ISBN: 978-3-95450-211-0 doi:10.18429/JACoW-SRF2019-TUP057

STUDY OF FLUX TRAPPING VARIABILITY BETWEEN BATCHES OF TOKYO DENKAI NIOBIUM USED FOR THE LCLS-II PROJECT AND SUBSEQUENT 9-CELL RF LOSS DISTRIBUTION BETWEEN THE BATCHES*

A. D. Palczewski†, Jefferson National Laboratory, Newport News, VA, USA
O. S. Melnychuk, and D. A. Sergatskov, Fermi National Accelerator Laboratory, Batavia IL, USA
D. Gonnella, SLAC National Accelerator Laboratory 2575 Sand Hill Road, Menlo Park CA, USA

Abstract

During the LCLS-II project, a new batch of niobium was procured from Tokyo Denkai Co Ltd to make additional cavities. The original production material came from two vendors, Tokyo Denkai Co., Ltd. (TD) and Ningxia Orient Tantalum Industry Co., Ltd. (OTIC/NX)). It was found TD niobium required a lower annealing temperature (900 °C) to obtain satisfactory flux expulsion characteristics compared to NX, which required a slightly higher annealing temperature (950–975 °C). To ensure the new TD material performed equivalent to the niobium produced three years ago after 900 °C annealing; each heat lot of niobium had its flux expulsion characteristics parametrized using single cell cavities and custom thermal treatments developed for each lot. Subsequent pure heat lot 9-cell cavities were made and tested. We will look at the flux expulsion characteristics of each lot through single cell cryogenic cycling, and RF loss of the 9-cell cavities produced using the individual heat lots.

INTRODUCTION

The SLAC National Accelerator Laboratory is currently constructing a major upgrade to its accelerator, the Linac Coherent Light Source II (LCLS-II). Several Department of Energy national laboratories, including the Thomas Jefferson National Accelerator Facility (JLab) and Fermi National Accelerator Laboratory (FNAL), are participating in this project. The 1.3-GHz cryomodules for this project consist of eight cavities produced by two vendors; Research Instruments GmbH in Germany (RI) and Ettore Zanon S.p.a. in Italy (EZ) using niobium cell material from Tokyo Denkai Co., Ltd. (TD) and Ningxia Orient Tantalum Industry Co., Ltd. (OTIC/NX)).

The initial production cavities showed multiple deficiencies, including manufacturing flaws from one vendor, high rates of field emission from both vendors, and Qo reduction from a previously unknown flux trapping/pinning material dependence [1]. Additional new cavities material from Tokoyo Denkai (TD) was purchased; TD showed the best flux expulsion and RF performance at the lowest heat treatment temperature from the original two niobium vendors. This material was used to make new cavities for five spare modules as well as for replacement cavities that had unrepairable manufacturing defect early in production [2]. Because there is still not a full understanding of the reason why the TD material performed better than the OTIC material at lower annealing temperatures; single cell cavities from each new niobium heat treatment lot are made ahead of the 9-cell cavities. To ensure the new TD material performed equivalent to the niobium produced 3 years before after 900°C annealing; each heat lot of niobium had its flux expulsion characteristics parametrized and custom thermal treatments developed for each lot using the single cell cavities. Subsequent pure heat lot 9 cell cavities were made and tested. We will look at the flux expulsion characteristics of each lot, and RF loss of the 9-cell cavities produced using the individual heat lots.

NIOBIUM UNDER ANALYSIS

The additional material procured during the LCLS-II production run used the DESY/XFEL specification [3]. The intent was to order identical TD niobium used for the initial production run.

Lot Information

Niobium delivered from Tokyo Denkai and procured under the LCLS-II contract came in multiple sets of heat lots because of the large quantity needed for the project. Heat lots contain ~ 140-150 sheet or enough material to make up to 8 cavities depending on RF sorting and loses during manufacturing. Within a single heat lot, two mother ingots are used and processed together. TD supplies two sets of information important to this paper. One is the RRR of each mother ingot, and two is the characterization of 2 sheets from each heat lot. The hardest and the softest sheet from the heat lot (containing two ingots) is selected from all sheet in the lot. Hardness testing is done one every sheet. These two sheets may come from the same ingot or one from each ingot depending on the final hardness measurements. The assumption is these sheets are the outliers of the lots, and all other sheets will lye somewhere in-between the two sheets. We do not fully agree with the last statement as the surface hardness is more a function of the final leveling pass than the post-annealing state, but in general, has been enough to certify the lots for RF

