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THE ROLE OF OXYGEN CONCENTRATION IN ENABLING HIGH GRADIENTS IN NIOBIUM SRF CAVITIES

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Abstract

We studied the role of O concentration with depth in the performance of Nb SRF cavities. An ensemble of electropolished 1.3 GHz cavities, which initially showed high field Q-slope (HFQS), was subjected to sequential testing and treatment with *in-situ* low temperature baking at various temperatures. We find that increasing the bake duration causes (i) an increase in the onset of HFQS until it is absent up to quench (ii) a non-monotonic relationship with the quench field (iii) an evolution of the R_{BCS} toward a non-equilibrium behavior that drives anti-Q slope. Our data is qualitatively explained by assuming an O diffusion model and suggests that the mitigation of HFQS that arises from 120 C *in-situ* LTB is mediated by the diffusion of O from the native oxide which prevents the precipitation of proximity-coupled Nb nano-hydrides, in turn enabling higher quench fields. The decrease in quench field for cavities in which O has been diffused >90 nm from the RF surface may be due to a reduction of the field limit in the SS bilayer structure. We also suggest that the evolution of the R_{BCS} occurs due to the absence of proximity coupled inclusions, bringing about non-equilibrium effects.

INTRODUCTION

Recent work by Romanenko *et al.* on Nb SRF cavity cutouts has highlighted the involvement of oxygen diffusion in the 120 C bake effect [1,2], which describes the mitigation of high field Q-slope (HFQS) [3] by *in-situ* low temperature baking (LTB) fully assembled and evacuated SRF cavities at 120 C for a period of 48 hours [4]. The work shows that the 120 C LTB diffuses oxygen from the native niobium oxide according to Fick's law [5] and traps free hydrogen present within the niobium lattice [6], thereby inhibiting the precipitation of poorly superconducting niobium nanohydrides upon cool down to cryogenic temperatures. This results in a lower volume fraction or elimination of nanohydrides that in turn yields a larger proximity breakdown field, shifting up or eliminating the onset of HFQS. However, a systematic study that investigates the evolution of HFQS and cavity performance with increasing oxygen diffusion depths has not yet been reported.

Using the findings of Romanenko *et al.* as a driver, in this contribution, we report a systematic study on the effect of gradually increasing the depth to which oxygen diffuses on the RF performance of 1.3 GHz bulk niobium TESLA-shaped SRF cavities and show that oxygen plays a key role in enabling exceptional cavity performance. We find that the field of HFQS onset varies linearly with increasing oxygen diffusion depth, supporting the Romanenko model of HFQS and the 120 C baking effect. We also show that there exists a non-monotonic dependence of the quench field with O diffusion depth. Furthermore, we present a gradual evolution of the RF performance toward a doped-like anti-Q slope behavior that stems from a decrease in the BCS resistance with field after *in-situ* baking at 200 C for 6 hours, suggesting that O impurities may bring about these effects.

EXPERIMENTAL

We used an ensemble of TESLA-shaped 1.3 GHz Nb single cell SRF cavities that were initially treated with a bulk removal via electropolishing (EP) from the inner RF surface post initial fabrication and an 800 C degas step [4]. All cavities were then subjected to a 40 μm removal of the inner surface via standard EP. The cavities were then assembled for testing and evacuated. Most cavities were tested at this point to get a baseline, which will be referred to as "EP."

Cavities were subjected to sequential rounds of *in-situ* low temperature baking at various temperatures ranging from 90 C up to 200 C, diffusing oxygen from the native oxide toward the bulk according to Fick's law. After each step of treatment, cavities were RF tested at a temperature of 2 K. Most tests were repeated at <1.5 K to enable decomposition of the BCS and residual resistances [7]. To minimize the possibility of trapping of magnetic flux through superconducting transition, we used Helmholtz coils to provide a zero field environment and a fast cool down protocol to produce a sufficient thermal gradient along the length of the cavity [8].

Some cavities developed field emission after several rounds of treatment and testing. We found that high pressure rinsing effectively removed field emission without deleteriously affecting cavity performance. Only tests that showed minimal field emission are presented here.

While we extensively tested 7 cavities, we explicitly report the results of only three and present the rest in the form of statistics in the Discussion section.

Effect of Sequential 120 C Baking

Due to its wide use, we first studied the effect that sequential baking at 120 C has on cavity performance; the results are shown in Fig. 1. The baseline EP test, which was only performed at 2 K, showed the expected onset of HFQS at 25 MV/m. The test was power limited. After baking at 120 C for 30 minutes, the HFQS onset shifted up to 27 MV/m but we were still limited by available RF power. Following an additional 30 minute bake, for a net treatment of 120 C for 1 hour, HFQS onset shifted further up to 29 MV/m and quench occurred at 34.4 MV/m. Subsequent treatment and testing showed that both the field of HFQS onset and quench field increased with integrated bake duration.

