

Healing Gradient Degradation in Nb₃Sn SRF Cavities Using a Recoating Method

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Despite having advantageous superconducting properties, Nb₃Sn superconducting radiofrequency (SRF) cavities still have practical challenges compared to Nb SRF cavities due to the brittle nature of Nb₃Sn. Performance degradation can occur when a Nb₃Sn SRF cavity experiences mechanical stresses such as during handling and tuning of the cavity. In this study, we present a potential treatment for SRF cavities that have experienced stress induced performance degradation that involves a recoating procedure. The degraded cavity is coated with a small amount of Sn using a single step vapor-diffusion methodology. Using this approach we can recover a significant portion of the lost performance of a Nb₃Sn SRF cavity.

I. INTRODUCTION

Niobium cavities have been extensively studied and treatments have been developed to optimize the accelerating gradient and quality factor [1–4]. The performance of niobium SRF cavities is limited by the material properties of niobium (Nb). A promising alternative to Nb is Nb₃Sn. There exists a large body of research on creating high performance Nb₃Sn superconducting radiofrequency (SRF) cavities[5, 6]. Desirable superconducting properties, such as higher superconducting transition temperature (T_c) and a higher superheating magnetic field (H_{sh})[7–10], make Nb₃Sn an attractive material for SRF applications. The material properties of Nb₃Sn, however, make it difficult to work with.

The brittleness of Nb₃Sn introduces new challenges to the cavity manufacturing process. Nb₃Sn must be deposited as a thin film on a bulk cavity substrate[11–13]. Because of the thin and brittle film, Nb₃Sn cavities are highly susceptible to mechanical stress. Nb₃Sn cavity performance is known to permanently degrade when stresses are applied to the cavity[14, 15]. This degradation is assumed to be caused by cracks in the brittle Nb₃Sn film caused by deformation of a cavity during processing such as tuning or assembly. Cavities that suffer from degradation are typically stripped and recoated with a new Nb₃Sn film, which is a time-consuming and expensive process.

In this current study we explore a new procedure to heal Nb₃Sn cavities whose performance has been degraded by deformation. This procedure utilizes a short Nb₃Sn recoating to attempt to heal cracks that have formed in the cavity without the need to remove the original film. This procedure was developed to restore the performance of a Nb₃Sn cavity which has undergone centrifugal barrel polishing[16]. The performance decrease measured on a polished cavity is like the above mentioned case of deformation-induced degradation. When employing this recoating procedure to a degraded cavity, we can recover a large portion of the performance with a simple furnace treatment. This discovery provides a valuable method for recovering degraded cavities without lengthy reprocessing which avoid subsequent thinning and frequency shifts.

II. EXPERIMENT

This study is performed on a Nb₃Sn, 1.3 GHz cavity coated using a high-temperature nucleation step to create a Nb₃Sn film with low surface roughness. An in-depth analysis of this cavity coating and the initial performance of the cavity can be found in reference [17].

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After initial testing, the cavity was transported to Cornell, after which performance decreased. The cavity was then returned back to FNAL for additional testing, which confirmed the performance degradation. We suggest that the degradation was caused by stresses applied to the cavity during transport, which led to the formation of cracks. This type of performance degradation has previously been observed during assembly of Nb₃Sn cavities [14], and when tuning Nb₃Sn cavities at room temperature [15]. In these cases, stresses applied to the cavity were suggested to be the main cause of the degradation. Cracks may also form in Nb₃Sn cavities as a result of stress concentrators such as foreign particles or impurities located in the Nb-Nb₃Sn interface. Another possible source of performance degradation is elastic deformation of the Nb₃Sn film caused by thermal contraction of the cavity during cooldown. This is, however, unlikely in this case since the thermal expansion coefficient of Nb and Nb₃Sn does not differ enough to cause degradation, and Nb₃Sn cavities have shown no signs of degradation due to thermal cycling.

To heal the cracks causing the performance degradation, we apply a recoating procedure. During this recoating procedure, the cavity was heated to 1000 °C and exposed to Sn vapor for 1 h. Sn vapor was provided by 0.85 g of Sn heated to 1250 °C. The reasoning behind these parameters is that only a small amount of Sn is necessary to fill the microscopic cracks in the film. Applying too much Sn causes the film to become too thick and negatively impacts the surface roughness of the film. During the coating process only a small fraction of the Sn evaporated leaving behind a large amount of the initial Sn still in the crucible.

