



Review of Thermal Treatments: Effects, Reliability, and Open Questions

Sam Posen SRF Conference 2023 June 28, 2023 Sincere thanks for contributions from Dan Gonnella, Kensei Umemori, Marc Wenskat, Pashupati Dhakal, Matthias Liepe, Fabien Eozenou, Hayato Ito, Genfa Wu, Feisi He

Review of Bulk Niobium Heat Treatments: Timeline



8/19/2023 Sam Posen - SRF'23 - Thermal Treatments

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• Nitrogen doping turns 10 this year!



Image from Amazon

Nitrogen Doping



• Nitrogen doping turns 10 this year!

"recipe" is typically referred to as the "2/6 N-doping" NIM-A, Vol. 883, pp. 143-150 (2018)

- Proposed birthday for N-doping:
 - February 6 (USA)
 - June 2 (everywhere else)



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Image from Amazon



- Nitrogen doping is now successfully implemented in production for LCLS-II, resulting in the highest Q₀ SRF linac to date
- Very high reliability is now achieved with industrial vendors, but it took substantial efforts
- Impetus and investments from LCLS-II efforts helped to drive progress in these challenges

Open question: Cause of lower quench field

Open question: Cause of anti-Q-slope – but there are theories (e.g. non-equilibrium) —

However, materials studies helped achieve a mature understanding of the **impact** of N-impurity profiles Superconductor Science and Technology

FAST TRACK COMMUNICATION

Nitrogen and argon doping of niobium for superconducting radio frequency cavities: a pathway to highly efficient accelerating structures

A Grassellino¹, A Romanenko¹, D Sergatskov¹, O Melnychuk¹, Y Trenikhina², A Crawford¹, A Rowe¹, M Wong¹, T Khabiboulline¹ and F Barkov¹

Published 22 August 2013 • © 2013 IOP Publishing Ltd

Superconductor Science and Technology, Volume 26, Number 10

Citation A Grassellino et al 2013 Supercond. Sci. Technol. 26 102001 DOI 10.1088/0953-2048/26/10/102001



Field-Enhanced Superconductivity in High-Frequency Niobium Accelerating Cavities

M. Martinello,^{*} M. Checchin, A. Romanenko, A. Grassellino, S. Aderhold, S. K. Chandrasekeran, O. Melnychuk, S. Posen, and D. A. Sergatskov *Fermi National Accelerator Laboratory, Batavia, Illinois 60510, USA*

Based on the experimental data, our hypothesis is that the frequency dependence of the temperature-dependent surface resistance may be a consequence of the quasiparticle distribution being out of equilibrium, with the extent of the nonequilibrium behavior depending on the resonant frequency and on the types and concentration of impurities.

Nitrogen-Doped Cavity Experience in LCLS-II





²⁰¹⁶ Heat Treatment for Flux Expulsion

- ~900 C heat treatment to reduce flux pinning, improve expulsion of ambient magnetic fields during cooldown
- Implemented successfully in LCLS-II
 production
- Improve Q by changing bulk, not surface
- Still challenging to find the right temperature to balance expulsion and mechanical properties, variations from one niobium production lot to another

Open question: specification to ensure strong flux expulsion without making material too soft





- Motivation for N-infusion: knew N-doping helped Q: try to combine with low T bake to get both high Q and high gradient
- Procedure: 1) 800 C+ to remove oxide, 2) plateau at 120 C for 48 hours with nitrogen injection
- Has led to very high cavity gradients
- Reproducibility in other labs has been challenging, likely due to no EP after 800 C in furnace -> even small contamination from furnace is a challenge
- To avoid degradation from contamination, great care is needed: clean furnace, caps

PAPER • OPEN ACCESS

Unprecedented quality factors at accelerating gradients up to 45 MVm⁻¹ in niobium superconducting resonators via low temperature nitrogen infusion

A Grassellino¹, A Romanenko¹, Y Trenikhina¹, M Checchin¹, M Martinello¹, O S Melnychuk¹, S Chandrasekaran¹, D A Sergatskov¹, S Posen¹, A C Crawford¹ + Show full author list Published 8 August 2017 · © 2017 IOP Publishing Ltd

Superconductor Science and Technology, Volume 30, Number 9

Focus on The Jan Evetts SUST Award 2017

Citation A Grassellino et al 2017 Supercond. Sci. Technol. 30 094004 DOI 10.1088/1361-6668/aa7afe







Results of Nitrogen-infusion at KEK

Example of good results



Example of bad results





- N-infusion at KEK showed improvement of Q-value, but not for gradient.
- Reproducibility of N-infusion was very poor.
- Very often, Q-degradation was observed. → Contamination from the furnace??
- Quality control was difficult.

