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OPTIMISATION OF NIOBIUM THIN FILM DEPOSITION PARAMETERS FOR SRF CAVITIES *

D. Seal[1]†, G. Burt[1], J. Conlon[2], O.B. Malyshev[2], K. Morrow[2], R. Valizadeh[2]
Cockcroft Institute, UKRI/STFC Daresbury Laboratory, Daresbury, Warrington, UK
[1]also at Engineering Department, Lancaster University, Lancaster, UK
[2]also at ASTeC, UKRI/STFC Daresbury Laboratory, Daresbury, Warrington, UK

Abstract

In order to accelerate the progression of thin film (TF) development for future SRF cavities, it is desirable to optimise material properties on small flat samples. Most importantly, this requires the ability to measure their superconducting properties. At Daresbury Laboratory, it has been possible for many years to characterise these films under DC conditions; however, it is not yet fully understood whether this correlates with RF measurements. Recently, a high-throughput RF facility was commissioned that uses a novel 7.8 GHz choke cavity. The facility is able to evaluate the RF performance of planar-coated TF samples at low peak magnetic fields with a high throughput rate of 2-3 samples per week. Using this facility, an optimisation study of the deposition parameters of TF Nb samples deposited by HiPIMS has begun. The ultimate aim is to optimise TF Nb as a base layer for multilayer studies and replicate planar magnetron depositions on split 6 GHz cavities. The initial focus of this study was to investigate the effect of substrate temperature during deposition. A review of the RF facility used and results of this study will be presented.

INTRODUCTION

Superconducting radio frequency (SRF) cavities are typically made from niobium (Nb) because it has the highest critical temperature (T_c) and the highest superheating field (H_{sh}) of any element. These cavities are usually made of bulk Nb. However, given the increasing cost of liquid helium (LHe), electricity and accelerator infrastructure, copper (Cu) cavities coated with thin film (TF) superconductors (SC) is an area of growing attention for the SRF community. One focus of this research is on Nb TFs, because they provide an attractive and economical alternative to bulk Nb cavities. Nb on Cu in comparison to bulk Nb benefits from improved thermal stability as a result of a higher thermal conductivity of Cu. It is also less sensitive to trapped magnetic flux during cooldown, and allows for operation at 4.2 K with lower BCS resistance (R_{BCS}) than bulk Nb.

Nb TF cavities have been used successfully at CERN since the early 1980s having first been installed in the Large Electron-Positron (LEP) collider [1] and most recently on the High Intensity and Energy Isotope mass Separator On-Line facility (HIE-ISOLDE) [2]. Typically, these cavities have exhibited high quality (Q) factors at low fields comparable with bulk Nb, however they have suffered from high RF losses in medium to high field operation, known as Q-slope. Recent improvements to deposition techniques have produced Nb TFs with RF properties comparable to bulk Nb [3], resulting in improved accelerating gradients (E_{acc}) and significant reduction of the Q-slope. However, more studies are needed to understand the relationship between deposition parameters and RF performance to achieve Q factors comparable to bulk Nb at high field. This demonstrates the importance of mastering TF Nb depositions if Nb on Cu cavities are to be used in facilities requiring high E_{acc}.

The simplest way of develop coating technology is to begin with planar samples prior to full cavity depositions. This allows the use of simple deposition facilities, a low-cost sample design, easy visual inspections before and after, and analysis with surface characterisation instruments. After deposition, the samples have mainly been used for DC superconducting property evaluation. At Daresbury Laboratory, facilities for quick DC analysis of TF samples have existed for many years [4]. These are used to measure, T_c, the residual resistance ratio (*RRR*), [5] and the field of full flux penetration (B_{fp}) [6, 7]. However, these facilities are unable to analyse the behaviour of films under RF conditions, which is arguably the most important measurement. As a result, a custom facility was developed to measure the average RF surface resistance (R_s) of TF SC samples. Most importantly, the facility has a very high sample throughput of up to 3 per week, allowing for quick optimisation of RF performance [8, 9].

