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THE COLLABORATIVE EFFECTS OF INTRINSIC AND EXTRINSIC IMPURITIES IN LOW RRR SRF CAVITIES*

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Abstract

The SRF community has shown that introducing certain impurities into high-purity niobium can improve quality factors and accelerating gradients. We question why some impurities improve RF performance while others hinder it. The purpose of this study is to characterize the impurity profile of niobium coupons with a low residual resistance ratio (RRR) and correlate these impurities with the RF performance of low RRR cavities so that the mechanism of impurity-based improvements can be better understood and improved upon. The combination of RF testing and material analysis reveals a microscopic picture of why low RRR cavities experience low BCS resistance behavior more prominently than their high RRR counterparts. We performed surface treatments, low temperature baking and nitrogen-doping, on low RRR cavities to evaluate how the intentional addition of oxygen and nitrogen to the RF layer further improves performance through changes in the mean free path and impurity profile. The results of this study have the potential to unlock a new understanding on SRF materials and enable the next generation of high Q/high gradient surface treatments.

INTRODUCTION

As we approach the theoretical limit of niobium for superconducting radio-frequency (SRF) cavities, the last decade has brought immense improvements in quality factor (Q_0) and accelerating gradients though intentionally added impurities into the niobium surface [1,2]. Many SRF studies follow a "clean bulk dirty surface" technique to optimize the BCS resistance (R_{BCS}) by adding extrinsic impurities to the surface layer of high purity niobium [3–5]. Advancements have been made with nitrogen through N-doping, where cavities experience an anti-Q_0 slope and record breaking Q_0's at mid fields [6–8]. Oxygen added through a low temperature bake (LTB) has also provided high Q_0's and mitigation of the high field Q_0 slope typically seen in electropolished (EP) niobium cavities [9, 10]. The performance of these surface treatments is shown in Fig. 1.

The success of intentionally added impurities to the niobium surface has drawn deeper questions about how these impurities affect cavity behavior, and has prompted an investigation of cavities with a low residual resistance ratio (RRR). Low purity niobium has been studied in the past for the purpose of cost reduction and possible high Q_0 [11].

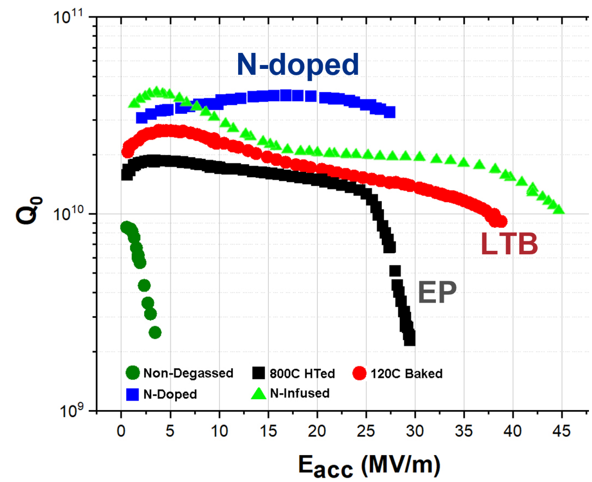

Figure 1: Comparison of quality factor versus gradient for surface treatments, adapted from Ref. [6].

In this study, we look to use the intrinsic impurities as a resource to optimize the R_{BCS} and understand the mechanism of impurity-based improvements. RRR and mean free path (mfp) have a direct relationship, so we might expect experience low R_{BCS} behavior at low RRR, as seen in Fig. 2. We ask if the intrinsic impurities can improve performance, as we observe in extrinsic impurities.

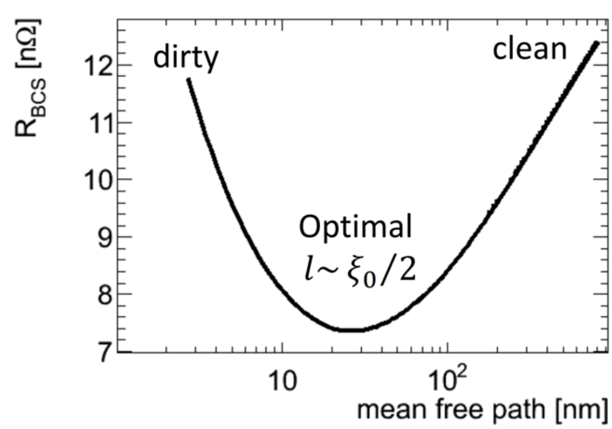

Figure 2: BCS resistance versus mean free path shows an optimization in BCS resistance for moderately dirty surface, adapted from Ref. [12].