*Authored by Jefferson Science Associates, LLC under U.S. DOE Contract No. DE-AC05-06OR23177. Additional support provide by LCLS-II project under US DOE Contract DE-AC02-766SF00515.
† ari@jlab.org

ISBN: 978-3-95450-211-0 doi:10.18429/JACoW-SRF2019-TUP057

performance [4]. The final crystallography of the heat lot determined by the manufacturer is supplied with the order. LCLS-II (using the XFEL spec) requires the final crystal structure to be between ASTM 5 and 7, with the majority being ASTM 6. The full summary table of the niobium, manufacturer supplied information, and flux expulsion/heat treatment temperature presented in the next sections is present in Table 1.

Single Cell Sheet Selection Requirements

Two sheets from the highest ASTM (smallest crystal) ingots in the lot were pulled at random to make single cell cavities. These sheets could come from anywhere in the ingot lot. The single cell cavities received the same processing step as the production 9-cells cavities including full chemistry and doping at 800°C during the 900°C thermal cycles [5]. The single cell cavity must then be characterized before the heat treatment of the 9-cell cavities from the lot. The single cell required 80% flux expulsion (~1.55 flux expulsion ratio) at a 5K delta to clear the 9-cells for thermal treatment at 900°C. Any single cell below this would require additional evaluation before the lot could be cleared.

9-cell Sorting

For the first time, the niobium for a mass-produced SRF cavities order was sorted into pure heat lot cavities. During the initial LCLS-II production the NX was sorted into ASTM lots, but not individual heat lots [1, 5]. The 9-cell made in this production run could only be made from a single heat lot, mixing of the two ingots within the lot was allowed. The sorting was done to only heat treat each lot at the highest temperature necessary to meet the LCLS-II flux expulsion specification without overheating a lot – reducing the yield strength unnecessarily. In the past sorting of niobium between lots was not see as a requirement, XFEL data, as well as LCLS-II quench field data, support the case when a single recipe is used for all cavities [1, 6].

There were two variances added to the cavity contract to allow a small amount of mixing between lots. One, the end-group material could be from the previous LCLS-II production run or the previous cavity lots already qualified. End-group manufacturing takes up a large portion of the total production time. The initial niobium deliveries did not line up with the need to start all end-group manufacturing of all lots. Two, the mixing of an ASTM 5/6 lot cavity with a couple of sheet from another lot 6 was allowed in one special case – one ASTM 5.75 average cavity.

SINGLE CELL FLUX EXPULSION RESULTS

The flux expulsion ratio curves for the 11 single cell cavities heat to 900°C are presented in Figure 1. Each ASTM is separated by color: ASTM 5 sheet red, ASTM 6 sheet black and ASTM 7 sheet blue. Details on the production of these curves and instrumentation techniques are published in previous works [5, 7]. The solid blue symbolled lots all required a higher heat treatment temperature to meet specification.

Table 1: Summary of All Niobium Analysed in This Paper and Flux Expulsion/Final 9-cell Thermal Treatment Needed for the Next Sections

Heat lot	Heat run	ASTM sheet	ASTM lot average	Howest ingot RRR	Highest ingot RRR	Flux expulsion ratio after 900°C @ 5K delta	Flux expulsion % after 900°C @ 5K delta	9 cell heat treatment
92	HT9-773	7	6	379	398	1.5	75	950/900°C
93	HT9-982	7	6.5	433	515	1.57	85	900°C
94	HT9-1002	7	6	356	395	1.6	90	900°C
95	HT9-1012	5	5	349	347	1.67	100	900°C
96	HT9-1013	NA	5.5	322	368	NA	NA	900°C
97	HT9-1025	6	6	344	367	1.57	85	900°C
98	HT9-1046	NA	7	402	415	NA	NA	900°C
99	HT9-1052	7	6.5	409	415	1.57	85	900°C
100	HT9-1065	7	6.5	395	399	1.55	82	925°C
101	HT9-1074	7	6.5	373	446	1.5	75	975°C
102	HT9-1081	6	6	339	340	1.595	89	900°C
103	HT9-1093	6	6	336	364	1.61	91	900°C
104	HT9-1101	7	6.5	410	433	1.62	93	900°C