With an RF facility now available for quick sample analysis, optimisation studies of the deposition parameters of Nb TFs can be performed. The coating technique for these TFs has been high-power impulse magnetron sputtering (HiPIMS). This technique can produce higher density plasmas and denser films compared with conventional pulsed DC magnetron sputtering [10]. The ultimate aim is to develop the best parameters to be used as a Nb TF baseline for multilayer studies. After planar sample depositon, these parameters can be repeated on split 6 GHz, also designed and tested in-house, to analyse TF performance on a cavity-like geometry [11, 12].

The first step to Nb TF optimisation has been the effect of substrate temperature during deposition on RF performance. Previous studies have shown effects on DC properties [10, 13], however limited RF measurements were possible. Ex-

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† daniel.seal@cockcroft.ac.uk

perimental details of the depositions and results from sample characterisation will be reported in this paper.

EXPERIMENTAL

Sample Preparation

Six Nb TF samples were deposited on Cu disks of 100 mm diameter and 3 mm thickness. Given the impact of the preparation of the surface of the Cu substrate on the superconducting properties [14], all disks had to be polished prior to deposition. All were mechanically polished at STFC RAL Space prior to deposition with a 1 hour treatment using a single-point diamond turning lathe. Note that this method would not be possible for a cavity-like geometry. However, for the optimisation of planar samples, this was the best method available to ensure a consistent and repeatable surface finish between substrates prior to deposition with an average roughness of ≈ of a few nanometres. This meant that measurements of the superconducting TF properties would be the result of changes in film quality resulting from changes in the deposition parameters.

Each Nb film was deposited by HiPIMS using a Nb target mounted to a planar magnetron. The sample substrate was mounted on a rotating sample holder approximately 10 cm from the target. The substrate could be heated from room temperature (RT) to ≈ 650 °C corresponding to an increase in the heater current from 0 to 35 A. With the exception of a sample deposited at RT, all substrates were heated for 20 hours with the required heater current prior to deposition. The target was controlled with an Ionautics HiPIMS power supply using the deposition parameters listed in Table 1. Based on previous depositions, the thickness of these films was ≈ 3 μm.

One can see that the only variable deposition parameter was the heater current. The 6 samples were deposited with heater currents of: 0, 10, 15, 23, 30 and 35 A. Temperature measurements were not possible during deposition; therefore, the sample temperature was estimated on the basis of past measurements. This will be accurately measured in a dedicated experiment in the future.

Table 1: The Deposition Parameters used for This Study

Deposition Parameter	**Value**
Base pressure (mbar)	10^{-9}
Initial heating time (h)	20
Initial heating current (A)	0 – 35
Pressure with heating (mbar)	10^{-7}
Average target power (W)	400
Pulse duration (μs)	80
Pulse frequency (kHz)	1
Discharge gas	Kr
Deposition pressure (mbar)	2×10^{-3}
Substrate heater current (A)	0 – 35
Substrate temperature (°C)	RT to ≈ 650
Deposition time (h)	4.5

RF Facility

After depositions, each sample was characterised under RF conditions by measuring R_s at sample temperatures 4.2 K $\leq T_s \leq$ 10 K and low peak magnetic fields, $B_{s,pk} \leq 1$ mT. The aim was to find the deposition parameters that minimise R_s close to R_{BCS}, whilst also minimising the temperature independent residual resistance, R_{res}, where $R_s = R_{BCS}(T) + R_{res}$. This is important to minimise RF losses. The values of R_{BCS}, were calculated in SRIMP [15, 16], for a typical set of TF Nb parameters, and are shown in Fig. 1. This shows a clear relation between R_{BCS} and *RRR*. The *RRR* is a measure of the purity of the film and the result of the deposition parameters, which is why it is important to optimise the substrate temperature for this study. An understanding of R_{res} gives a good indication of losses due to overall sample quality (e.g. cleanliness, roughness, oxides, etc.).