In this study, we investigate a single-cell TESLA-shaped 1.3 GHz cavity with RRR 61. First, the cavity receives EP treatment to make the surface layer and bulk uniform [13]. We measure Q_0 versus gradient at 2 K and low temperature (< 1.5 K) in the vertical test stand [2]. The surface resis-

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tance is the geometry factor of the cavity divided by the Q_0; this can be broken down into the residual resistance (R_{res}) and R_{BCS}. We compare the performance to its high RRR counterpart in EP condition to understand how the intrinsic impurities affect the bulk and surface behavior of the cavity. We perform a LTB at 120 °C for 48 hours and repeat the testing to evaluate how the addition of the surface oxide to the RF layer further affects performance. We additionally investigate the effect of adding nitrogen to the dirty bulk by performing N-doping with the standard 2/6 + 5 µm recipe [14]. Additionally, we performed secondary ion mass spectrometry (SIMS) on low and high RRR witness samples in EP, LTB, and N-doped conditions to characterize their impurity profiles. By correlating the concentration of impurities with the RF performance, we can gain a deeper understanding of the role of both intrinsic and extrinsic impurities.

RESULTS

Quality Factor

We measure the Q_0 at a given gradient by maintaining the cavity at its resonant frequency, inputting power via antenna, and then measuring the reflected and transmitted power [15]. The Q_0 is the ratio of the energy gain per RF period and dissipated power. The measurements of Q_0 at 2 K are graphed in Fig. 3. In general, the Q_0's of the low RRR tests are lower than their high RRR counterparts.

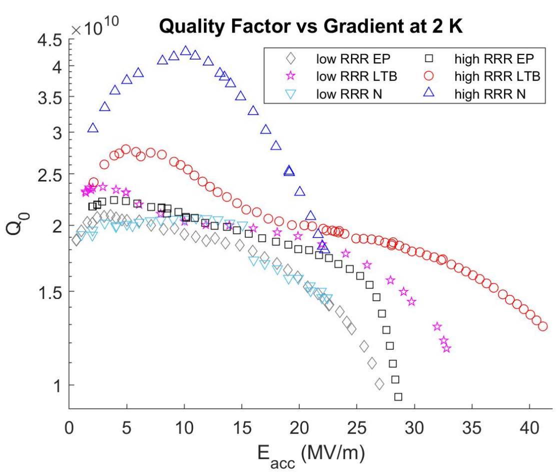

Figure 3: Quality factor at 2 K versus accelerating gradient for EP, LTB, and N-doping on low and high RRR.

The low RRR cavity after the LTB shows a slight increase in Q_0 at low gradients, as well as improved performance through higher gradients, compared to the EP test. The performance after EP and LTB treatments for high and low RRR is similar at mid gradients. Oxygen improves performance of low RRR cavity but with a weaker response than we see in high RRR cavities, as the LTB treatment delays Q_0 slope in low RRR with a less extreme difference than for high RRR. The low RRR cavity did not show a strong high field Q_0 slope in EP condition, so the transition to LTB was not as drastic. In the LTB test, the low RRR cavity does not experience the significant anti-Q_0 slope at low gradient seen on the high RRR. The weakened Q_0 slope suggests that the intrinsic impurities may capture the free H which is thought to exacerbate the high field Q_0 slope [16, 17]. We are also unable to reach as high gradient in the low RRR test in both EP and LTB, which is likely due to its higher concentration of intrinsic impurities which causes a lower thermal conductivity.

The performance after N-doping is quite similar to EP at low and high gradients. The cavity experienced multipacting quenches above 16 MV/m, which trapped magnetic flux and worsened the performance up to its ultimate quench at 22 MV/m. The quality factor of the N-doped low RRR cavity is significantly lower than that of the high RRR, but they reach similar maximum gradients. We observe a slight anti-Q_0 slope on the low RRR, but much less extreme than the high RRR. N-doping the cavity with the 2/6 + 5 µm recipe did not improve the Q_0 of the low RRR cavity, unlike high RRR N-doped cavities.

