

# REPRODUCIBILITY OF HIGH-Q SRF CAVITIES BY HIGH-TEMPERATURE HEAT TREATMENT\*

P. Dhakal<sup>#</sup>, G. Ciovati, P. Kneisel, G. R. Myneni, JLab, Newport News, VA 23606, USA

## Abstract

Recent work on high-temperature ( $> 600$  °C) heat treatment of ingot Nb cavities in a customized vacuum furnace for several hours showed the possibility of achieving  $Q_0$ -values of up to  $\sim 5 \times 10^{10}$  at 2.0 K, 1.5 GHz and accelerating gradients of  $\sim 20$  MV/m. This contribution presents results on further studies of the heat treatment process to produce cavities with high  $Q_0$  values for continuous-wave accelerator application. Single-cell cavities of different Nb purity have been processed through few cycles of heat-treatments and chemical etching. Measurements of  $Q_0$  as a function of temperature at low RF field and of  $Q_0$  as a function of the RF field at or below 2.0 K have been made after each treatment. Measurements by TOF-SIMS of the impurities' depth profiles were made on samples heat treated with the cavities.

## INTRODUCTION

An R&D program to improve the quality factor ( $Q_0$ ) of superconducting radio-frequency (SRF) niobium cavities was initiated at Jefferson Lab in 2010. A custom built ultra-high vacuum induction furnace with an all-niobium hot-zone was procured and installed at the lab with the intent to pursue systematic studies of the performance of ingot Nb cavities after high-temperature ( $> 600$  °C) heat-treatments without subsequent chemical etching [1]. The goal was to maximize the removal of hydrogen from the Nb and to prevent re-absorption by sealing the surface in order to increase  $Q_0$ . In February 2012 it was discovered that, after heat treatment (HT) at 1400 °C for 3 h, the  $Q_0(2.0$  K) of a 1.47 GHz cavity increased up to  $\sim 5 \times 10^{10}$  at an accelerating gradient  $E_{\text{acc}} = 20$  MV/m (corresponding to a peak surface magnetic field  $B_p \cong 90$  mT) [2]. Such high  $Q_0$ -value at 2.0 K was unprecedented at such accelerating gradients. Measurements on Nb samples heat-treated with the cavity at 1400 °C showed, compared to those with no HT or heat-treated at lower temperatures [3]:

- The presence of titanium oxide on the surface and a concentration of  $\sim 1$  at.% of Ti, diffusing to  $\sim 1$   $\mu\text{m}$  depth
- Reduced broadening of the peaks in the electronic density of states at the edges of the energy gap.

It was then realized that the titanium had sublimated from the cavity flanges which are made of Ti45Nb. Removal of  $\sim 1$   $\mu\text{m}$  surface layer by buffered chemical

polishing (BCP) reduced  $Q_0(2.0$  K) to  $\sim 2 \times 10^{10}$ , nearly independent of field up to 80 mT.

As a next step, three 1.5 GHz single-cell cavities made of fine-grain and ingot Nb and with Nb flanges were heat treated at 1400 °C and 1600 °C and the measurements of  $Q_0(B_p)$  at 2.0 K did not show the extended  $Q_0$ -increase observed in the cavity with Ti45Nb flanges [4].

In this contribution we present the measurement results from four 1.47 GHz single-cell cavities with Ti45Nb flanges after heat-treatment at  $\geq 1250$  °C and results from analysis of the surface impurities by time-of-flight secondary ion mass spectrometry (TOF-SIMS) done on samples which had been heat treated with the cavities.

## CAVITIES TEST RESULTS

The following 1.47 GHz single-cell Nb cavities (original Cornell shape,  $B_p/E_{\text{acc}} = 4.43$  mT/(MV/m),  $E_p/E_{\text{acc}} = 1.78$ ) with Ti45Nb flanges were heat-treated and tested:

- Two Nb cavities from different ingot material supplied by CBMM, named G1G2 and H1H2.
- Two Nb cavities, one made of RRR $\sim 40$  material supplied by Cabot, one made of ingot Nb of RRR $\sim 120$  supplied by Tokyo-Denkai.