Slide from Kensei Umemori

Lessons learned on furnace infrastructure

"Forensic" study to identify key process parameters

- Studies show a correlation between furnace pressure and/or partial pressure of CO/CO₂ to surface resistance
 - \rightarrow lower pressures resulted in higher \textbf{Q}_{0}
- Clear excess of carbon in hot spots vs. cold spots shown by SIMS, SEM and EBSD/EDX studies
- No nitrogen observed in any samples from infusion runs
- Conclusions:
 - RGA analysis is mandatory low partial pressure of CO and CO₂ is crucial.
 - Extreme cases of carbon contamination cause grain decoupling.
 - Minor carbon contamination will prevent N adsorption and limit performance improvement



Related publications:

Marc Wenskat et al 2020 Supercond. Sci. Technol. 33 115017 Arti Dangwal Pandey et al 2021 Appl. Phys. Lett. 119 194102

C. Bate, PhD Thesis 2021

Slide from Marc Wenskat



JLAB Infusion Recipe and Results P. Dhakal Talk: 06/27/2023 10:30 AM E_{acc} (MV/m) **Cavity Surface Temperature** 18 24 42 6 12 30 36 10^{11} Hold (800C/3hrs) Temperature N₂ Inject to furnace at different temperature during ramp down (YYY °C) Ramp Up $\ddot{\mathcal{O}}$ 10¹⁰ Hold (120-200 ° C/24, 48hrs) 0.75 GHz 1.3 GHz 1.5 GHz 3.0 GHz Time 10^{9} 25 50 75 100150 175 125 () 0.75 – 3.0 GHz single-cell fine-grain Nb cavities (RRR>300) • $B_{n}(mT)$ **N-infusion**: 800 °C/3h, N₂ at ~25 mTorr ~300 °C, cooling to hold temperature maintained for 48 h. •

• R&D to understand the mechanism and depth of the N2 diffusion (in any) on going (SIMS and XPS)

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• Multi-cell are being prepared to valid the recipe.

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Low-T Doping at Cornell



Treatment	Anneal #1	Anneal #2	Infusion	Gas	Anneal #3	Chemical Etching ^{1,2,3}
C1P1	800 °C (3 hr)		160 °C (48 hr)	$Ar + CO_2$	_	HF
C1P2	800 °C (3 hr)	_	160 °C (48 hr)	$Ar + CO_2$	—	HF + OP (27 nm)
C1P3	800 °C (3 hr)	160 °C (3 hr)	160 °C (24 hr)	N_2	_	_
C1P4	800 °C (3 hr)	160 °C (3 hr)	160 °C (24 hr)	N_2	—	OP (54 nm)
C2P1	900 °C (3 hr)	160 °C (3 hr)	160 °C (4.5 dy)	N_2	—	_
C2P2	900 °C (3 hr)	160 °C (3 hr)	160 °C (4.5 dy)	N_2	—	HF
C2P3	900 °C (3 hr)	160 °C (3 hr)	160 °C (4.5 dy)	N_2	—	HF (×2)
C3P1	800 °C (3 hr)	160 °C (3 hr)	160 °C (48 hr)	N_2	—	_
C3P2	800 °C (3 hr)	160 °C (3 hr)	160 °C (48 hr)	N_2	—	HF (×2)
C3P3	800 °C (3 hr)	160 °C (3 hr)	160 °C (48 hr)	N_2	—	HF (×2) + EP (100 nm)
C4P1	800 °C (5 hr)		160 °C (48 hr)	$Ar + CO_2$	—	OP (54 nm)
C5P1	800 °C (12 hr)		160 °C (48 hr)	N_2	160 °C (168 hr)	_
C5P2	800 °C (12 hr)	—	160 °C (48 hr)	N_2	160 °C (168 hr) + 75 °C (6 hr)	—

- Performed various 160C bakes in N₂ and Ar+CO₂
- All cavities showed strong reduction in BCS resistance with field
 - *HF rinsing required in some cases to remove surface contamination (Ti)*



²⁰¹⁸ **2-Step Low-T Bake**

arxiv > physics > arXiv:1806.09824

 4×10^{10} 3×10^{10}

2 × 10¹⁰

Physics > Accelerator Physics

[Submitted on 26 Jun 2018]

Accelerating fields up to 49 MV/m in TESLA-shape superconducting RF niobium cavities via 75C vacuum bake

A. Grassellino, A. Romanenko, D. Bice, O. Melnychuk, A. C. Crawford, S. Chandrasekaran, Z. Sung, D.A. Sergatskov, M. Checchin, S. Posen, M. Martinello, G.Wu