The cavity performance was tested using a vertical testing stand (VTS). The cavity was tested before degradation, after degradation, and after recoating at 4 K and at 2 K. The cavity was cooled down below its superconducting transition temperature at 16 K at a slow cooling rate of 0.1 K min⁻¹ to minimize trapped flux. Between each test the cavity was brought up to a temperature above its superconducting transition temperature and cooled back down using the slow cooling rate to eliminate any trapped flux caused by cavity quenching. Temperature mapping was performed during the VTS test at 2 K.

III. RESULTS

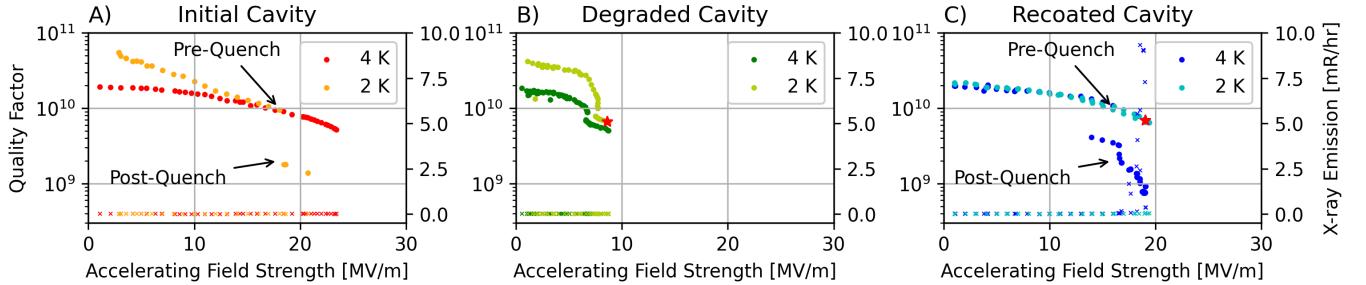


FIG. 1. The quality factor, indicated by dots, and X-ray emissions, indicated by x, versus the accelerating gradient of the cavity after the initial coating (A), after the degradation (B), and after the recoating (C). The quality factor and accelerating gradient of the T-maps in figure 2 are indicated by a red star.

After initially coating the cavity it achieves a peak accelerating field of 24 MV m⁻¹ and a maximum Q of 2×10^{10} at 4 K. The peak accelerating gradient after degradation is 8 MV m⁻¹ and a maximum Q of 2×10^{10} at 4 K. The cavity displays a decrease in the quality factor at around 6 MV m⁻¹ before a quench. A similar decrease in the quality factor is observed for other cavities affected by performance degradation[14, 15] and in Nb₃Sn cavities treated with centrifugal barrel polishing[16]. Temperature mapping is displayed in figure 2 and is utilized to locate the quench source responsible for the performance degradation. A single hot spot on the equator of the cavity is present. Visual inspection of the cavity did not display visible defects near the quench location.

After the recoating procedure is applied the cavity's performance increases. The cavity experiences an initial quench at 16 MV m⁻¹ resulting in trapped flux which decreases the quality factor. The cavity is still able to reach a peak accelerating gradient of 19 MV m⁻¹. At 2 K the cavity does not quench until reaching the maximum gradient of 19 MV m⁻¹. The quality factor after recoating is 2×10^{10} at 4 K and the Q slope seen in the degraded cavity is not present. Temperature mapping of the cavity after recoating demonstrates that the initial hot spot is healed with no detectable heating from that area. This indicates that the defect causing the performance degradation is repaired by recoating. At higher gradients another small hotspot appears in a new location close to the equator, and there is also a larger hot spot closer to the iris, which appears just before the cavity quench. We also see a spike in the x-ray emissions from the cavity just before the final quench at 4 K. Although we are not certain what the cause of

