

FUNDAMENTAL STUDIES OF IMPURITY DOPING IN 1.3 GHz AND HIGHER FREQUENCY SRF CAVITIES*

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Abstract

As the demand for more powerful, more efficient, and smaller superconducting RF accelerators continues to increase, both impurity doping and high-frequency cavities (>1.3 GHz) have become hot topics for fundamental research because of their potential to significantly decrease surface losses and cost respectively. In this report, we present recent experimental and theoretical results on undoped and nitrogen-doped high-frequency cavities and on alternative doping agents in traditional 1.3 GHz cavities, with a focus on understanding the fundamental science of impurity doping.

“2/6 DOPING” AT 2.6 GHz

Impurity doping is a technique for preparing niobium superconducting radio-frequency (SRF) cavities that can increase the quality factor Q by a factor of two or more at low field and cause an “anti- Q -slope” in which Q rises as the cavity field increases [1–3]. This Q rise has been shown to increase in strength with increasing frequency [4]. At IPAC’18, we presented Cornell’s first results of an impurity-doped 2.6 GHz niobium cavity [5, 6]. This ILC/TESLA-shape cavity [7] ($B_{pk}/E_{acc} = 4.28 \text{ mT/(MV/m)}$) was treated with the so-called “2/6 doping”: a degas step in vacuum for 5 hours, followed by an injection of 40 mTorr (6 Pa) of continuously flowing N₂ gas for 2 minutes, followed by a vacuum anneal step for 6 minutes, all at 800 °C. This is followed by 6 μm of surface removal by vertical electropolish (VEP). In the initial test, the cavity showed a strong field-dependent reduction in the temperature-dependent “BCS” portion of the surface resistance.

In this paper, we present the results of additional study of this cavity, investigating the sensitivity of the temperature-independent “residual” surface resistance R_0 to trapped magnetic flux. This sensitivity has been studied extensively in 1.3 GHz nitrogen-doped cavities, where strongly doped cavities (electron mean free path $\ell < 100 \text{ nm}$) exhibit up to 5 nΩ of residual resistance per mG of magnetic flux trapped during cooldown [3, 8]. To determine this sensitivity, we record R_0 as a function of field for several different amounts of trapped flux $B_{trapped}$ and subtract the non-flux-dependent R_0 present in a cooldown with no flux trapped (typically a “fast” cooldown with a cross-cavity temperature gradient $\nabla T > 200 \text{ K/m}$). We then take $R_0/B_{trapped}$ and perform a linear fit over the RF field strength to yield an overall offset

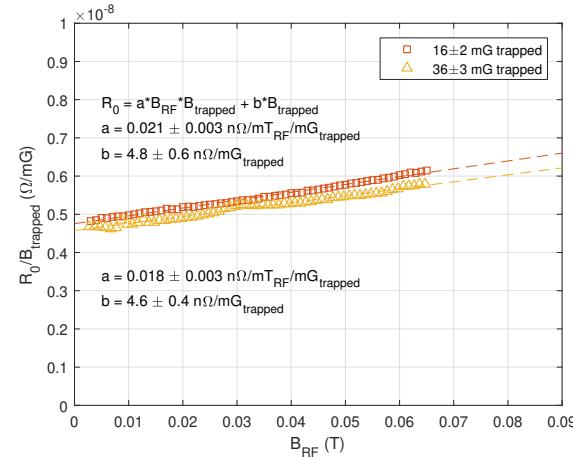


Figure 1: Sensitivity of the residual resistance due to trapped magnetic flux for two cooldowns of a single 2/6-doped 2.6 GHz niobium cavity.

sensitivity as well as a field-dependent sensitivity. Previous analyses of 1.3 GHz N-doped cavities largely considered only the offset parameter, *e.g.* the 5 nΩ/mG figure above; analyses of 1.3 GHz Nb₃Sn cavities have included similar two-parameter linear fits [9].

Figure 1 shows these results for two cooldowns of the 2/6-doped 2.6 GHz cavity, which had an $\ell = 46 \text{ nm}$. The linear fit parameters for both tests (with $16 \pm 2 \text{ mG}$ and $36 \pm 3 \text{ mG}$ trapped, respectively) were consistent within uncertainty. Averaging the results yields a sensitivity of $R_0 = aB_{RF}B_{trapped} + bB_{trapped}$ with $a = 0.020 \pm 0.003 \text{ nΩ/mT}_RF/\text{mG}_{trapped}$ and $b = 4.7 \pm 0.5 \text{ nΩ/mG}_{trapped}$.