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The middle plot in Fig. 1 shows that residual resistance remains below $\sim$5 nΩ for most tests before the sharp takeoff due to HFQS occurs. Like the 2 K Q_0 vs E_{acc} tests, the point of HFQS onset increases with integrated bake time. Note that the <1.5 K test post 120 C × 8 hours showed a reduced quench field that occurred after quenching at 2 K.

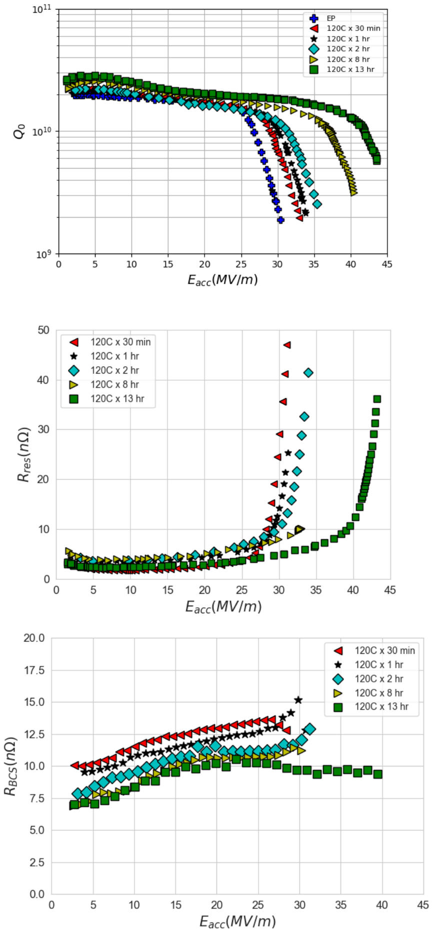

Figure 1: Q_0 vs E_{acc} at T = 2 K, R_{res} a T < 1.5 K, and R_{BCS} at T = 2 K of TE1RI003 after sequential *in-situ* baking at 120 C. Legend durations represent integrated baking times.

The lower plot in Fig. 1 shows the decomposed BCS resistance at 2 K. The test after 120 C × 30 min showed the otherwise expected increase in R_{BCS} with field due to greater dissipation introduced by increased screening currents. Baking for an additional 30 minutes, for a total of 1 hour, showed a slight decrease in R_{BCS} at all fields while maintaining an almost linear increase with field. After an additional hour of baking, for a total of 2 hours, the BCS resistance decreased further. At this point, there was an onset of a slight "knee" at about 22 MV/m, showing a saturation of the BCS resistance up to 26 MV/m before increasing again. After baking for a total of 8 hours, the cavity displayed a slightly lower BCS resistance and maintained this saturating behavior. The test after a net treatment of 120 C × 13 hours showed that the resistance *decreased* after the knee at 22 MV/m and leveled off. This slight decrease in R_{BCS} is reminiscent of the decrease in BCS resistance with field in nitrogen doped cavities [9] that has been suggested to have origins rooted in non-equilibrium superconductivity [10].

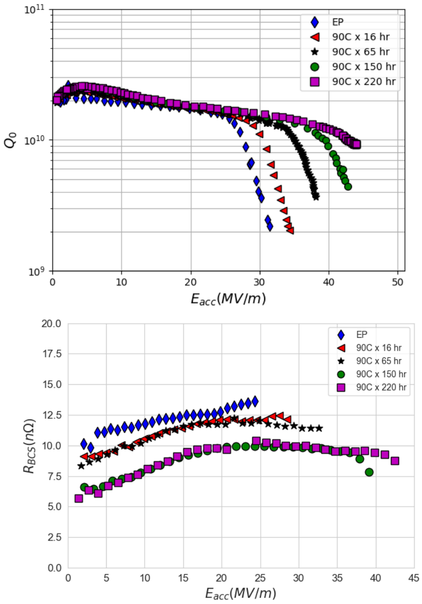

Figure 2: Q_0 and $R+BCS$ vs E_{acc} at T = 2 K of TE1AES014 after sequential *in-situ* low temperature baking at 90 C.