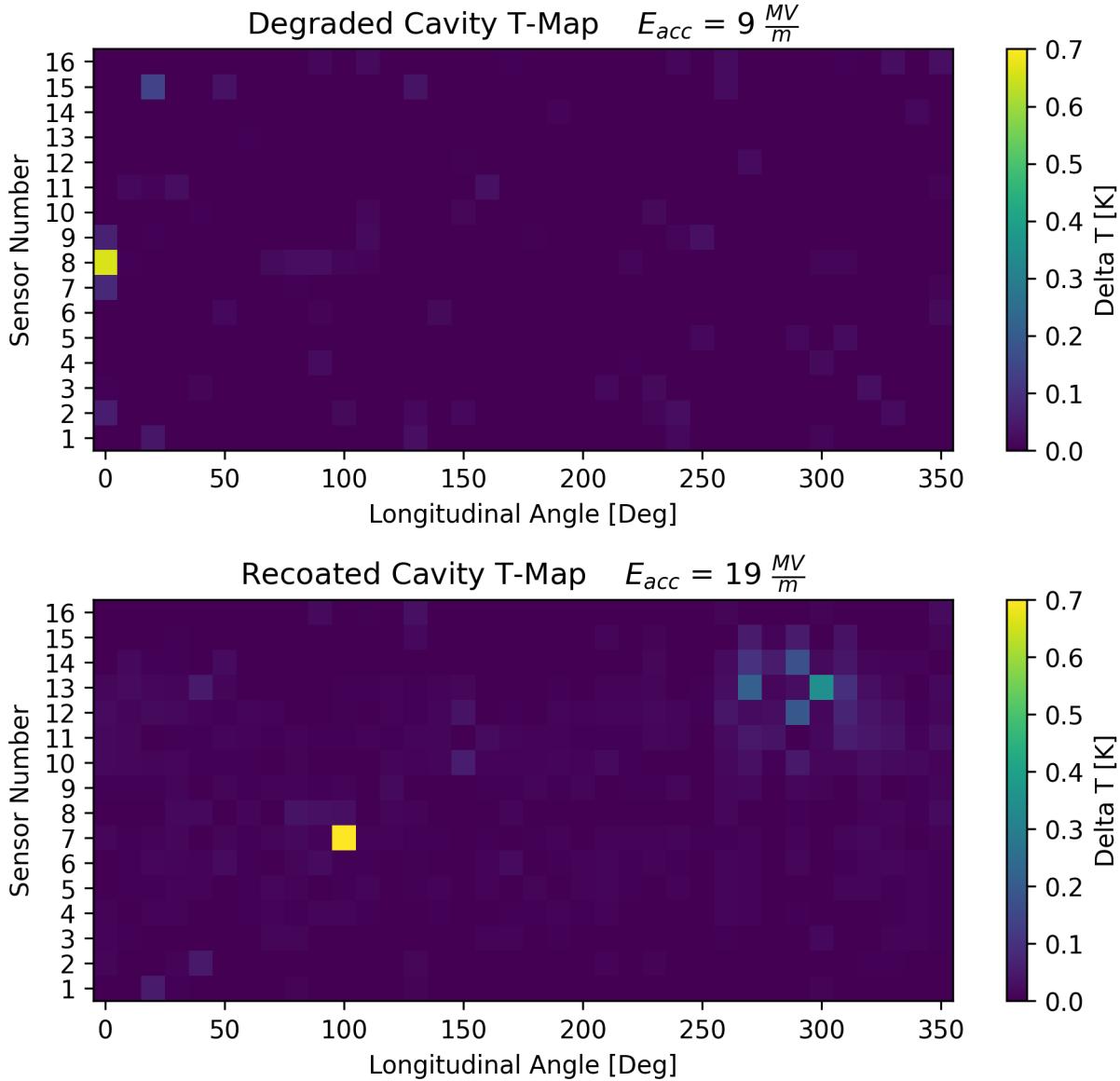


FIG. 2. Temperature maps of a cavity's surface prior to quench as measured before (top) and after (bottom) the recoating is applied. The temperature maps are measured at 2 K. The sensor number corresponds to different regions of the cavity. Sensor 1 and 16 are near the top and bottom iris while sensor 8 is on the equator. The quality factor and accelerating gradient of the cavity during the measurement is shown by the red star in figure 1. The temperature of the hot spot near the equator of the recoated cavity exceeds the maximum value of the color bar and achieves a maximum value at 3 K

this spike is, it does not seem to be caused by the cracks nor the recoating. This particular cavity has shown x-ray emissions in the past even before the degradation and recoating occurred.