ISBN: 978-3-95450-211-0 doi:10.18429/JACoW-SRF2019-TUP057

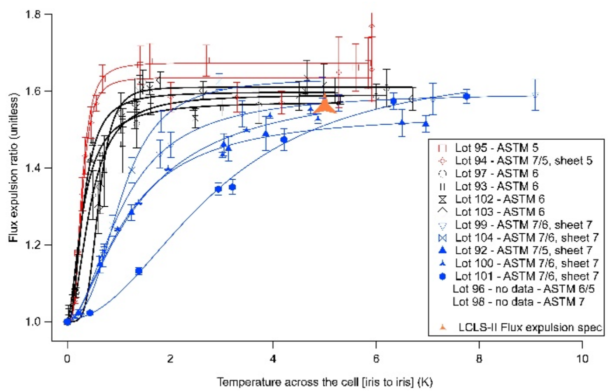

Figure 1: Flux expulsion vs. thermal difference (temperature across the cell) between iris's when the cells equator reached 9.25K. Each color represent each ASTM grain size defended by the manufactured lot, and the symbols are for each lot. Each curve is fit with a Hill-equation as a guide to the eye.

During the manufacturing of the single cell, an error occurred which at the time we though ruined three cavities. The error required 2 of the lots to be evaluated with 9-cell cavities (see next section) as no other spare material was available, and one of the single cell from - Lot 94 - was remade. In the case of lot 94, ASTM 5 sheets were used for the single cells, this material was kept as a backup and not used except for 2 sheets.

There are multiple insights we can extract from the single cell data set.

- There is a strong scaling in the flux expulsion ratios vs grain size, where the largest grain (smallest ASTM number) niobium produces the best flux expulsion results
- The plateau in the ASTM 5 and ASTM 6 material, although clustered at 5K delta, does not appear to have any other scaling.
- The ASTM 5 and ASTM 6 niobium flux expulsion is the same for a 2K delta or 5K delta (plateaued ay 2K).
- The ASTM 7 material has a very broad distribution in flux expulsion ratios.
- The worst material, lot 101, was reheat treated at 925°C with no change (not shown).

9 CELL VERTICAL TEST RF FLUX EXPULSION RESULTS

Two of the lots, 96 and 98, required 9-cell analyses as the single cell for these two lots (and lot 94) sustained damage during manufacturing and no spare material remained to make new single cells. For these lots, the first 9-cell cavity in each lot was heat treated to 900°C and then sent without a helium tank installed to JLab for testing. Instead of the normal flux expulsion measurements, RF tests in a 5 mG and then a 20 mG field cool were used. The trapped flux losses (change in surface resistance) and therefore flux expulsion would then be extrapolated RF change using a 1 nΩ/mG trapped flux loss [5, 8]. A summary of lot 96 and 98 results are shown in Table 2.

Table 2: RF Loss Analysis from Trapped Flux for Lots 96 and 98 Test - 9 Cell Cavities *

	Q0 @ 16MV/m 5mG field	Q0 @ 16MV/m 20mG field	Change in RS	% field trapped
Lot 96	3.70E+10	2.75E+10	2.53E-09	17%
Lot 98	2.9E+10*	2.45E+10	NA	NA

Lot 96 (ASTM 5.5) showed an expected flux expulsion (% trapped) close to expectations for ASTM 5.5 material with a surface resistance change of ~ 2.5 nΩ, corresponding to ~17% trapping or a flux expulsion ratio of ~1.6. Lot 98 (ASTM 7) had mixed results with the 5mG cooldown stalling close to Tc where the bottom of the cavity would trap all fields, giving an unrealistic low Q0 expectation, for production. Because of schedule constraints, the test was not redone. The 20 mG field cool proceeded and the test barely missed the cavity spec of 2.5×10^{10}. Since the cryomodule spec is 5 mG, the material was deemed "good enough" and the heat treatment was left at 900 °C. In hindsight and with the full data set now complete we would have re-done the 5 mG field cool and the batch should have been heated at a slightly higher temperature to give more flux expulsion headroom – see next section and lots 100.

9 CELL PRODUCTION RF RESULTS

A total of 50 of the 86 9-cell cavities were tested as of June 1 2019. Of the 36 not tested 16 were transferred to the LCLS-II HE project (Lot 100 and 101, two of the most interesting ASTM 6.5 lots) and there results will be published under that project [9]. The remaining 20 should have their testing completed by the end of July 2019 and published in a future article. For the cavities already tested only 2 had their heat treatment temperature changed. These were 2 cavities from Lot 92. At the time we could sort lot 92 into individual ingots as well as heat lot, 2 cavities from the ASTM 7 ingot heated to 950 °C and 2 from an ASTM 6 ingot left at 900°C.