Effect of Sequential 90 C Baking

To ensure that the results observed in the previous section were reproducible with other temperatures, we treated cavity TE1AES014 with sequential bakes at 90 C. The results are shown in Fig. 2. The baseline EP test showed the expected onset in HFQS at 26 MV/m at <1.5 K and the maximum achievable gradient was limited by available power from the amplifier. After baking the cavity at 90 C for 16 hours, the HFQS onset increased by 2 MV/m and the cavity quenched at 34.3 MV/m. Similar to what was observed in Fig. 1, increasing the bake duration showed continual increase in both HFQS onset and quench field. An integrated bake duration of 220 hours showed almost no HFQS up to quench. The BCS resistance also followed the trends observed in Fig. 1, evolving toward lower values at all fields and developing a knee at 22 MV/m after which the BCS decreased. While not shown here, the R_{res} also showed all expected behaviors.

Effect of Large O Concentrations/Diffusion Depths

Cavity Measurements We next investigated the effect that diffusing large levels of oxygen from the oxide has on cavity performance. We began by subjecting cavity TE1AES019 to a 90 C *in-situ* low temperature bake for 12 hours after the initial EP step. The Q_0 vs E_{acc} results in blue in Fig. 3 showed HFQS onset at 29 MV/m; the cavity quenched at 36.2 MV/m at low temperature (not pictured). The cavity then underwent a very long bake at 90 C with an

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integrated duration of 384 hours which showed *no HFQS up to quench at* 43 MV/m. We then baked the cavity further at 200 C for 1 hour. The RF results in Fig. 3 showed that the quench field reduced to 38 MV/m. Furthermore, the Q_0 at all fields decreased and the curve shape showed a slight rounding at mid-field. Continued baking at this temperature for an additional 5 hours showed the emergence of anti-Q slope and a further reduction of the quench field. These behaviors agree with data presented in [11]. We note that the cavity was never HPR'd or opened after any of these tests, preventing the formation of a new oxide.

The R_{res} increased substantially with treatment, likely due to the growth of suboxides [12]. This well explains the moderately low Q_0 after the +200 C × 6 hour treatment.

The R_{BCS} evolved according to the trends observed in Figs. 1 and 2 wherein a knee at 22 MV/m emerged after baking at 90 C for 384 hours. Baking for an additional 200 C for 1 hour showed that this knee shifted down to ~12 MV/m and a substantial reduction in R_{BCS} occurred up to 30 MV/m. The final round of treatment showed that the knee was replaced by a reversal of the BCS resistance with field which explains the origin of the anti-Q slope observed in the corresponding Q_0 vs E_{acc} curve.

SIMS Measurements As such anti-Q slope behavior is characteristic of cavities processed with surface treatments that introduce uniform and dilute concentrations of nitrogen into the RF layer (nitrogen doping [9] and mid-temperature baking [13]), we suspected that the 200 C low temperature baking treatment was somehow introducing nitrogen into the niobium lattice. To investigate, we performed time-of-flight secondary ion mass spectrometry (TOF-SIMS) on an as received EP cavity cutout after baking the sample *in-situ* at 205 C for 3 hours and 19 hours without ever losing vacuum, replicating the surface treatments used on TE1AES019. We used the same IONTOF TOF-SIMS system described in [1] with a liquid Bi^+ ion beam to perform secondary ion measurements while sputtering with a Cs^+ ion gun with energy 1 keV to obtain impurity profiles with depth. The results are shown in Fig. 4.

While the as received EP sample in Fig. 4 exhibited little to no oxygen ion signal just below the oxide, *in-situ* baking at 205 C for increasing durations showed an increasing oxygen signal with depth, following a Fick's diffusion profile. Longer durations showed a slight decrease in O^- ion signal just below the native oxide. The NbH^-/Nb^- signal, which we interpret as a measure of the concentration of free hydrogen in the niobium lattice, showed a very slight decrease with increasing bake durations. On the other hand, the OH^-/Nb^- signal, which may reflect the oxygen bound hydrogen signal, increased with bake duration and showed good correspondence with the oxygen signal, achieving bulk value concentrations after similar sputter times. Surprisingly, the NbN^-/Nb^- ion signal, which we use as a measure of nitrogen concentration, changed very little throughout the tests. After an integrated bake duration of 19 hours at 205 C, the NbN^-/Nb^- ion signal increased just slightly above the as received measurement. We note that while the native niobium pentoxide ion signal decreased after baking, it remained appreciable, suggesting that the native oxide layer