The offset sensitivity b here is approximately two times larger than the sensitivity we reported at 1.3 GHz for cavities with similar ℓ ; in the previously reported model [8], a 1.3 GHz cavity with $\ell = 46 \text{ nm}$ would have sensitivity $b = 2.4 \text{ nΩ/mG}_{trapped}$. This indicates a linear scaling of this effect in 2/6-doped cavities with frequency $b \propto \omega$; other doping protocols may give different scaling. See also [10] for discussion of possible scaling mechanisms.

160 °C “N-INFUSION” AT 2.6 GHz

We also prepared a 2.6 GHz ILC-shape cavity with the “low-temperature dope” or “nitrogen-infusion” procedure, which consists of an 800 °C degas step followed by a ramp down to 160 °C and a 48-hour doping step with 40 mTorr (6 Pa) of continuously flowing N₂ gas. The cavity received no chemical treatment after doping. At 1.3 GHz, this treat-

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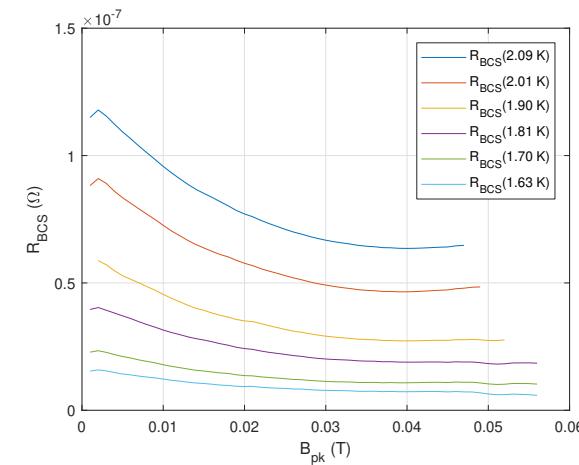


Figure 2: BCS surface resistance of the 2.6 GHz 160 °C-doped cavity.

ment has been shown to give results similar to the 800 °C N-dope [11, 12].

Under RF testing, this cavity performed with a strong anti-Q-slope at all temperatures below 2.17 K. Analysis yielded an electron mean free path ℓ of 2.8 nm and a critical temperature of 9.2 K. There was also a high R_0 , with a low-field minimum of 18 nΩ rising to 32 nΩ at $B_{pk} = 55$ mT, possibly due to a furnace contamination issue. Figure 2 shows the field-dependent BCS surface resistance for the cavity, which exhibits a drop of approximately 50% from low field to a minimum near 40 mT. This drop is similar in magnitude to our previous observations for 160 °C-doped 1.3 GHz cavities, both with N₂ gas [12, 13] and with a CO₂ gas mixture in an Ar carrier [5, 14], but more dramatic in slope; tests at 1.3 GHz showed the minimum in BCS resistance to be at 60 mT or higher field.

As with the 2/6-doped 2.6 GHz cavity [5], this slope is steeper than can be predicted by the Gurevich theory of the anti-Q-slope which has produced successful fits for many high- and low-T doped cavities at 1.3 GHz [3, 5, 15]. Moreover, the slope is steeper than that of the 2/6-doped 2.6 GHz cavity; qualitatively, this stronger slope in resistance for this cavity compared with the high-temperature-doped cavity with longer ℓ is consistent with observations at 1.3 GHz, serving as further evidence that cavities with shorter ℓ show stronger anti-Q-slope.

This test result also provides insight into the continuing discussion of the significance of impurity species in the low-T bakes [14]. Figure 3 shows the result of secondary ion mass spectrometry (SIMS) measurements of atomic concentration of impurities in a witness sample baked alongside the 2.6 GHz cavity during the 48-hour 160 °C bake. Similar to our previous report, there is very little N in the material, except for a low concentration $< 10^{20}$ at/cc in the first 10 nm; on the other hand, there is a significant amount of C and O through the RF penetration layer, possibly from impurities in the N₂ gas supply. As in our previous report, this indicates that the C and O impurities are responsible for the