IV. EVIDENCE OF CRACK HEALING MECHANISM IN Nb_3Sn

To study the healing mechanism of the recoating procedure, we purposefully introduce cracks into Nb_3Sn coated Nb wires. 3 mm diameter low RRR Nb wires were coated with Nb_3Sn using Sn vapor diffusion. Before the coating, the Nb wires are electropolished and anodized in the same way as Nb_3Sn cavities. Cracks are created by elongating the wires using an Instron tensile testing machine. We then cut the wires into two pieces and treat one half of the wires with the same recoating recipe shown in section II. The cracks are analyzed before and after recoating using

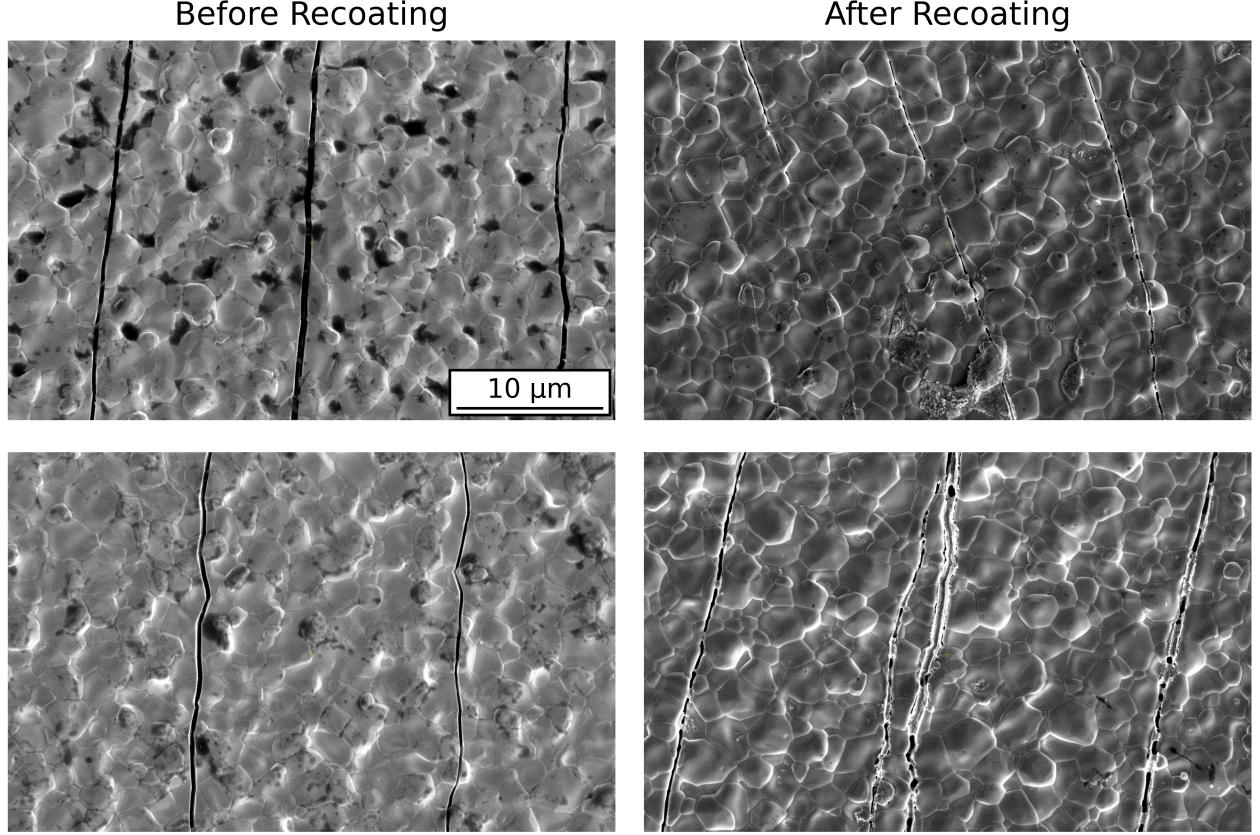


FIG. 3. This figure shows micrographs of elongated Nb₃Sn coated wires. The left side shows the samples before treatment with recoating and the right side shows the samples after recoating. After the recoating treatment the cracks appear to be partially filled with new Nb₃Sn.

scanning electron microscopy (SEM) and energy dispersive x-ray spectroscopy (EDS).