### Ingot Nb cavity G1G2

After the first 1400 °C HT resulting in the exceptionally high- $Q_0$  reported in [3], the cavity was subjected to a series of rinses with HF and oxypolishing, followed by electropolishing (EP). The results from the RF tests after these treatments were reported in [4]. After EP, the cavity was processed further as follows:

- 2<sup>nd</sup> HT at 1400 °C/3 h, standard degreasing with ultrasonic agitation and high-pressure water rinse (HPR).
- $\sim 1$   $\mu\text{m}$  BCP, followed by HPR.
- 3<sup>rd</sup> HT at 1400 °C/3 h, degreasing, HPR.
- $\sim 25$   $\mu\text{m}$  BCP (inside),  $\sim 20$   $\mu\text{m}$  BCP (outside), HPR.

It was noticed that the Nb susceptor and Nb stand in the induction furnace had become contaminated with Ti. For example, the top part of the Nb stand, closer to the cavity, had a yellowish colour which was determined to be titanium oxide, as measured by ToF-SIMS on a sample cut from the stand. We decided to reduce the HT temperatures below 1400 °C in order to reduce the Ti vapour pressure in the furnace.

The next sequence of cavity treatments was:

- 1250 °C/3 h HT, degreasing, HPR.
- $\sim 10$   $\mu\text{m}$  BCP, 1300 °C/3 h HT, degreasing, HPR.

RF tests in liquid He followed each preparation step listed above and the  $Q_0(B_p)$  curves measured at 2.0 K are shown in Fig. 1.

\*This manuscript has been authored by Jefferson Science Associates, LLC under U.S. DOE Contract No. DE-AC05-06OR23177. The U.S. Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce this manuscript for U.S. Government purposes.

<sup>#</sup>dhakal@jlab.org

Content from this work may be used under the terms of the CC BY 3.0 licence (© 2014). Any distribution of this work must maintain attribution to the author(s), title of the work, publisher, and DOI.

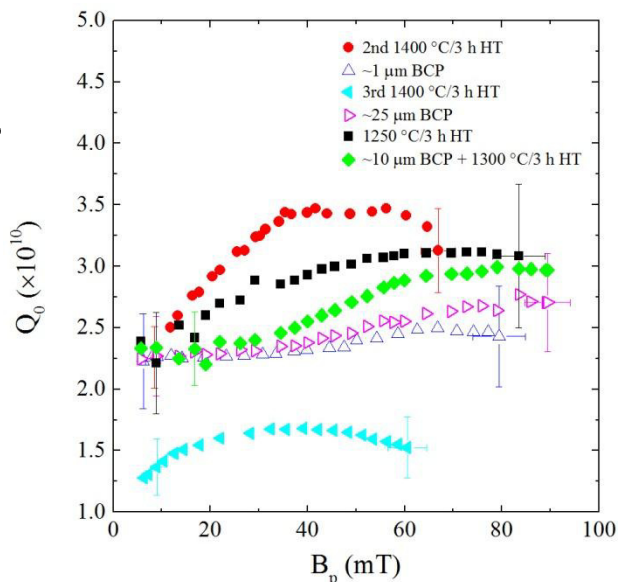


Figure 1:  $Q_0(2.0\text{ K})$  vs.  $B_p$  measured for 1.47 GHz single-cell G1G2 made of ingot Nb after several treatments listed chronologically from top to bottom in the legend. In all cases, the cavity was limited by quench, with no field emission.