Residual Resistance

The residual resistance (R_{res}) taken at low temperature is temperature-independent, and comes from impurities in the superconducting lattice as well as any trapped flux from cooldown or quench. The R_{res} measurements are shown in Fig. 4. We observe a significant offset in R_{res} between low and high RRR for all surface treatments, especially at mid gradient. This may suggest that the oxide structure of the low RRR cavity is different or that the intrinsic impurities may drive additional losses.

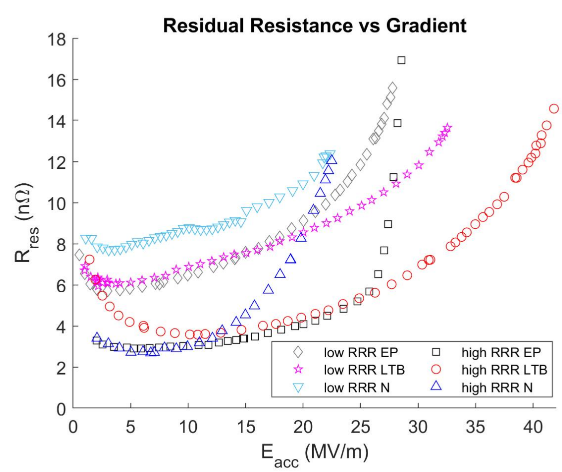

Figure 4: Residual resistance (at low T) versus accelerating gradient for low and high RRR.

The low RRR EP and LTB curves are nearly equal at low and mid gradients until around 20 MV/m, after which oxygen enables lower R_{res}. It is reassuring that the addition of oxygen to the RF layer did not further increase the resistive effect of the intrinsic impurities in the material. This split is analogous to that of the high RRR EP and LTB which occurs around 25 MV/m at a lower resistance. The offset of the low and high RRR LTB curves clarifies the effect of a uniform distribution of impurities in the bulk.

The low RRR N-doped curve is slightly higher than the corresponding EP and LTB curves. It is also larger than the high RRR N-doped curve, except at high gradient. Because N-doping introduces impurities further into the bulk than LTB, it is possible this caused the increase in R_{res}. Another possible cause is the flux trapped through the multipacting quenches during the 2 K test.

BCS Resistance

The R_{BCS} is calculated by taking the difference between the total surface resistance at 2 K and low T. This temperature-dependent component of the resistance is caused by the breakdown of cooper pairs with increasing temperature [3, 14]. In Fig. 5, we highlight the low R_{BCS} behavior of the low RRR cavity.

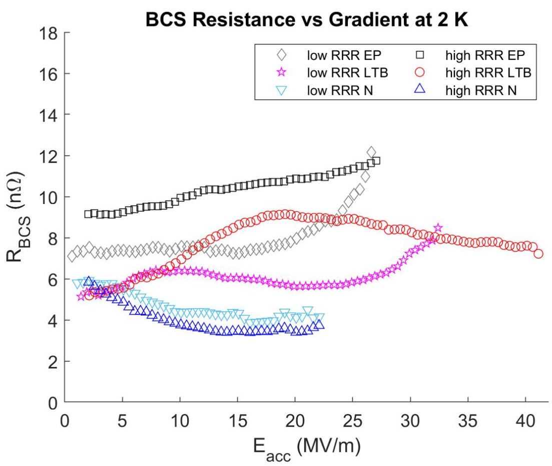

Figure 5: BCS resistance versus accelerating gradient at 2 K for low and high RRR.

At all points of the EP and LTB tests, the low RRR R_{BCS} is equal to or below that of its high RRR counterpart. The benefit of the low RRR with these treatments is most prominent at mid gradients and is completely lost at high gradients. The LTB high and low RRR are equal until 10 MV/m, but then show a similar behavior of the local maximum and then decrease. It is promising that the LTB lowered the R_{BCS} at all gradients from the EP test, so making the surface even dirtier allowed for lower BCS resistance even with a less clean bulk. It is not clear yet if we have reached the optimized surface dirtiness or if we could go even further.