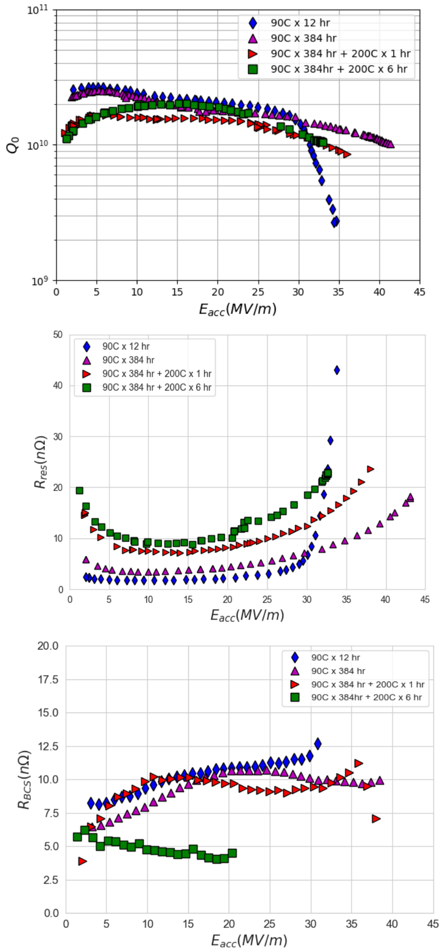

Figure 3: Q_0 and R_{BCS} vs E_{acc} at T = 2 K and R_{res} at <1.55 K of TE1AES019 after *in-situ* baking treatments.

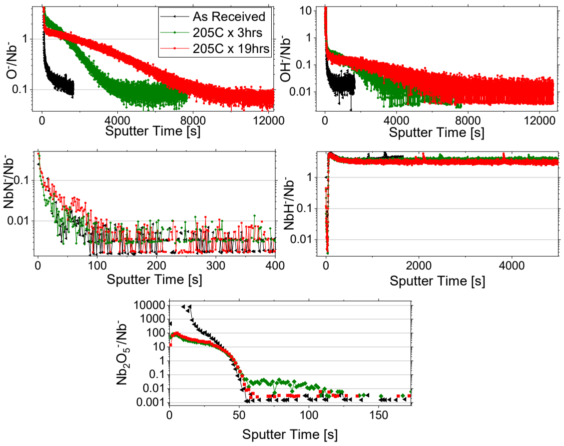

Figure 4: TOF-SIMS acquired depth profiles on an as received EP cavity cutout after *in-situ* baking at 205 C. All signals are normalized point-by-point to the Nb^- ion signal.

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stayed mostly intact throughout the measurements, and likely served as a passivating layer against external impurities.

To summarize the SIMS results, for the durations studied here, the 205 C LTB does not break up the native oxide and introduce nitrogen into the RF surface, thus suggesting the role of oxygen in the doping behavior observed in Fig. 3.

DISCUSSION

Using the diffusion equations presented in [5] with a built-in bulk concentration term, we calculated the O concentration depth profile and plotted the depth at which it achieved the bulk value (see inset in Fig. 5) against the field of HFQS onset at $T < 1.5$ K and the highest achieved quench field for each cavity test; results are shown in Fig. 5. We note that the O depth profiles were calculated assuming that the concentration at the oxide-metal interface remained constant at the solubility limit C_0, which was observed in Fig. 4 to not be strictly true for elevated temperatures and durations.

We find that the field of HFQS onset varies linearly with the depth to which oxygen diffuses for all studied temperatures. This supports the role of oxygen in the mitigation of HFQS in the 120 C baking effect as presented by Romanenko *et al.* and suggests that as oxygen diffuses deeper, it captures free hydrogen and inhibits the formation of proximity coupled nano-hydrides. The observations made from SIMS further support this hypothesis.

We mention that the usual 120 C bake for 48 hours introduces oxygen to a depth of ~100 nm; extrapolating from Fig. 5 shows that HFQS should be absent up to 50 MV/m in these cavities, which agrees with experimental data [14].

We also find that the quench field follows a non-monotonic relationship with the calculated depth at which oxygen concentration achieves the bulk value, showing a peak near 90 nm. The initial increase in quench may be understood by assuming the hydride model: fewer hydrides in the RF layer results in less RF heating that might otherwise drive thermo-magnetic quench. The subsequent decrease in quench may be explained by the reduction of the theoretical field limit due to an increasingly thicker dirty layer in the superconductor-superconductor (SS) bilayer structure, which achieves peak field when the dirty layer is roughly half the thickness of the effective penetration depth [15]. As such, the quench mechanism in this regime may stem from magnetic origins, and the non-monotonic dependence of the quench field may be due to the interplay between these two regimes.