### Ingot Nb cavity H1H2

The inner surface of the cavity was mechanically polished by centrifugal barrel polishing in three steps: 10 h with coarse media (ceramic angle-cut triangles), 16 h with medium media (ceramic with binder cones), 24 h with fine media (800 mesh alumina powder and wood blocks in water). The total material removal was estimated to be  $\sim 150\ \mu\text{m}$  from the change in resonant frequency. The cavity was then heat treated at  $800\ \text{°C}/2\ \text{h}$  in the production vacuum furnace to degas hydrogen, followed by the next treatments steps:

- $\sim 48\ \mu\text{m}$  BCP, HPR.
- $\sim 20\ \mu\text{m}$  EP, degreasing, HPR.
- $1400\ \text{°C}/30\ \text{min}$  HT, degreasing and HPR.
- $1400\ \text{°C}/3\ \text{h}$  HT, degreasing, HPR

The  $Q_0(B_p)$  curves measured at 2.0 K after each processing step are shown in Fig. 2.

### Fine-Grain Reactor Grade Nb Cavity

The cavity was built from reactor-grade (RRR $\sim 40$ ) Nb from Cabot, USA. After fabrication the cavity was etched by BCP removing  $\sim 100\ \mu\text{m}$  followed by hydrogen degassing at  $600\ \text{°C}/10\ \text{h}$  in the production vacuum furnace, additional  $\sim 50\ \mu\text{m}$  removal by BCP and HPR. The  $Q_0(2.0\ \text{K})$  was  $\sim 1.2 \times 10^{10}$ , nearly constant up to a quench field of  $\sim 89\ \text{mT}$ . The cavity was heat-treated at  $800\ \text{°C}/3\ \text{h}$  in the production vacuum furnace followed by  $20\text{-}30\ \mu\text{m}$  etching by BCP and HPR. The RF test at 2.0 K did not show any significant difference of  $Q_0(B_p)$  from the previous test.

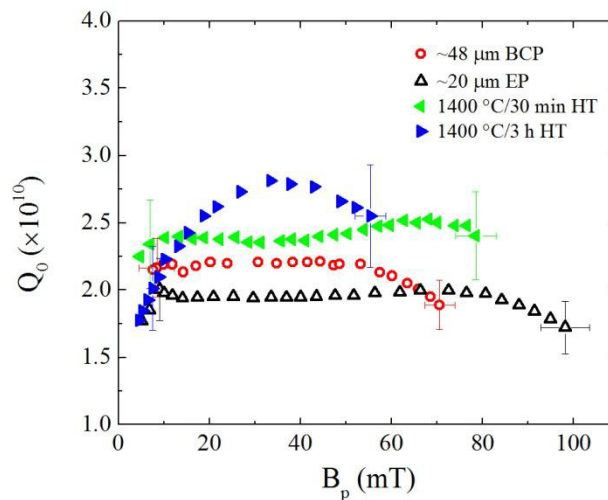


Figure 2:  $Q_0(2.0\text{ K})$  vs.  $B_p$  measured for 1.47 GHz single-cell H1H2 made of ingot Nb after several treatments listed chronologically from top to bottom in the legend. In all cases, the cavity was limited by quench, with no field emission.

The next processing steps were as follows:

- Post-purification in a Ti box at  $1250\ \text{°C}/3\ \text{h}$ ,  $\sim 60\ \mu\text{m}$  BCP, HPR.
- $1400\ \text{°C}/3\ \text{h}$  HT, degreasing and HPR.
- $\sim 20\ \mu\text{m}$  BCP, HPR
- $1250\ \text{°C}/3\ \text{h}$  HT, degreasing and HPR.

The  $Q_0(B_p)$  curves measured at 2.0 K after each processing step are shown in Fig. 3. The RRR of the cavity after post-purification is estimated to be  $\sim 100$  [5].