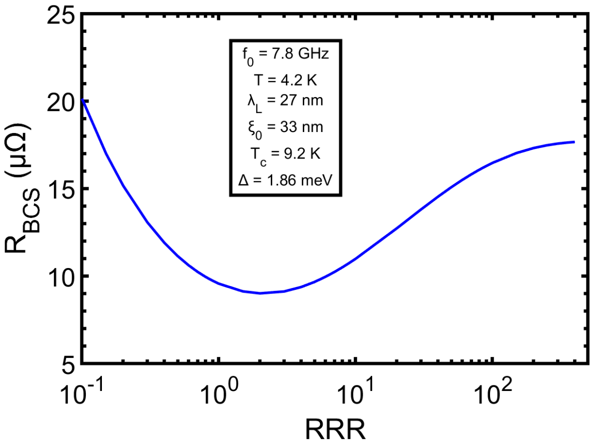

Figure 1: BCS resistance as a function of *RRR* calculated in SRIMP with the material parameters shown [15, 16].

Details of the facility used are detailed in Refs. [8, 9]. It uses a two-part test cavity: a choke cavity made up of a half-cell elliptical bulk Nb cavity surrounded by quarter-wavelength RF chokes, and the TF coated Cu disk. It operates at 7.8 GHz in the TM_{010} mode. The purpose of RF chokes is to allow the choke cavity and sample to be physically and thermally isolated whilst minimising RF leakage. This enables a simple measurement of R_s using an RF-DC compensation method (described in Ref. [17]). Given the wide bandwidth of the test cavity (≈ few kHz), the RF power was input from a VNA without the need for a phase-locked loop.

The test cavity is mounted in a LHe-free cryostat operated with a Gifford-McMahon cryocooler. Figure. 2 shows the choke cavity and sample, as well as their mounting to the stage 2 plate. The samples are indium-brazed to a Cu sample holder prior to mounting. Simple mounting and removal of the samples, as well as the use of a LHe-free cryocooler, allowed for a rapid 2 day turnaround time per sample. This allowed a quick direct comparison of R_s of each of the samples.

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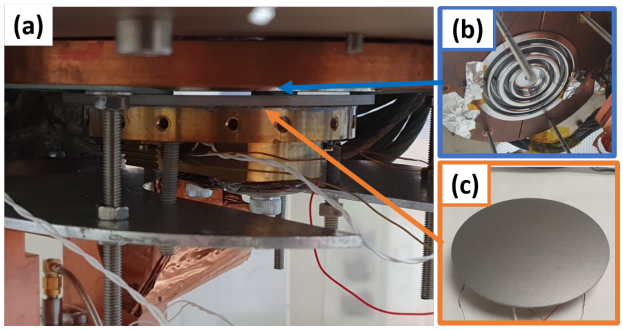

Figure 2: The RF facility for planar sample testing: (a) the choke cavity mounted to stage 2 with the sample underneath, (b) choke cavity, (c) a TF Nb sample to be tested.

In addition to measurements of R_s, a low-power sweep with increasing T_s close to T_c allows one to track the shift in the resonant frequency (Δf). This is carried out while keeping the temperature of the choke cavity constant. From this an estimation of T_c can be made from where Δf starts to level out. A good-quality film would probably have a $T_c \approx 9.2$ K (i.e. close to bulk Nb). It is also possible to extract material parameters from a fit of this curve [18], however, this will only be performed after further superconducting analysis, which will be reported at a later date.

Other Facilities

Additional analysis of these samples has been carried out. Before and after deposition, the samples were analysed using a white-light interferometer (SmartWLI). Measurements of the mean arithmetic roughness (S_a) and the root mean squared roughness (S_q) were calculated. The samples for these averages were taken using a 20 x objective that allowed areas of 909.4 µm × 567.5 µm to be measured at 10 mm intervals from 25 to 75 mm from the centre of the sample.