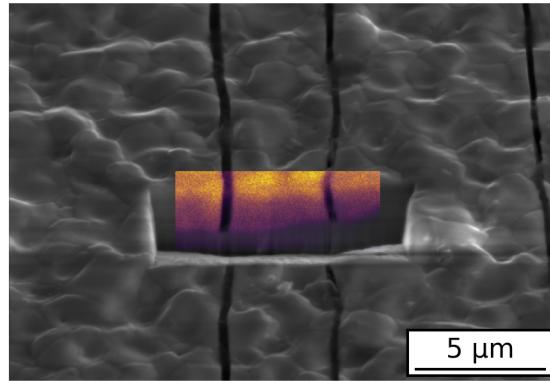
The elongation of the wires causes intragranular cracks to form perpendicular to the direction of applied stress as shown in figure 3. A cross section of the sample, as seen in figure 4, show that the cracks penetrate all the way through the Nb₃Sn film and stop at the Nb substrate giving the crack a rectangular profile with sharp 90° edges. These cracks can be considered an extreme example of the cracks we expect to see in Nb₃Sn cavities since the wires are heavily deformed in the plastic regime. In contrast, cracks formed in Nb₃Sn cavities are formed during elastic deformation of the niobium substrate or only slight plastic deformation which does not significantly impact the geometry of the cavity. Therefore, we expect cracks in degraded cavities to be smaller than what is seen in this study.

After the recoating, the cracks appear to be partially healed. Figure 3 shows some of the cracks being partially filled in with new material creating a discontinuous crack. Additionally, the sharp edges of the untreated cracks have been smoothed out due to deposition of new material. The cross section of the sample in figure 4 shows that new Nb₃Sn is created both within the crack and at the exposed Nb substrate at the base of the crack. Since the cracks produced by elongating the wires are larger than what we expect in Nb₃Sn cavities, it is likely that the cracks formed in cavities can be completely healed using this recoating recipe. It may also be possible to increase the recoating duration or temperature to allow larger cracks to heal completely as well.

DISCUSSION

Recoating a damaged Nb₃Sn cavity can have a major impact on its performance. The recoating process was able to mostly recover both the maximum accelerating gradient and quality factor of the cavity. The degradation in quality factor seen at 2K at high fields seen in figure 1, C is caused by a quench at 16 MV m⁻¹ resulting in trapped flux. However, the cavity was able to reach a higher gradient of 19 MV m⁻¹ after the initial quench. This is close to the

0.5% Elongation Before Recoating



0.5% Elongation After Recoating

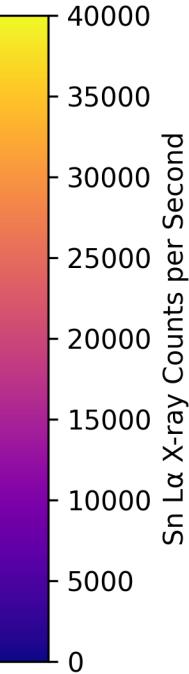
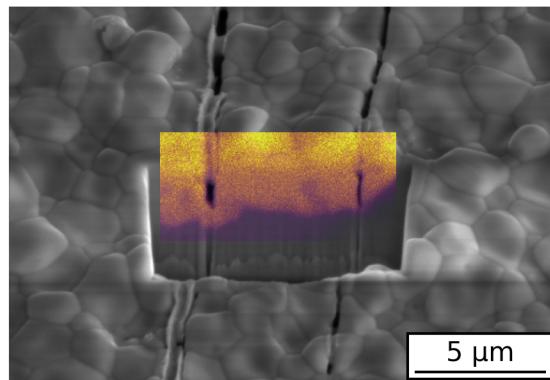


FIG. 4. The cross section of the film cracks is imaged using FIB/SEM before and after recoating. The Sn content of the cracks is measured qualitatively using Energy dispersive X-ray Spectroscopy (EDS). We find that there is new Nb₃Sn material both in the crack and in the substrate at the base of the crack. There is no evidence of Sn rich phases in the crack.