- Accidental oven setting, adding 4 hours at 75 C before 48-hour 120 C bake, led to improved performance
- Unprecedented gradients in 1.3 GHz single cell TESLA cavities, up to 50 MV/m
- Consistently high gradients in single cells observed at Fermilab (always combined with cold EP) – reproduced in vertical tests at other labs

Open questions: cause of improvement still unclear - needs to be studied; also bifurcation





2-Step Baking results on 1AC3 1300MHz 1Cell

- Cavity used for different R&D experiments (alternative acid concentration, doping...)
- Best gradient: 37MV/m after standard baking (48h @120°C)
- Last treatment: + VEP 15µm (T<15°C, 19V)
- 2-step baking : Field Emission observed
- + HF rinse: significant improvement: Gradient > 50 MV/m achieved
- Cavity tested again after new cleanroom assy and modified calibration

4h @ 75°C + 48h @120 °C



1C cavity on VEP set-up





Slide from Fabien Eozenou Ryo Katayama, TTC@Aomori, 2022 [2-step baking & mid-T baking of 9-cell SRF cavities]

Result of cold-EP + 2-step bake (70 °C 4 h) at KEK

- Comparison of Q-E between "STD EP + bake" and "cold EP + 2-step bake" is shown below.
 - Red: KEK STD EP (25-30 °C) + STD bake (120 °C 48 h bake).
 - Blue: Cold EP(~14°C) + 2-step bake (70 °C 4 h bake + 120 °C 48 h bake).

Cavity temperature during measurement

- R8 STD ... at 2.07K
- Other cases ... at 2.0 K (2.00~2.01 K)



- Higher Q tends to be obtained for 2-step baked cavity.
- On the other hand, improvement of gradient can not be obtained.
 - ⇒ Why higher gradient for high-Q/high-G recipe is difficult at KEK? Cavity? EP? Furnace? Baking procedure? Cooling procedure at VT?

Slide from Kensei Umemori

High Gradient Cryomodule Collaborative Effort

 Eight 9-cell cavities under consideration for HGC reached an average of 41.0 MV/m, several now tanked





- In-situ mid-T bake: cavity assembled
- Developed initially for quantum applications to remove Nb₂O₅ layer, enabling high Q at mK and low photon counts
- SIMS studies determined 300-450 C temperature $_{\circ}$
- Accelerator regime: anti-Q-slope similar to Ndoping; high Q even after oxide regrows
- Currently understood mechanism is adding impurities like N-doping, but in this case oxygen
- Advantages compared to N-doping:
 - No post-doping light EP: key for complex geometries where uniform EP is challenging; removes step
 - Lower temp reduces risk of contamination

PHYSICAL REVIEW APPLIED

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Ultralow Surface Resistance via Vacuum Heat Treatment of Superconducting Radio-Frequency Cavities

S. Posen, A. Romanenko, A. Grassellino, O.S. Melnychuk, and D.A. Sergatskov Phys. Rev. Applied **13**, 014024 – Published 14 January 2020



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Three-Dimensional Superconducting Resonators at $T < 20~{\rm mK}$ with Photon Lifetimes up to $\tau = 2~{\rm s}$

A. Romanenko, R. Pilipenko, S. Zorzetti, D. Frolov, M. Awida, S. Belomestnykh, S. Posen, and A. Grassellino Phys. Rev. Applied **13**, 034032 – Published 12 March 2020





Q-E curve with various baking temperature

٠.

25

35

30

••

20

o°

10

 5×10^{1}

4×10¹

3×10¹

 2×10^{10}

1010

5×10⁸

4×10⁶

3×10⁶

2×10⁶

120C 48h baking (R-9 15th VT

200C 3h baking (R-4 11th VT)

250C 3h baking (R-4 16th VT) 300C 3h baking (R-4 10th VT)

300C 3h baking (R-8 13th VT) 350C 3h baking (R-4 13th VT)

500C 3h baking (R-8 16th VT)

600C 3h baking (R-4 12th VT)

800C 3h baking (R-8c 5th VT)

Radiation of 350C 3h baking Radiation of 400C 3h baking

Radiation of 500C 3h baking

Badiation of 300C 3h baking (B-4)

10

15

Havato Ito. 2023/06/13

400C 3h baking (TE1AES018 7th V



Cavity temperature during measurement

- 120 ~ 600°C baking ... at 2.0 K (2.00~2.01 K)
- 800°C baking ... at 2.1 K (2.07K)

$250 \sim 400^{\circ}$ C 3 h

[hSv/h]

≥

×

 10^{2}

- Extremely high Q value and anti-Q slope are observed
- Highest Q value at 2.0 K is ~ 5E10 for 300°C baked cavity