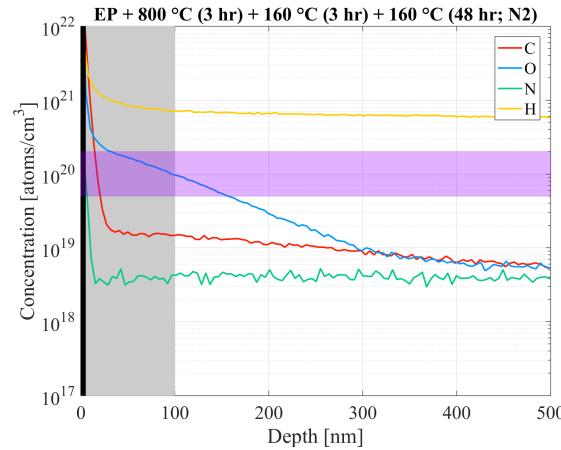


Figure 3: Results of SIMS analysis for a witness sample baked with the 1.3 GHz and 2.6 GHz cavities treated with the 48 hr 160 °C dope. The black line indicates the 5 nm oxide layer at the surface, the gray bar indicates the approximate RF penetration region of 100 nm, and the purple bar indicates the typical nitrogen concentration of a 2/6-doped cavity.

short ℓ of the cavity. Further, because the short ℓ correlates with stronger anti-Q-slope, this serves as more evidence for the suggestion that C and O impurities, rather than N, are responsible for the anti-Q-slope in the 160 °C-doped cavities.

160 °C “N-INFUSION” AT 1.3 GHz

In research efforts for the LCLS-II HE upgrade, we have tested several 1.3 GHz cavities with variations of the 160 °C doping treatment. Here we report on three recent tests: a 2-day doping (baked alongside the 2.6 GHz cavity discussed above), a 4-day doping, and the 4-day doping followed by approximately 5 nm surface removal by hydrofluoric acid (HF) rinse.

Figure 4 shows the BCS surface resistance of the 1.3 GHz cavity doped at 160 °C for 48 hours; Fig. 5 shows the same for the 96-hour dope. For both these cavities, unlike in the case of the 2.6 GHz cavity, RF test results did not show a strong anti-Q-slope. Instead, the BCS resistance showed a general field-dependent rise with a small region of decreasing resistance approximately 20 mT $< B_{pk} < 40$ mT. These cavities also showed high residual resistance, $R_0 \sim 15$ nΩ, which may indicate titanium contamination from the NbTi flanges of these cavities or other furnace contamination, as in the 2.6 GHz 160 °C-doped cavity above.

Fortunately, the third cavity test discussed in this study yielded more positive results, shown in Fig. 6 along with theoretical fits (discussed below). After a ~ 5 nm removal by HF rinse, the cavity performed with a strong anti-Q-slope, reaching a minimum resistance near $B_{pk} = 60$ mT. The low-field peak in BCS resistance was slightly lower, by $\sim 10\%$, and the residual resistance was lower than in the test before the HF rinse by a factor of 2.

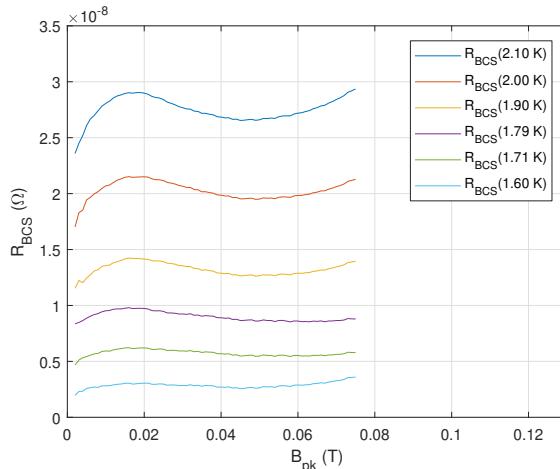


Figure 4: BCS surface resistance of the 1.3 GHz 48-hr 160 °C-doped cavity, with $\ell = 5.3$ nm.

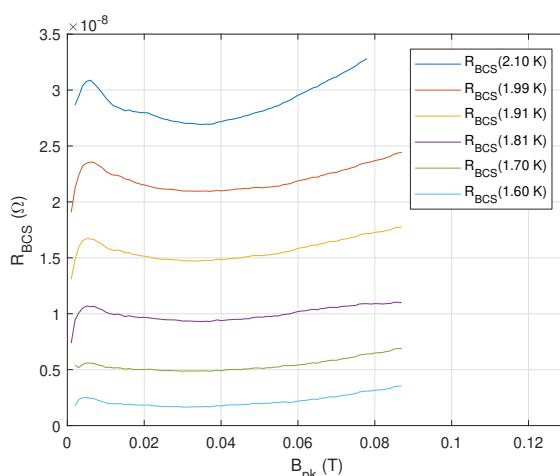


Figure 5: BCS surface resistance of the 1.3 GHz 96-hr 160 °C-doped cavity, with $\ell = 1.9$ nm.