The N-doped test of the low RRR cavity showed similar R_{BCS} than that of the high RRR, but significantly reduced from the EP and LTB tests.The decrease of R_{BCS} with field emphasizes the anti-Q_0 slope of the N-doped low RRR, which is difficult to discern from the Q_0 curve alone. N-doping showed additional improvement of the R_{BCS} from the EP and LTB tests, but it is surprising that the low RRR is larger than its high RRR counterpart. A possible explanation is that the 2/6 + 5 µm recipe produces an "overdoped" effect on a cavity with more intrinsic impurities, since this recipe was optimized for a high RRR material.

Impurity Profiles

The SIMS data is measured as the intensity of each ion versus sputtering time. The impurity profiles shown in Figs. 6–9 are the most relevant ions found showing the differences between the surface treatments in low and high RRR. The x axes are normalized by the noise floor of the Nb_2O_5 signal at 10 counts of intensity corresponding to 5nm depth into the samples [18]. The y axes are normalized by the niobium signal point-to-point for each coupon. We found no obvious impurities which explain the dramatically lower RRR, so we consider that other factors, such as grain size, may govern the RRR. This will be the subject of further study.

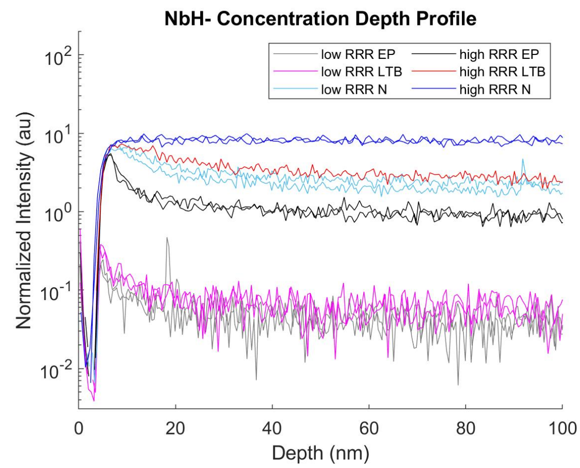

Figure 6: Impurity profile of NbH- on variously treated Nb samples.

In Fig. 6, we observe that the low RRR samples have less NbH-, which suggests that some impurity is trapping the free hydrogen. This aligns with the weakened Q_0 slope seen in the low RRR cavity. The low RRR EP and LTB are nearly identical, whereas there is a larger gap for their high RRR counterparts. This could be from the low RRR EP surface being dirtier to begin with than the high RRR EP. We also see that N-doping increases NbH- in both high and low RRR. Further studies needed to understand this heightened NbH- signal from introducing nitrogen into the RF layer.

In Fig. 7, we observe that the low RRR EP and LTB samples have more C- than their high RRR counterparts. However, the N-doped samples do not follow this trend, where the low and high RRR samples appear to have traded places. This is surprising, as the low RRR samples were all cut from the same sheet metal and N-doping treatment is unlikely to remove carbon from the surface. However, the high RRR samples were not necessarily from the same sheet metal, so some variation is to be expected. While carbon may be a noticeable impurity in the low RRR material, the C- signal alone cannot explain the drastic difference in RRR. Further studies of carbon-containing ions may help provide a better explanation.

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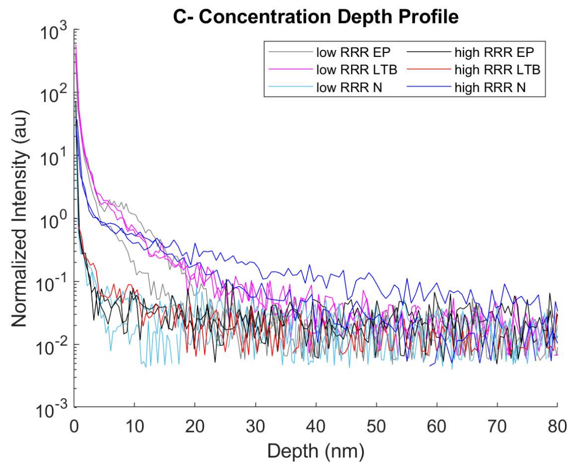

Figure 7: Impurity profile of C- on variously treated Nb samples.