RESULTS

RF

Measurements of R_s as a function of T_s for the 6 samples are shown in Fig. 3. A substrate heater current of 0 A corresponds to a room temperature deposition, while 35 A is ≈ 600 – 650 °C. It can be seen that the 30 A deposition overall produced the lowest R_s closest to R_{BCS} while the 0 A deposition produced the highest R_s, indicating a significant R_{res} contribution. At 4.2 K, $R_s = 19.9 \pm 2.7\,\mu\Omega$ for the 30 A sample compared to almost an order of magnitude higher for the 0 A sample with $R_s = 181.2 \pm 13.8\,\mu\Omega$.

Given that the aim is to operate TF Nb cavities at 4.2 K, R_s as a function of the substrate heater current has been compared at this temperature. Figure 4 shows a summary of R_s as a function of the substrate heater current for the 6 samples. In addition to 4.2 K, measurements are also shown at T_s = 7 K and 9 K. The reported R_s measurements have uncertainties of ≈ 15%.

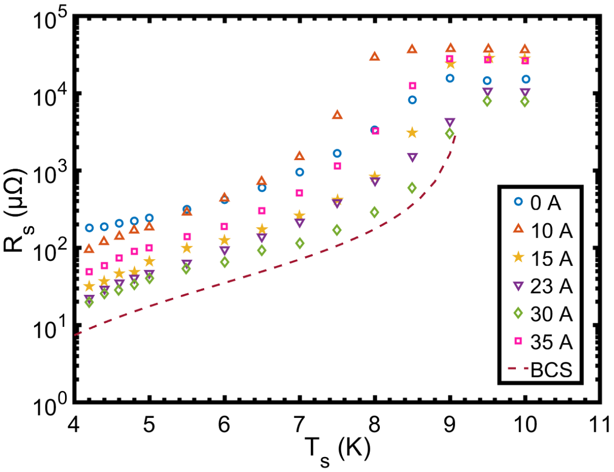

Figure 3: Surface resistance as a function of sample temperature for all of the samples.

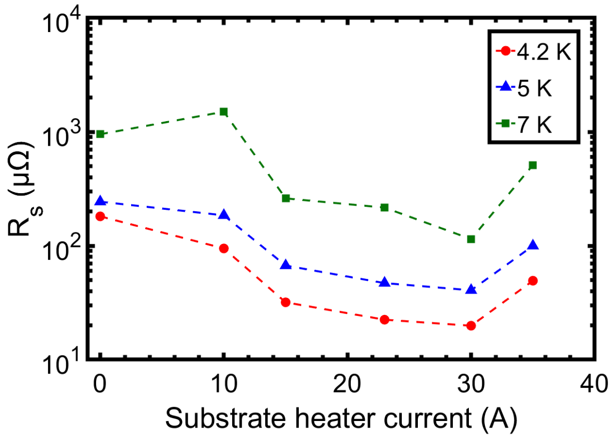

Figure 4: Surface resistance as a function of substrate heater current measured at sample temperatures of 4.2 K, 5 K, and 7 K.

These results show a relationship between substrate temperature during deposition and R_s. The sample deposited with a heater current of 30 A has the lowest R_s for all three sample temperatures shown. In addition to an increase in R_s for heater currents below 30 A, an increase in R_s was observed for higher temperature deposition using a heater current of 35 A (corresponding to ≈ 575 – 625 °C). For this sample $R_s = 49.3 \pm 7.0\,\mu\Omega$ at 4.2 K. Based on the results of the sample deposited at 23 A, it can be inferred that a substrate heater current of 23 – 35 A produced the lowest R_s, corresponding to a deposition temperature of ≈ 375 – 625 °C.

The final set of RF measurements is shown in Fig. 5. This shows the measurements of Δf as a function of T_s. The largest maximum Δf = - 460 kHz was observed for the 10 A sample, which also had the lowest $T_c \approx 8.2$ K. The lowest maximum Δf = - 90 kHz for the 30 A sample, which also had a $T_c \approx 9.1$ K.