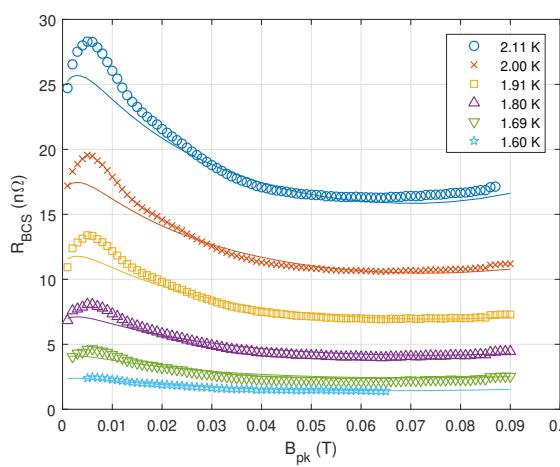


Figure 6: BCS surface resistance of the 1.3 GHz 96-hr 160 °C-doped cavity followed by ~ 5 nm HF rinse, as well as theoretical fit results, with $\ell = 1.7$ nm.

Using a recently improved version of our model [3] implementing the Gurevich anti-Q-slope theory [15], which now considers depth-dependent material properties, includes a model of heat transport away from the RF surface and into the liquid helium bath, and treats the quasiparticles as in thermal equilibrium through the RF layer, we have fitted these cavity test data. These fits are shown in Fig. 6. In general, the fit shows good agreement with experiment, except for a “bump” of higher resistance with peak at $B_{pk} \sim 5$ mT. This bump is also present in the test before the HF rinse.

Because of the thermal equilibrium in the updated model, the fit parameter is not directly comparable to the parameters from previous fits, but the very low level of quasiparticle overheating and strong anti-Q-slope are consistent with previous tests and fits of 160 °C-doped cavities and 800 – 1000 °C-doped cavities with mean free path < 10 nm [3, 5].

These initial results show promise for the 160 °C doping treatment. Though it appears that the treatment is susceptible to surface contamination from the furnace or otherwise, HF rinsing seems to alleviate the high residual resistance and restore the anti-Q-slope in cases of mild contamination. This bodes well for the treatment’s potential for future accelerators such as the LCLS-II HE upgrade, especially in terms of re-treatment of cavities: the gentle baking recipe and simple electrolyte-free chemistry can be performed on cavities without removing the welded helium jacket, greatly simplifying re-treatment procedures.

This still leaves the question of what specifically causes the difference in performance between the results in Figs. 5 and 6. The fact that the HF rinse removes only the first several nm suggests that it indeed is a light surface contamination, either present in the ~ 5 nm oxide layer or the first few nm of the Nb surface. For these two cavities, it is possible that the contamination comes from titanium from the NbTi flanges, as mentioned above. In this case, production accelerator cavities using the popular NbTi flanges with diamond-profile aluminum gaskets may suffer from similar contamination in the case of flanges which outgas Ti significantly. Thankfully, HF rinsing may offer a “cure” for such contamination, as indicated by our initial results here. Further study will be required to understand the source and nature of this occasional reduced performance for 160 °C-doped cavities.

CONCLUSIONS

We have studied flux trapping sensitivity in a 2.6 GHz 2/6-doped cavity, finding a sensitivity twice that of 1.3 GHz cavities with the same electron mean free path. We have studied the 160 °C doping at 2.6 GHz, finding a drop in resistance of comparable magnitude and steeper slope than that at 1.3 GHz. SIMS analysis indicates that C and O impurities caused the anti-Q-slope in this cavity. We also studied 160 °C doping at 1.3 GHz, finding poor performance from possible surface contamination, and finding that HF rinsing “cured” the poor performance of one cavity and restored a strong anti-Q-slope.