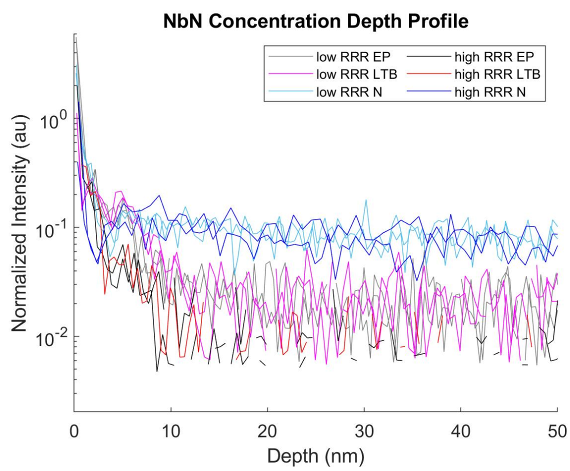

Figure 8: Impurity profile of NbN on variously treated Nb samples.

In Fig. 8, we note nitrogen diffuses similarly for low and high RRR. This aligns with their similar R_{BCS}. It appears that the nitrogen introduced via doping and the intrinsic impurities do not interact, as we observed no combined effect of these impurities like we had for oxygen. We also observe some nitrogen in bulk of low RRR EP and LTB which does not occur in the corresponding high RRR samples. This shows nitrogen could be one of the intrinsic impurities in the low RRR material, but not so much that it would account for the difference in RRR.

In Fig. 9, we observe oxygen diffuses similarly during LTB for high and low RRR. The O- profiles do not explain the difference in the LTB tests, suggesting another impurity is responsible for the different R_{BCS}. We also note that the oxygen profile is quite similar for high and low RRR EP samples, so oxygen is not an intrinsic impurity in the low RRR material.

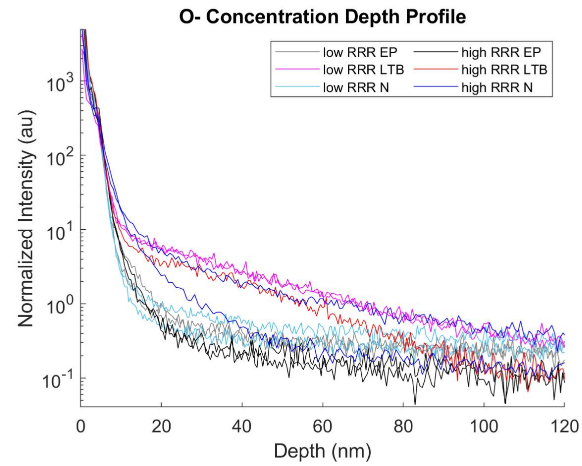

Figure 9: Impurity profile of O- on variously treated Nb samples.

CONCLUSION

The low RRR cavity behaves quite differently than high RRR cavities, with lower R_{BCS}, larger R_{res}, lower Q_0, and lower gradients in general. The intrinsic impurities affect the performance of the cavity for all surface treatments examined. Making the surface even dirtier allowed for lower R_{BCS}, even with a less clean bulk.

This difference is most notable in the EP testing, as the intrinsic impurities protect the cavity from a high field Q_0 slope and significantly improve the R_{BCS}. There is more similarity in the performance of the LTB cavities in terms of the offset of the R_{res}, the shape of the R_{BCS} curves, and the oxygen diffusion profiles. It is an important result that oxygen and the intrinsic impurities appear to collaborate, enabling higher Q_0 and gradients. It appears that the LTB brought the low RRR cavity closer to the optimization of the R_{BCS}. The N-doping test showed increased R_{res} from the other low RRR tests, but also showed a further decrease in the R_{BCS}. The similar diffusion of nitrogen, along with the similar R_{BCS} shows that N-doping is a robust treatment in different purity SRF cavities, as well as suggesting that nitrogen and the intrinsic impurities have minimal interactions. By understanding how oxygen and nitrogen interact with the intrinsic impurities, we can gain insight how to develop a future high Q_0/high gradient surface treatment involving these impurities.

REFERENCES

[1] S. Belomestnykh *et al.*, “Key directions for research and development of superconducting radio frequency cavities”, in *Proc. Snowmass 2021*, 2022. doi:10.48550/arXiv.2204.01178

[2] M. Checchin, “Physics of Limiting Phenomena in Superconducting Microwave Resonators: Vortex Dissipation, Ultimate Quench and Quality Factor Degradation Mechanisms”, Ph.D. thesis, Phys. Dept., Illinois Institute of Technology, 2016.