Standard recipe (120°C 48 h), 200°C 3 h

- 200°C baked cavity follows the standard recipe (120°C 48h)
- Q-E behavior at low Eacc is slightly different

500 ~ 800°C 3 h

High Q value wasn't observed





JOURNAL ARTICLE

Influence of furnace baking on Q–E behavior of superconducting accelerating cavities a

H Ito 💌, H Araki, K Takahashi, K Umemori

Progress of Theoretical and Experimental Physics, Volume 2021, Issue 7, July 2021,

071G01, https://doi.org/10.1093/ptep/ptab056

- Under US/Japan collaboration. process was shared with KEK and they adjusted it to their facility
- Furnace mid-T bake: unassembled cavity in furnace
- More accessible than in-situ mid-T bake, just use normal furnace!



Slide from Marc Wenskat

Heat treatment R&D is ongoing and reproducible

Furnace upgrades improve research capabilities

- Two very good furnaces at DESY and U Hamburg
 - New single-cell furnace & refurbished niobium retort furnace for 9-cell cavities.
- 2-step bake

DESY.

- 4h@75°C/24@130°C as standard treatment in the last 4 years.
- Thorough analysis not done yet no obvious improvement compared to regular 120°C bake.
- Mid-T heat treatment
 - Reproducible and stable process.

R

- RF tests of 14 single-cell cavities, treated in three furnaces.
- All succeeded in improving R_{BCS} (anti-Q-slope) and resulted in varying maximum accelerating field.
- Partially enhanced R_{res} is under investigation.

für Bildung

und Forschung

Related contributions at SRF: C. Bate – Poster ID 2030 R. Ghanbari – Poster ID 2101 A. Zaidman – Poster ID 2421 M. Wenskat – Poster ID 2054 & Talk 2901





All in DESY Niobium retort furnace medium temperature treated cavities. Open markers depict curves which were intentionally stopped to avoid quenching. Page 19

Baking 300 °C – 400 °C at JLab

Slide from Pashupati Dhakal

For More info: See Poster by E. Lechner



- SIMS measurements show that O in the main impurity for mid-T baked cavities [1]
- SIMS measurements show fewer impurities introduced during vacuum annealing vs N doping
- Baking too long or too high in temperature shows EP-like performance (O absorbed into bulk)
- Sensitive to multipacting at ~20 MV/m
- High Q₀ but low E_{max} observed in multi-cell cavities

Lechner, E. M., et al. *Applied Physics Letters* 119.8 (2021): 082601.
 Angle, Jonathan W., et al. *Journal of Vacuum Science & Technology B* 41.3 (2023).



Mid-T Bake for Fermilab PIP-II LB650 Cryomodule

- High Q recipe using Mid-T furnace baking was endorsed by a review committee and adopted for LB650 cryomodules
- Uniform few-micron EP proved challenging on LB650 geometry, but mid-T bake doesn't need it!



LB650 Cavity Processing Final Design Review Report, April 4, 2023



G. Wu | LB650 Processing FDR | Cavity Processing Development



Mid-T furnace baking of 1.3GHz 9-cell & 650MHz cavities

- 14 1.3GHz-9cell cavities was mid-T baked at 300C for 3hrs upto now. Average Q0~4.5e10 at 16&21MV/m (ss flange loss corrected), average Eacc ~ 26MV/m
- After mid-T baking (300 C for 3 h), 650 MHz 1cell cavities achieved state-of-the-art Q0, which reached 6.4E10 @ 30 MV/m (at 2.0 K).





First cryomodule of 8 mid-T cavities tested at IHEP

- IHEP is developing the cryomodule for the DALS project
- Dynamic heat load at 133MV is 61W (Q0~3.6±0.5e10 at 16MV/m)
- Dynamic heat load at 174MV is 104W (Q0~3.6±0.4e10 at 21MV/m)
- Usable voltage up to now (stable>1hr, <1mSv/h) is 185MV (~22MV/m), which is limited by 5.2kW power source.
- More details will be published later, after all measures are finished.



Slide from Feisi He





8/19/2023 Sam Posen - SRE'23 - Thermal Treatments

Review of Bulk Niobium Heat Treatments: Timeline





- In the last decade, several thermal treatments were developed, leading to unprecedented Q and gradient
- Not all are reproduceable everywhere high gradient heat treatments N-infusion & 2-step bake have shown ~50 MV/m in some cases, others not
- N-doping took a lot of development to make reproducible but is now industrialized. Still more development was needed for LCLS-II-HE
- Furnace Mid-T bake has shown to be comparatively straightforward to reproduce successfully, even when scaling up to multicells or to complex geometries
- Mid-T bake benefits a lot from the R&D done on Ndoped cavities for reducing trapped flux dissipation