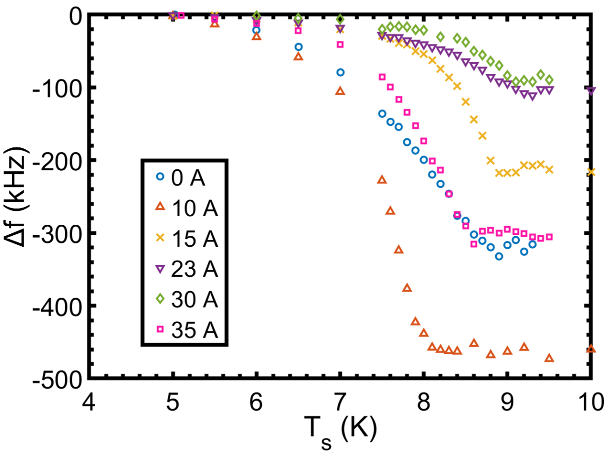

Figure 5: Resonant frequency shift as a function of sample temperature for all of the samples.

Surface Analysis

Prior to deposition, the average roughness of a sample of the diamond turned Cu substrates was measured. The results of these measurements showed that the disks were consistently polished to a low roughness of $S_s = 2 - 3$ nm and $S_q = 5 - 6$ nm.

After deposition and post-RF characterisation, the average roughness of all 6 samples was re-measured and the results are presented in Fig. 6. Both the average S_a and S_q had increased after deposition as expected. Now, $S_a = 11.8 - 44.3$ nm and $S_q = 29.6 - 204.9$ nm. Overall, there was an increase in average roughness as a function of the heater current to a maximum of $S_a = 44.3 \pm 3.1$ nm and $S_q = 204.9 \pm 43.1$ nm at 30 A. The sample deposited at 10 A had the lowest average roughness of $S_a = 11.8 \pm 0.1$ nm and $S_q = 29.6 \pm 1.5$ nm.

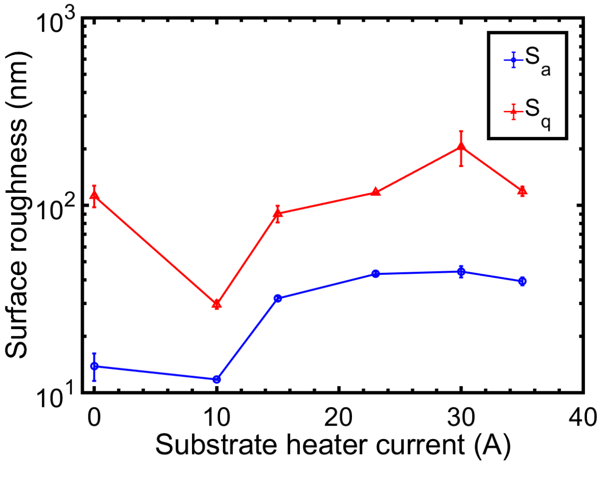

Figure 6: Surface roughness as a function of substrate heater current showing both S_a and S_q.

DISCUSSION

For the first time, it has been possible at Daresbury Laboratory to carry out a study on TF optimisation with the focus being on RF performance. A study of this kind has demonstrated the enormous benefit of having a high turnover facility for a rapid RF evaluation. The testing of up to 3 samples per week has allowed for quick analysis of how substrate temperature affects RF performance. A total of 6 Nb on Cu films have been reported in this paper; therefore, the total RF testing time, including sample handling, facility operation, and measurements was only 2-3 weeks.

The results have shown that the sample deposited with a substrate heater current of 30 A performed best under RF conditions with an R_s close to theoretical R_{BCS}. A clear relationship has been demonstrated between R_s and the heater current of the substrate, although there is quite a large variation between the samples, which was somewhat unexpected. The sample deposited at RT (0 A) had an order of magnitude higher R_s than the sample deposited in 500 – 550 °C (30 A). A visual inspection of the RT sample found significant pitting on the film that could have contributed to increased losses. However, given that the sample with the highest S_a and S_q was also the sample with the lowest R_s, it is unlikely that the variation in roughness will explain the differences in R_s alone. It is possible that a roughness effect is seen at higher RF fields; however, this was not possible with this facility. Ultimately, further surface analysis will need to be performed to fully understand these results.