ISBN: 978-3-95450-234-9 ISSN: 2673-5504 doi:10.18429/JACoW-SRF2023-MOPMB032

[3] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, "Theory of Superconductivity", *Phys. Rev.* vol. 108, p. 1175, 1957. doi:10.1103/PhysRev.108.1175

[4] T. Kubo, "Effects of Nonmagnetic Impurities and Subgap States on the Kinetic Inductance, Complex Conductivity, Quality Factor, and Depairing Current Density", *Phys. Rev. Applied*, vol. 17, p. 014018, 2022. doi:10.1103/PhysRevApplied.17.014018

[5] V. Ngampruetikorn and J. A. Sauls, "Effect of inhomogeneous surface disorder on the superheating field of superconducting RF cavities", *Phys. Rev. Res.*, vol. 1, p. 012015, 2019. doi:10.1103/PhysRevResearch.1.012015

[6] A Grassellino *et al.*, "Nitrogen and argon doping of niobium for superconducting radio frequency cavities: a pathway to highly efficient accelerating structures", *Supercond. Sci. Technol.* vol. 26, p. 102001, 2013. doi:10.1088/0953-2048/26/10/102001

[7] D. Gonnella *et al.*, "Industrialization of the nitrogen-doping preparation for SRF cavities for LCLS-II", *Nucl. Instrum. Methods Phys. Res., Sect. A*, vol. 882, pp. 143–150, 2018. doi:10.1016/j.nima.2017.11.047

[8] P. Dhakal, "Nitrogen doping and infusion in SRF cavities: A review", *Phys. Open*, vol. 5, p. 100034, 2020. doi:10.1016/j.physo.2020.100034

[9] D. Bafia, A. Grassellino, O. Melnychuk, A. Romanenko, Z. H. Sung, and J. Zasadzinski, "Gradients of 50 MV/m in TESLA Shaped Cavities via Modified Low Temperature Bake", in *Proc. SRF'19*, Dresden, Germany, Jun.-Jul. 2019, pp. 488–493. doi:10.18429/JACoW-SRF2019-TUP061

[10] D. Bafia, A. Grassellino, and A. S. Romanenko, "The Role of Oxygen Concentration in Enabling High Gradients in Niobium SRF Cavities", in *Proc. SRF'21*, East Lansing, MI, USA, Jun.-Jul. 2021, pp. 871–875. doi:10.18429/JACoW-SRF2021-THPTEV016

[11] G. Ciovati, P. Dhakal, and G. R Myneni, "Superconducting radio-frequency cavities made from medium and low-purity niobium ingots", *Supercond. Sci. Technol.*, vol. 29, p. 064002, 2016. doi:10.1088/0953-2048/29/6/064002

[12] A. Miyazaki, "Basics of RF superconductivity and Nb material", SRF2021 Tutorial, East Lansing, Michigan, Jun. 23-25, 2021. https://indico.frib.msu.edu/event/38/attachments/159/1139/SRF2021_lecture_miyazaki_ver4.pdf

[13] F. Furuta, D. J. Bice, A. C. Crawford, and T. J. Ring, "Fermilab EP Facility Improvement", in *Proc. SRF'19*, Dresden, Germany, Jun.-Jul. 2019, pp. 453–455. doi:10.18429/JACoW-SRF2019-TUP022

[14] D. Bafia, "Exploring and Understanding the Limitations of Nb SRF Cavity Performance", Ph.D. thesis, Phys. Dept., Illinois Institute of Technology, 2020.

[15] H. Padamsee, J. Knobloch, and T. Hays, *RF Superconductivity for Accelerators*, Second Edition, Wiley-VCH Verlag GmbH and Co., KGaA, Weinheim, 2008.

[16] A. Romanenko, "Surface Characterization of Nb Cavity Sections - Understanding the High Field Q-Slope", Ph.D. thesis, Phys. Dept., Cornell University, 2009.

[17] F. Barkov, A. Romanenko, Y. Trenikhina, and A. Grassellino, *J. Appl. Phys.*, vol. 114, p. 164904, 2013. doi:10.1063/1.4826901

[18] Y. Trenikhina, A. Romanenko, J. Kwon, J.-M. Zuo, and J. F. Zasadzinski. "Nanostructural features degrading the performance of superconducting radio frequency niobium cavities revealed by transmission electron microscopy and electron energy loss spectroscopy", *J. Appl. Phys.*, vol. 117, p. 154507, 2015. doi:10.1063/1.4918272