* The cooldown of Lot 98 test cavity in a 5mG field stalled close to Tc, flux expulsion ratio close to zero.

ISBN: 978-3-95450-211-0 doi:10.18429/JACoW-SRF2019-TUP057

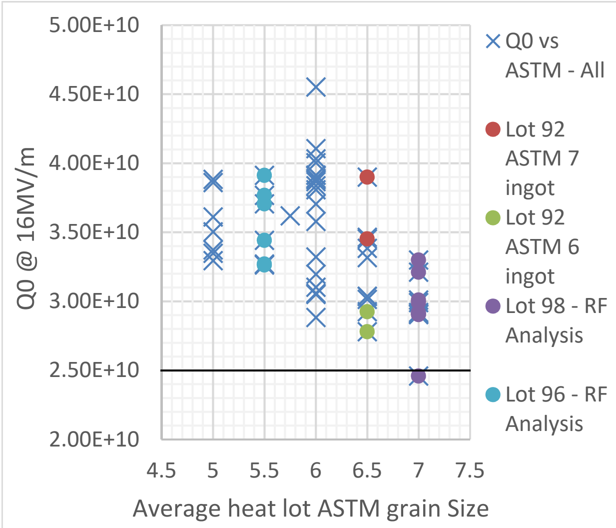

Figure 2: Q0 @ 16MV/m vs Average heat lot ASTM grain size. The light blue "x" is all data available, with other subsets of data overlapped in closed circles.

Q0 vs ASTM Grain Size

Figure 2 shows the complete Q0 measurements @ 16 MV/m for all cavities. The data is sorted by the average ASTM grain size for the heat lot (2 manufacturer test sheets average grain size, **not** single cell sheet ASTM). The specification for the project is 2.5×10^{10} @ 16MV/m - horizontal (black line). All but one cavity meets the vertical test specification, and that cavity had a Q0=2.45×10^{10}. The cavity was tested in D5 at JLab which technically has higher magnetic fields than the specified for the project – next section.

There are multiple insights we can extract from the 9-cell data set.

- There is a clear scaling of ASTM grain size and Q0 results after a 900 °C heat treatment on TD niobium, starting with ASTM 6 there is a dropoff in performance, but within specification.
- If optimal performance is required ASTM 6 and above niobium requires a heat treatment temperature greater than 900 °C.
- Lot 98, no single cell flux expulsion test only 9 cell RF data, in hindsight should have been heat-treated at 925 °C to 950 °C to guarantee 100% pass rate but was still within spec for LCLS-II.
- Lot 92's ASTM 6 ingot cavities should have been heat-treated at 950 °C like the ASTM 7 cavities to maximize performance.
    - There appears to be a stronger correlation between heat lots than ingot lot for flux expulsion characteristics. This suggests the processing, and not the mother material is a larger driver of flux expulsion performance.
    - All future heat lots should receive the same annealing temperature to maximize performance independent of the lower ASTM ingot in the lot.

Results vs. Background Magnetic Field

To first order, the use of single-cell cavities flux expulsion results to predict the trapped RF losses seems reasonable with the higher ASTM material showing slightly higher loses. The ASTM 6 niobium especially produces a broad distribution in Q0, even if we remove the one outlier above 4.5×10^{10}. To understand if these results are within expectations the test setups background magnetic field and associated trapped flux losses must also be taken into account.

Within the total of 50 RF tests, 43 had active magnetic field sensors monitoring the axial field direction during the cooldowns. - 13 at FNAL and 30 at JLab. These tests were performed in 5 different Dewar; 3 at JLab and 2 at FNAL. A summary table of the 43 RF tests is in Table 3.

Table 3: Test Setup, Estimated Constant Perpendicular Field Used for Trapped Flux Loss Estimations and Total Cavity Count in Each Setup for Figure 3.

Dewar number	D7	D8	D5	D2	D3
location	JLab	JLab	JLab	FNAL	FNAL
Number of cavities during the test	1	1	3	3	3
Estimated perpendicular background fields	1 mG	2 mG	5 mG	2 mG	5 mG
Test count	2	5	22	4	9

For each RF test we extrapolated the total trapped field using the average of the three magnetometers monitoring the axial field in the Dewar right before Tc, and the measured average perpendicular fields from a survey from previous tests - which are **not** normally monitored during LCLS-II testing as they do not change between tests compared to the axial field. We then scaled the total trapped fielded using the flux expulsion results in Figure 1, and the 9-cell data from lots 96 and 98 (estimated) in Table 2. This trapped field data is plotted vs. Q0 to see if the losses are within expectations – Fig. 3. Three lines on the plot are for 0.8 nΩ/mG, 1 nΩ/mG and 1.2 nΩ/mG, flux trapping RF losses. The three losses is the range expected for the 2N6 doping recipes; the zero points were moved to the nominal 5nΩ surface resistance doping level [5, 10].