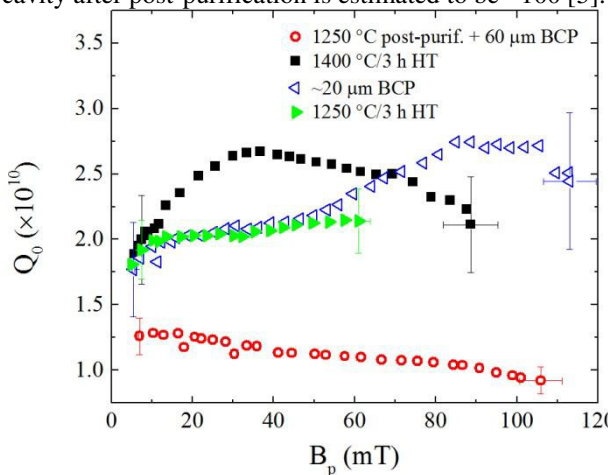


Figure 3:  $Q_0(2.0\text{ K})$  vs.  $B_p$  measured for a 1.47 GHz single-cell cavity made of fine-grain reactor-grade Nb after several treatments listed chronologically from top to bottom in the legend. The cavity was always limited by quench, with no field emission.

### Low RRR, Ingot Nb Cavity

The cavity was built from ingot Nb of RRR $\sim 120$  from Tokyo-Denkai, Japan. After fabrication, the cavity was etched by BCP, removing  $\sim 100\ \mu\text{m}$ , followed by hydrogen degassing at  $800\ \text{°C}/2\ \text{h}$  in the production vacuum furnace. The subsequent processing steps were:

- ~50  $\mu\text{m}$  BCP, HPR
- 120  $^{\circ}\text{C}/\sim 24$  h “in situ” bake.
- Post-purification in a Ti box at 1250  $^{\circ}\text{C}/3$  h, ~60  $\mu\text{m}$  BCP, HPR.
- 1400  $^{\circ}\text{C}/3$  h HT, degreasing and HPR.
- ~1  $\mu\text{m}$  BCP, HPR.

The  $Q_0(B_p)$  curves measured at 2.0 K after each processing step are shown in Fig. 4.

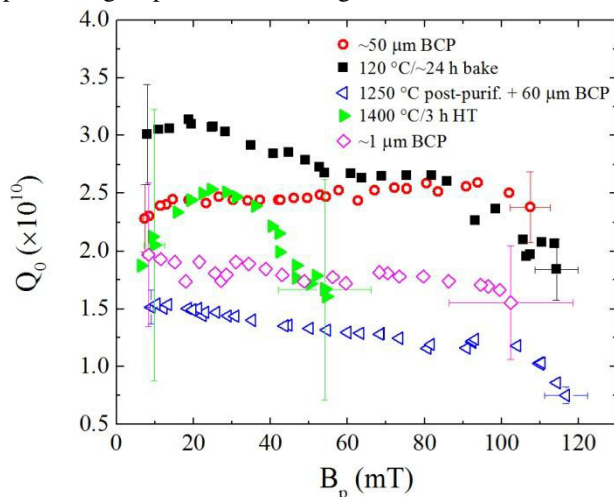


Figure 4:  $Q_0(2.0\text{ K})$  vs.  $B_p$  measured for a 1.47 GHz single-cell cavity made of medium-purity (RRR~120) ingot Nb after several treatments listed chronologically from top to bottom in the legend. The cavity was always limited by quench. Some multipacting was present above 90 mT in the test after 120  $^{\circ}\text{C}$  bake and some field emission was detected above 100 mT in the test after post-purification and BCP.