It should also be noted that the samples were not deposited in order of increasing the heater current. The order of depositions was: 0, 30, 35, 23, 15 and 10 A. Unfortunately, after the 10 A sample, it was observed that the Nb target had worn through to the magnets behind and thus further depositions were not possible. The original plan was to find whether the minimum R_s occurred for substrate heater currents in the range of 23 – 30 A or 30 – 35 A; however, this was not possible. The measurements in Fig. 4 show a deviation from the trend for the 10 A sample, which could have been due to the target failure that affected the quality of the film. It is not yet known whether the target, which was close to failure, caused an increase in R_s for all samples. A second Nb target will be used for further depositions to compare the results with those shown in this paper and to analyse the repeatability of the best-performing sample.

Having analysed the RF performance and roughness, samples will be cut to meet the sample size requirements for the DC analysis and surface analysis facilities at Daresbury Laboratory. This has also been made possible by using a simple, cost-effective sample design. DC measurements of B_{fp} will be performed, as well as the analysis of *RRR* and T_c, which will give a better understanding of how these properties are affected by the depositon temperature. In addition, in depth surface analysis (i.e. XPS, SIMS, XRD, SEM etc.) of these cuts will allow for analysis of surface morphology and chemistry to see whether any correlations can be made with RF results.

The ultimate reason for this study was to enable the quick development of the best performing Nb film under RF conditions (i.e. lowest R_s at 7.8 GHz at 4.2 K). However, despite the high throughput rate of the RF facility, the measurements were limited to low field ($B_{s,pk} < 1.2$ mT) due to the high cavity resonant frequency and the limited thermal capacity of the cryocooler. Future upgrades and moving to an RF test bunker will allow for fields up to 15 mT, overlapping with QPR. Despite this, the optimisation study performed has been shown to be a useful way to rapidly analyse the best deposition temperature for Nb films in order to optimise RF performance. Overall, a test such as this will serve two purposes in comparing planar magnetron depositions:

1. The best deposition parameters can be repeated on a quadrupole resonator (QPR), allowing high-field measurements at three frequencies close to typical cavity frequencies [19].
2. The best deposition parameters can be repeated on a 6 GHz split cavity to analyse the RF performance on a curved cavity-like geometry [11, 12, 20], while also allowing visual inspection of the film.

Once the Nb deposition parameters have been optimised for low-field RF performance, the best parameters will be used as a baseline TF layer for multilayer studies. In this, higher T_c SC depositions will be performed on top (e.g. NbTiN, Nb_3Sn, V_3Si [21]) with the aim of screening magnetic fields $B > B_{sh,Nb}$. SC optimisation studies will then be performed, starting with the choke cavity.

CONCLUSIONS

For the first time at Daresbury Laboratory, it has been possible to use a recently developed RF facility as the primary means of characterising thin films under RF conditions as step to optimising deposition parameters. The use of a novel choke cavity as well as a simple sample design has allowed for a rapid sample turnover of 2-3 samples per week. This has enabled the rapid optimisation of one of the deposition parameters, namely the substrate temperature, against the RF properties of the deposited Nb thin films. Each sample was tested under low-power RF conditions using the choke cavity at 7.8 GHz, $B_{s,pk} \leq 1.2\,\text{mT}$ and $4.2\,\text{K} \leq T_s \leq 10\,\text{K}$. The lowest $R_s = 19.9 \pm 2.7\,\mu\Omega$ was shown for the sample deposited with a sample heating current of 30 A ($\approx 500 - 550\,^\circ\text{C}$). Results were compared with roughness measurements, which showed no correlation with R_s, therefore further surface analysis will have to be performed in the future. In addition, SC DC analysis as well as an accurate calibration of the sample temperature as a function of the heater current will be performed. A second set of Nb on Cu samples with a new target are being prepared for future analysis and comparison using the same procedures.

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