REFERENCES

- [1] A. Grassellino, A. Romanenko, D. Sergatskov, O. Melnychuk, Y. Trenikhina, A. Crawford, A. Rowe, M. Wong, T. Khabibouline, and F. Barkov. Nitrogen and argon doping of niobium for superconducting radio frequency cavities: a pathway to highly efficient accelerating structures. *Superconductor Science and Technology*, 26(10):102001, 2013.
- [2] P. Dhakal, G. Ciovati, G. R. Myneni, K. E. Gray, N. Groll, P. Maheshwari, D. M. McRae, R. Pike, T. Proslier, F. Stevie, R. P. Walsh, Q. Yang, and J. Zasadzinski. Effect of high temperature heat treatments on the quality factor of a large-grain superconducting radio-frequency niobium cavity. *Phys. Rev. ST Accel. Beams*, 16:042001, Apr 2013.
- [3] J. T. Maniscalco, D. Gonnella, and M. Liepe. The importance of the electron mean free path for superconducting radio-frequency cavities. *Journal of Applied Physics*, 121(4):043910, 2017.
- [4] M. Martinello, S. Aderhold, S.K. Chandrasekaran, M. Checchin, A. Grassellino, O. Melnychuk, S. Posen, A. Romanenko, and D.A. Sergatskov. Advancement in the understanding of the field and frequency dependent microwave surface resistance of niobium. In *Proceedings of SRF2017, Lanzhou, China*, pages 364–267, 2017.
- [5] J. T. Maniscalco, P. N. Koufalis, and M. Liepe. Modeling of the frequency and field dependence of the surface resistance of impurity-doped niobium. In *Proceedings of IPAC2018, Vancouver, Canada*, pages 2471–2474, 2018.
- [6] P. N Koufalis, M. Liepe, J. T. Maniscalco, and T. Oseroff. Experimental results on the field and frequency dependence of the surface resistance of niobium cavities. In *Proceedings of IPAC2018, Vancouver, Canada*, pages 2451–2454, 2018.
- [7] B. Aune, R. Bandelmann, D. Bloess, B. Bonin, A. Bosotti, M. Champion, C. Crawford, G. Deppe, B. Dwersteg, D. A. Edwards, H. T. Edwards, M. Ferrario, M. Fouaidy, P.-D. Gall, A. Gamp, A. Gössel, J. Gruber, D. Hubert, M. Hüning, M. Juillard, T. Junquera, H. Kaiser, G. Kreps, M. Kuchnir, R. Lange, M. Leenen, M. Liepe, L. Lilje, A. Mattheisen, W.-D. Möller, A. Mosnier, H. Padamsee, C. Pagani, M. Pekeler, H.-B. Peters, O. Peters, D. Proch, K. Rehlich, D. Reschke, H. Safa, T. Schilcher, P. Schmüser, J. Sekutowicz, S. Simrock, W. Singer, M. Tigner, D. Trines, K. Twarowski, G. Weichert, J. Weisend, J. Wojtkiewicz, S. Wolff, and K. Zapfe. Superconducting TESLA cavities. *Phys. Rev. ST Accel. Beams*, 3:092001, Sep 2000.
- [8] D. Gonnella, J. Kaufman, and M. Liepe. Impact of nitrogen doping of niobium superconducting cavities on the sensitivity of surface resistance to trapped magnetic flux. *Journal of Applied Physics*, 119(7), 2016.
- [9] D.L. Hall, M. Liepe, and R. Porter. Field-dependence of the sensitivity to trapped flux in Nb₃Sn. In *Proceedings of SRF2017, Lanzhou, China*, pages 844–847, 2017.
- [10] M. Checchin, M. Martinello, A. Grassellino, S. Aderhold, S. K. Chandrasekaran, O. S. Melnychuk, S. E. Posen, A. Romanenko, and D. A. Sergatskov. Frequency dependence of trapped flux sensitivity in SRF cavities. *Applied Physics Letters*, 2017.
- [11] A. Grassellino, A. Romanenko, Y. Trenikhina, M. Checchin, M. Martinello, O. S. Melnychuk, S. K. Chandrasekaran, D. A. Sergatskov, S. E. Posen, and A. C. Crawford. Unprecedented quality factors at accelerating gradients up to 45 MVm⁻¹ in niobium superconducting resonators via low temperature nitrogen infusion. *Superconductor Science and Technology*, 2017.
- [12] P. N. Koufalis, J. T. Maniscalco, and M. Liepe. Low temperature doping of niobium cavities: What is really going on? In *Proceedings of SRF2017, Lanzhou, China*, pages 353–358, 2017.
- [13] P.N. Koufalis, F. Furuta, J.J. Kaufman, and M. Liepe. Impact of the duration of low temperature doping on superconducting cavity performance. In *Proceedings of SRF2017, Lanzhou, China*, pages 750–753, 2017.
- [14] P.N. Koufalis and M. Liepe. Insights into the role of C, N, and O introduced by low temperature baking on niobium cavity performance. In *Proceedings of IPAC2018, Vancouver, Canada*, pages 2455–2457, 2018.
- [15] A. Gurevich. Reduction of dissipative nonlinear conductivity of superconductors by static and microwave magnetic fields. *Phys. Rev. Lett.*, 113:087001, Aug 2014.