### SAMPLES TOF-SIMS ANALYSIS

Two Nb samples ( $5 \times 7 \times 3\text{ mm}^3$ ) were cut by wire-electrodischarge machining from a half-cell, etched to remove ~80  $\mu\text{m}$  by BCP and hydrogen degassed at 800  $^{\circ}\text{C}/2$  h. The surface of one side of the samples was nanopolished at Wah-Chang, USA. The samples were H-degassed once more at 800  $^{\circ}\text{C}/2$  h followed by ~20  $\mu\text{m}$  removal by BCP. The samples were then heat-treated at 1400  $^{\circ}\text{C}/3$  h, one with the second HT of cavity G1G2 (sample “B10”), the other one with cavity H1H2 (sample “B3”). The depth profiles of impurities such as H, N, C, O and Ti were measured, together with a non-heat-treated sample, by TOF-SIMS (TOF.SIMS 5, ION-TOF, Chestnut Ridge, NY) at North Carolina State University. The analysis chamber pressure is maintained below  $5.0 \times 10^{-9}$  mbar. For the depth profiling, 3 keV low energy  $\text{Cs}^+$  with 20 nA current was used to create a  $240\text{ }\mu\text{m} \times 240\text{ }\mu\text{m}$  area, and the middle  $50\text{ }\mu\text{m} \times 50\text{ }\mu\text{m}$  area was analysed using 0.2 pA  $\text{Bi}^{3+}$  primary ion beam.

The thickness of the oxide layer was ~4 nm for samples B3 and B10, compared to ~8 nm for the non-heat-treated one. The concentrations of C, O, and N were comparable

among all samples. The concentration of H was a factor of ~50 lower in the heat-treated samples. The concentration of Ti at a depth of ~20 nm is  $\sim 8 \times 10^{20}$  atoms/cm<sup>3</sup> and  $\sim 6 \times 10^{20}$  atoms/cm<sup>3</sup> in samples B3 and B10, respectively. These values are larger than  $\sim 3 \times 10^{20}$  atoms/cm<sup>3</sup> measured at the same time and with the same instrument, on a sample which was subjected to the first HT of cavity G1G2 at 1400  $^{\circ}\text{C}/3$  h [3]. Higher Ti concentration seems to correlate with lower quench field.

### CONCLUSION

Niobium cavities of different grain structure and purity have been heat-treated at high-temperature ( $\geq 1250\text{ }^{\circ}\text{C}$ ) without subsequent chemistry and tested at 2.0 K to investigate the reproducibility of high- $Q_0$ .  $Q_0$  increasing with field up to  $\sim 2.5 \times 10^{10}$  at ~60 mT has been typically obtained. The increase of  $Q_0$  extending to higher  $B_p$ -values than previously achieved could be an effect of broadening of the peaks in the electronic density of states at the gap edges [6]. Residual resistance values of ~1-2 n $\Omega$  were typically obtained after BCP or EP and ~2-4 n $\Omega$  after HT and will be discussed in more details in a future publication.

The HT method followed in this work proved it difficult to control the amount of Ti diffusing in the Nb, partly because of contamination of the Nb chamber in the induction furnace. However, it was established that doping of the penetration depth with Ti has a beneficial effect on  $Q_0$ . Future studies will rely on sublimation from a rod of pure Ti inserted inside the cavity which will be closed with Ta disks and heat-treated in the furnace.

### ACKNOWLEDGMENT

We would like to acknowledge J. Follkie, P. Kushnick, D. Forehand, B. Clemens, G. Slack, B. Martin, A. Palczewski and T. Harris of JLab for helping with EP, HPR, HT, CBP and cavity fabrication. We would also like to thank E. Zhou of the Analytical Instrumentation Facility at NCSU for the TOF-SIMS measurements.

### REFERENCES

- [1] P. Dhakal et al., Rev. Sci. Instr. 83, 065105 (2012).
- [2] P. Dhakal, G. Ciovati and G. R. Myneni, “A Path to Higher  $Q_0$  with Large Grain Nb Cavities,” IPAC’12, New Orleans, May 2012, p. 2426 (2012); <http://www.JACoW.org>
- [3] P. Dhakal et al., Phys. Rev. ST Accel. Beams 16, 042001 (2013).
- [4] P. Dhakal et al., “Analysis of Post-Wet-Chemistry Heat Treatment Effects on Nb SRF Surface Resistance,” TUIOC04, SRF’13, to be published.
- [5] P. Kneisel, J. Less-Common Metals 139, 179 (1988).
- [6] G. Ciovati, P. Dhakal and A. Gurevich, Appl. Phys. Lett. 104, 092601 (2014).