24 MV m⁻¹ maximum field of the cavity before the degradation occurred. It is possible that the cavity may recover more of its performance by using a longer recoating or other surface treatments such as mechanical polishing[16].

The healing mechanism of the recoating is heretofore unknown. From our observations it appears that the healing occurs via two different processes, the creation of new Nb₃Sn within the crack and the creation of Nb₃Sn at the exposed Nb substrate at the base of the crack. This phenomenon has been studied in other thin film system and is known as self healing [18]. Here we propose two mechanisms to explain the self healing observed in our experiments.

The first mechanism for self healing is creation of newly formed Nb₃Sn within the crack. When a cavity is exposed to Sn, a thin layer of liquid Sn coats the surface which thereby fills the cracks. Nb₃Sn is then formed in the cracks by diffusion of Nb from the old Nb₃Sn into the liquid Sn creating new Nb₃Sn. The diffusion rate of Nb is relatively slow compared to the diffusion of Sn through the grain boundaries during normal film growth, however if the cracks are small, less than a few 100 nm, Nb may have sufficient time to diffuse into the crack. The diffusion of Nb into the crack may be aided by dissolution of Nb₃Sn into the liquid Sn layer above 910 °C which would greatly boost the diffusion rate. Since Nb is diffusing from the Nb₃Sn film into the crack, there is the possibility of creating non-superconducting Sn rich Nb-Sn phases such as Nb₆Sn₅ and NbSn₂. However, we see no evidence of these phases on the length scales measurable using EDS. Further analysis is required using higher resolution techniques such as transmission electron microscopy (TEM-EDS) to verify the stoichiometry of the newly created Nb₃Sn material.

The second mechanism involves the diffusion of Sn into the Nb substrate through the crack. Since the crack penetrates the Nb₃Sn film, the liquid Sn layer can come into contact with the crack and react with the Nb substrate creating a region of new Nb₃Sn. This new region acts as a bridge for electrical currents to flow through the film and prevents current from flowing through the Nb substrate, which has a higher resistivity than does Nb₃Sn.

V. CONCLUSION

Using a low temperature (1000°C), short duration (1 h) Sn recoating process, we are able to heal a degraded Nb_3Sn cavity that suffered damage during transportation. The recoating process improved the maximum gradient of the cavity from 8 MV m^{-1} to 19 MV m^{-1} , which is close to the initial performance of the cavity of 24 MV m^{-1} . Temperature mapping measurements of the cavity demonstrate that a single hot spot on the equator of the cavity was responsible for the performance degradation. After the recoating process this defect becomes healed leading to less heating and a higher maximum electric field gradient. Ultimately, the performance is limited by a second hot spot.

This discovery provides a new approach which applies to similarly degraded SRF cavities to recover their performances. This approach saves time and money which would otherwise be spent removing the Nb_3Sn coating and then applying a new coating. This self healing process makes Nb_3Sn cavities more viable for real-world accelerator applications by reducing their manufacturing costs.