The scaling of the total trapped flux vs. Q0 for all but a couple of cavities are within our expectations. The surface resistance appears to only scale with the amount of trapped flux and not with the ASTM grain size. The spread in Figure 3 could have some other embedded uncertainties as well. For instance, we know the doping level RF losses can vary by about 1nΩ independent of the remnant field during testing; which would shift the zero point of the trapped magnetic field losses up and down. The worst material – higher flux trapping % - could have an even higher spread

ISBN: 978-3-95450-211-0 doi:10.18429/JACoW-SRF2019-TUP057

coming from other remnant fields which were trapped; for instance, the sometimes magnetic return te [5].

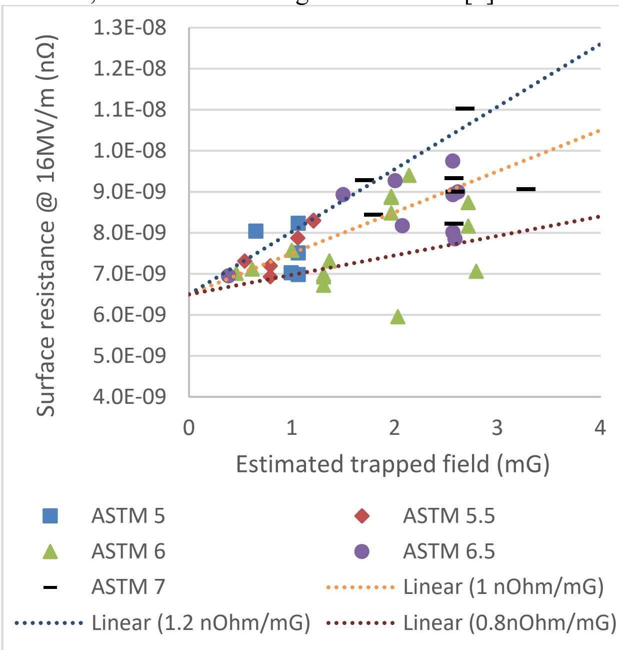

Figure 3: Estimated trapped magnetic field vs Q0 @ 16MV/m for the 43 9-cell cavities with magnetic field monitoring during cooldown. Each symbol represents a different average ASTM grain size. The three linear curves are the trapped magnetic field losses for 2N6 doping (centroid and error).

ADDITIONAL CONSIDERATION FOR FUTURE PROJECTS

For the first time niobium for a mass-produced SRF cavity order required sorting of pure heat lot cavities. In the past sorting of niobium between lots was not see as a requirement and only sorting between niobium vendors. The sorting created a unique logistical requirement for the cavity vendors. For instance, enough spare material from each lot must be made available to the cavity vendors to compensate for variance in the RF stack-up and general production losses during manufacturing; both add additional cost to production [2, 7]. The need for extra spare material was learned after one batch of single cell from lots 94, 96 and 98 needed remanufactured and there was no spare material for lots 96 and 98. In addition, in our opinion, the next High Q0 project, most likely LCLS-II HE, should change the flux expulsion specification from 80% expulsion at 5K delta, to 80% flux expulsion at 2K delta, as this would allow a loosening of the cryomodule cooldown rate [7, 11].

Keep in mind that increasing the cavity annealing temperature will inherently lower the yield strength of niobium; current research point to grain growth to increase flux expulsion [8, 12, 13]. Care must always be taken as the cavity temperature is increased to compensate for poor flux expulsion that the cavities may become too soft for handling. LCLS-II performed shipping tests on higher temperature heat treated cavities to ensure they were not too soft to ship but not to the extent performed for the XFEL production [14].

CONCLUSIONS

The use of single-cell cavities to analysis the flux trapping properties of a heat lot of high RRR niobium appears to be an excellent way to extrapolate the added RF losses on High Q0 9-cell cavities. Careful analysis of the test setup for the 9 -cell RF results yield the expected trapped flux losses for most of the cavities.