The transition from magnetically driven to thermo-magnetically driven quench for varying concentrations and/or dirty layer thicknesses agrees with previous TMAP studies after sequential nitrogen treatments [16]. A cavity post N-doping that originally showed quench stemming from magnetic origins and anti-Q slope later exhibited HFQS and evidence for a thermo-magnetically driven quench after removing part of the RF surface *via* EP and thus eliminating some of the dirty layer/decreasing the concentration of impurities in the RF layer. This suggests that the interplay between magnetic and thermo-magnetic quench mechanisms may exist for niobium SRF cavities that contain either oxygen or nitrogen impurities and that both impurities play similar roles in cavity performance.

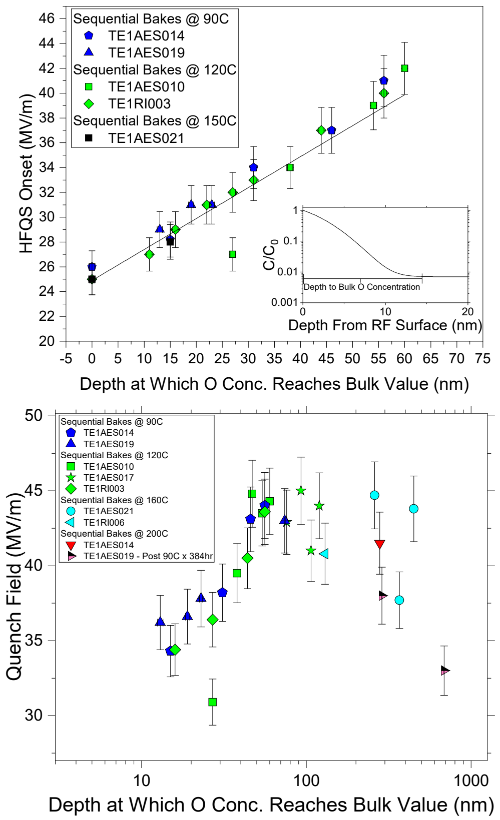

Figure 5: (Upper) The field of high field Q-slope onset at T < 1.5 K and (lower) quench field of cavities subjected to sequential *in-situ* baking at various temperatures plotted against the calculated depth to which the diffused O achieved the bulk value. Points at 0 nm come from baseline EP tests.

Furthermore, the observed evolution of the Q_0 and R_{BCS} behaviors of TE1AES019 in Fig. 3 towards those that resemble N-doped cavities *in the absence of N* suggests that diffused O may drive the non-equilibrium BCS behavior that enables the anti-Q slope in SRF cavities in the same way that interstitial nitrogen produced *via* N-doping does, which is consistent with what has been recently shown in [17].

One possible mechanism that could drive such doping behavior may stem from the theory that both oxygen and nitrogen capture free hydrogen present in the niobium lattice, as suggested by references [1, 6] and Fig. 4, and may prevent the precipitation of niobium nano-hydrides. Thus, the anti-Q slope (or non-equilibrium BCS behavior) may be an inherent behavior to "pristine" Nb that is void of deleterious proximity coupled inclusions. This is reinforced by the observation that N-doped cavities that exhibit this non-equilibrium behavior have been found to exhibit lower inelastic scattering parameters than EP cavities that exhibit the HFQS phenomenon [18, 19].

We note that the present contribution does not fully decouple the role of O concentration in the RF layer and the depth to which oxygen diffuses in the observed doping behavior present after low temperature baking. The full delineation of these two parameters will be a subject of future work.

CONCLUSIONS

In this work, we have investigated the role of oxygen in the performance of niobium SRF cavities after *in-situ* low temperature baking and found that it plays a crucial role in mitigating HFQS, driving high quench fields, and maybe even enabling anti-Q slope after these treatments. Our data is qualitatively well explained by O diffusing from the native niobium oxide and capturing free H that results in the elimination or lower volume fraction of proximity coupled niobium nano-hydrides. Our work shows that any *in-situ* LTB treatment may be used to eliminate HFQS as long as it introduces O sufficiently deep toward the bulk from the native Nb oxide. We show that the quench field follows a non-monotonic relationship with the depth to which oxygen diffuses and suggest a possible interplay of thermo-magnetic and magnetic quench mechanisms. In addition, cavity studies and TOF-SIMS performed on as received cavity cutouts show that *in-situ* LTB at 200 C for 6 hours introduces a large concentration of oxygen deeper toward the bulk of the niobium matrix and effectively recreates RF performance that is characteristic of N-doped cavities. This suggests that oxygen impurities may play a role in the performance of SRF cavities after low temperature baking that is similar to that of nitrogen after nitrogen doping. In light of this, we suggest that the anti-Q slope may be an inherent behavior to SRF cavities that arises in the absence of proximity coupled niobium nano-hydrides, which are mitigated by doping with oxygen, nitrogen, or possibly other impurities.

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