VI. ACKNOWLEDGEMENTS

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- [1] A. Romanenko, A. Grassellino, F. Barkov, A. Suter, Z. Salman, and T. Prokscha, “Strong Meissner screening change in superconducting radio frequency cavities due to mild baking,” *Applied Physics Letters*, vol. 104, p. 072601, 02 2014.
- [2] M. Martinello, A. Grassellino, M. Checchin, A. Romanenko, O. Melnychuk, D. A. Sergatskov, S. Posen, and J. F. Zasadzinski, “Effect of interstitial impurities on the field dependent microwave surface resistance of niobium,” *Applied Physics Letters*, vol. 109, p. 062601, 08 2016.
- [3] E. M. Lechner, J. W. Angle, F. A. Stevie, M. J. Kelley, C. E. Reece, and A. D. Palczewski, “RF surface resistance tuning of superconducting niobium via thermal diffusion of native oxide,” *Applied Physics Letters*, vol. 119, p. 082601, 08 2021.
- [4] A. Dangwal Pandey, T. F. Keller, M. Wenskat, A. Jeromin, S. Kulkarni, H. Noei, V. Vonk, W. Hillert, D. Reschke, N. Walker, H. Weise, and A. Stierle, “Grain boundary segregation and carbide precipitation in heat treated niobium superconducting radio frequency cavities,” *Applied Physics Letters*, vol. 119, p. 194102, 11 2021.
- [5] C. Becker, S. Posen, N. Groll, R. Cook, C. M. Schlepütz, D. L. Hall, M. Liepe, M. Pellin, J. Zasadzinski, and T. Proslier, “Analysis of Nb_3Sn surface layers for superconducting radio frequency cavity applications,” *Applied Physics Letters*, vol. 106, p. 082602, 02 2015.
- [6] S. Posen, M. Liepe, and D. L. Hall, “Proof-of-principle demonstration of Nb_3Sn superconducting radiofrequency cavities for high Q applications,” *Applied Physics Letters*, vol. 106, p. 082601, 02 2015.
- [7] D. B. Liarte, S. Posen, M. K. Transtrum, G. Catelani, M. Liepe, and J. P. Sethna, “Theoretical estimates of maximum fields in superconducting resonant radio frequency cavities: stability theory, disorder, and laminates,” *Superconductor Science and Technology*, vol. 30, no. 3, p. 033002, 2017.
- [8] G. Catelani and J. P. Sethna, “Temperature dependence of the superheating field for superconductors in the high- κ london limit,” *Physical Review B*, vol. 78, no. 22, p. 224509, 2008.
- [9] F. P.-J. Lin and A. Gurevich, “Effect of impurities on the superheating field of type-II superconductors,” *Physical Review B*, vol. 85, no. 5, p. 054513, 2012.
- [10] T. Kubo, “Superfluid flow in disordered superconductors with dynes pair-breaking scattering: Depairing current, kinetic inductance, and superheating field,” *Physical Review Research*, vol. 2, no. 3, p. 033203, 2020.
- [11] S. Posen and D. L. Hall, “ Nb_3Sn superconducting radiofrequency cavities: fabrication, results, properties, and prospects,” *Superconductor Science and Technology*, vol. 30, no. 3, p. 033004, 2017.
- [12] U. Pudasaini, G. V. Eremeev, J. W. Angle, J. Tuggle, C. E. Reece, and M. J. Kelley, “Growth of Nb_3Sn coating in tin vapor-diffusion process,” *Journal of Vacuum Science & Technology A*, vol. 37, no. 5, 2019.
- [13] R. Porter, T. Arias, P. Cuevas, J. Ding, D. Hall, M. Liepe, D. Muller, N. Sitaraman, *et al.*, “Update on Nb_3Sn progress at Cornell University,” *Proc. of IPAC 2018*, 2018.
- [14] G. Eremeev, U. Pudasaini, S. Cheban, J. Fischer, D. Forehand, S. Posen, A. Reilly, R. Rimmer, and B. Tennen, “Preservation of the high quality factor and accelerating gradient of Nb_3Sn -coated cavity during pair assembly,” *arXiv preprint arXiv:2307.10268*, 2023.
- [15] G. Eremeev, W. Crahen, J. Henry, F. Marhauser, U. Pudasaini, and C. Reece, “Rf performance sensitivity to tuning of Nb_3Sn coated cebaf cavities,” in *Proc. SRF’19*, no. 19 in International Conference on RF Superconductivity, pp. 55–59, JACoW Publishing, Geneva, Switzerland, aug 2019. <https://doi.org/10.18429/JACoW-SRF2019-MOP015>.
- [16] E. Viklund, D. N. Seidman, D. Burk, and S. Posen, “Improving Nb_3Sn cavity performance using centrifugal barrel polishing,” *Superconductor Science and Technology*, vol. 37, no. 2, p. 025009, 2024.

- [17] S. Posen, J. Lee, D. N. Seidman, A. Romanenko, B. Tennis, O. Melnychuk, and D. Sergatskov, “Advances in nb3sn superconducting radiofrequency cavities towards first practical accelerator applications,” *Superconductor Science and Technology*, vol. 34, no. 2, p. 025007, 2021.
- [18] W. G. Sloof, *Self Healing in Coatings at High Temperatures*, pp. 309–321. Dordrecht: Springer Netherlands, 2007.