No field emission was detected.

Nb/Cu RF cavity results, shown in Fig. 6, exhibit low field Q values in the mid to high 10^9 range and performing up to ~21 MV/m. After high pressure water rinsing, the Nb/Cu cavities had no detectable particulates during assembly, anecdotally relating to the fact that no field emission was detected during RF testing. The first Nb/Cu coating (LSFC 12.1) was hindered by a misalignment of the Nb cathode during coating, i.e. a non-uniformity in coating thickness. After cathode alignment corrections, the same coating parameters were applied to LSFC 9.2 which showed a net improvement. However, the film had a T_c of 9.5 K suggesting significant intrinsic stress in the film. In an effort to reduce the stress in the film, the next coating was performed at a lower bias (-25 V). The cavity (LSFC 12.2) exhibited a slight improvement at low field but had a mid-range Q-slope that seems to be substrate related.

It is important at this point to make a note on substrate manufacturing and preparation, it was discovered after these tests that continued removal from acid etching, in preparation for the next cavity coatings, revealed cracks in the cavity beam pipe seam welds, resulting from non-fully penetrating welds, which were able to be seen by eye; in some cases extending almost all the way to the iris. In response, the Cu cavities were re-inspected using a cavity optical inspection tool at JLab, an example of which is shown in Fig. 7. The results showed macroscopic pits and cracks present even in areas that they were not visible by eye.

This anecdotal and qualitative finding brings into question the reported RF results and may imply that these are lower bounds to the actual film performance. Cracks present during deposition could cause locally poor microstructure and, more importantly, areas where the film thickness is significantly thinner than expected. Areas with extremely thin Nb films or, in the extreme case, even uncoated Cu showing, could have the RF field probe the normal conducting Cu substrate resulting in significant deterioration of the measured RF performance.

Upon this discovery, cavity depositions were paused to allow time for extra electron beam welding to be performed on all Cu cavity substrates to fix the known bad seam welds and give confidence in welds where no cracks had been discovered yet, since the operating assumption at the time was that all seam welds would reveal cracks at some point since they were all manufactured using the same welding parameters. Even with the underlying substrate issues, these results show a gradual increase in performance of films deposited in this system and hold promise for future film manipulation for RF performance improvement.

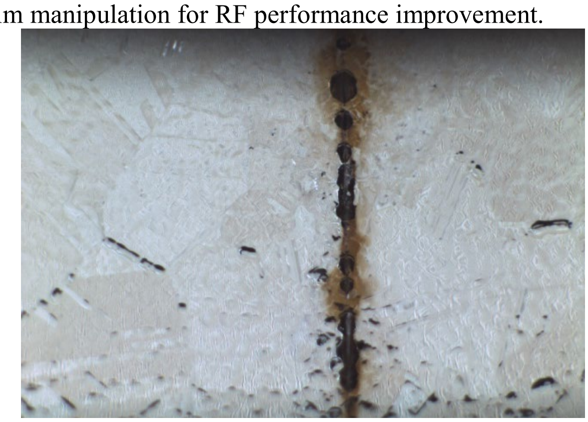

Figure 7: Representative image of seam weld cracks discovered in low surface field Cu cavities, LSFC-12 shown here, after SUBU acid etching.

CONCLUSION

The results presented here have shown that HiPIMS has great promise for high quality SRF thin films. Small coupon samples were produced with bulk-like lattice parameters and low roughness with implications for future surface engineering. System performance and reliability was confirmed after required system maintenance. The preliminary results on cavities have also shown great promise. We have achieved Nb thin film cavities with reasonable low-field Q values and remaining relatively flat up to moderate accelerating gradients. The reported Nb/Cu cavities exhibit performance in line or better than

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previously reported results at similar deposition parameters; even with using now known bad Cu substrates. Plans for the future of this project involve overcoming many known challenges such as: applying Cu electropolishing to cavity geometry, which is far superior to SUBU, and changing film deposition parameters throughout the coating process for active engineering of film properties throughout the thickness of the film. These results give a starting point to begin pursuing control over the flaws observed in the technology over the last 40 years leveraging the increased film deposition control and quality offered by energetic condensation methods.

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