The larger variability in the flux expulsion of ASTM 6.5 and ASTM 7 heat lots needs more careful analysis to understand if this variability could be removed in the manufacturing stage. If one wanted to maximize the performance in these materials from TD, all batch would need a higher heat treatment than 900°C, at the cost of reducing the yield strength of the cavity. Later lots 100 and 101 annealing temperature increased to 925°C and 975°C after more information was known, and the softening of TD cavities was better understood.

Poorer, yet within LCLS-II specification ASTM 6.5 and 7 niobium heat-treated at 900°C, may show slightly lower results in the cryomodules where the 5K delta equivalent cooling is hard to archive in all portion of the LINAC.

ACKNOWLEDGEMENTS

Thank you to Sam Posen and the Applied Physics and Superconducting Technology Division at Fermi National Laboratory for the flux expulsion results of lot 101 single cell after 925°C – these are not shown as the flux expulsion did not change after the additional thermal treatment.

REFERENCES

[1] D. Gonnella *et al.*, “Industrialization of the nitrogen-doping preparation for SRF cavities for LCLS-II,” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 883, pp. 143-150, 2018.

[2] D. Gonnella *et al .,* “Industrial Cavity Production: Lessons Learnt to Push the Boundaries of Nitrogen-Doping”, presented at the 19th Int. Conf. RF Superconductivity (SRF'19), Dresden, Germany, Jun.-Jul. 2019, paper FRCAA3.

[3] W. Singer *et al.,* “Superconducting cavity material for the European XFEL,” *Superconductor Science and Technology*, vol. 28, no. 8, p. 085014, 2015.

[4] C. Antoine, M. Foley, N. Dhanaraj , “Physical properties of niobium and specifications for fabrication of superconducting cavities”, Fermi National Accelerator Laboratory Technical Division Note, TD-06-048. August 25, 2006. .

[5] A. D. Palczewski, F. Marhauser, A. Grassellino, and S. Posen, “Material Qualification of LCLS-II Production Niobium Material Including RF and Flux Expulsion Measurements on Single Cell Cavities”, in *Proc. LINAC'16*, East Lansing, MI, USA, Sep. 2016, pp. 199-202. doi:10.18429/JACoW-LINAC2016-MOPLR026

[6] W. Singer *et al.*, “Production of superconducting 1.3-GHz cavities for the European X-ray Free Electron Laser,”

ISBN: 978-3-95450-211-0 doi:10.18429/JACoW-SRF2019-TUP057

Physical Review Accelerators and Beams, vol. 19, no. 9, p. 092001, 2016.

[7] S. Posen *et al.,* "Flux Expulsion Lessons Learned from LCLS-II", presented at the 19th Int. Conf. RF Superconductivity (SRF'19), Dresden, Germany, Jun.-Jul. 2019, paper TUP039.

[8] S. Posen *et al.,* "Efficient expulsion of magnetic flux in superconducting radiofrequency cavities for high Q0 applications," *Journal of Applied Physics*, vol. 119, no. 21, p. 213903, 2016.

[9] D. Gonnella *et al.*, "The LCLS-II HE High Q and Gradient R&D Program", presented at the 19th Int. Conf. RF Superconductivity (SRF'19), Dresden, Germany, Jun.-Jul. 2019, paper MOP045. .

[10] M. Martinello *et al.,* "Trapped Flux Surface Resistance Analysis for Different Surface Treatments", in *Proc. 17th Int. Conf. RF Superconductivity (SRF'15*), Whistler, Canada, Sep. 2015, paper MOPB015, pp. 115-119.

[11] S. Posen *et al.*, "Role of magnetic flux expulsion to reach Q0>3×10^10 in superconducting rf cryomodules," *Physical Review Accelerators and Beams*, vol. 22, no. 3, p. 032001, 2019.

[12] H. Jiang *et al*., "Mechanical Properties of High RRR Niobium with Different Texture", *IEEE Transactions on Applied Superconductivity*, vol. 17, issue 2, pp. 1291-1294, 2007.

[13] P. D. A. Palczewski, F. Marhauser and C. Reece, "Flux expulsion as a function of grain growth and defect density of niobium for SRF application," to be published, 2019.

[14] A. Schmidt, R. Bandelmann, H. Brueck, A. Matheisen, and J. Schaffran, "Studies on Transportation of Superconducting Resonators and Beam Position Monitors - Quadrupol Units for the XFEL Project", in *Proc. 15th Int. Conf. RF Superconductivity (SRF'11*), Chicago, IL, USA, Jul. 2011, paper TUPO050, pp. 504-507.