Pyrochlore Ingot Niobium SRF Technology for Next Generation Continuous Wave Accelerators

Ganapati Myneni

LBNL

June 3, 2013

Overview

✓Introduction

✓Fine grain - ingot niobium technologies

✓Cavity process optimization

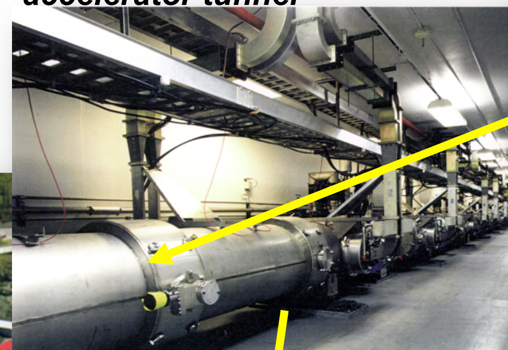
✓Accelerator cost and sustainable operation

✓Summary and outlook

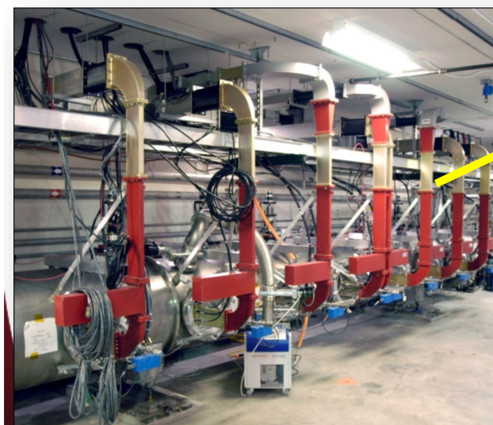
Jefferson Lab Accelerator Complex

Hall D (new construction)

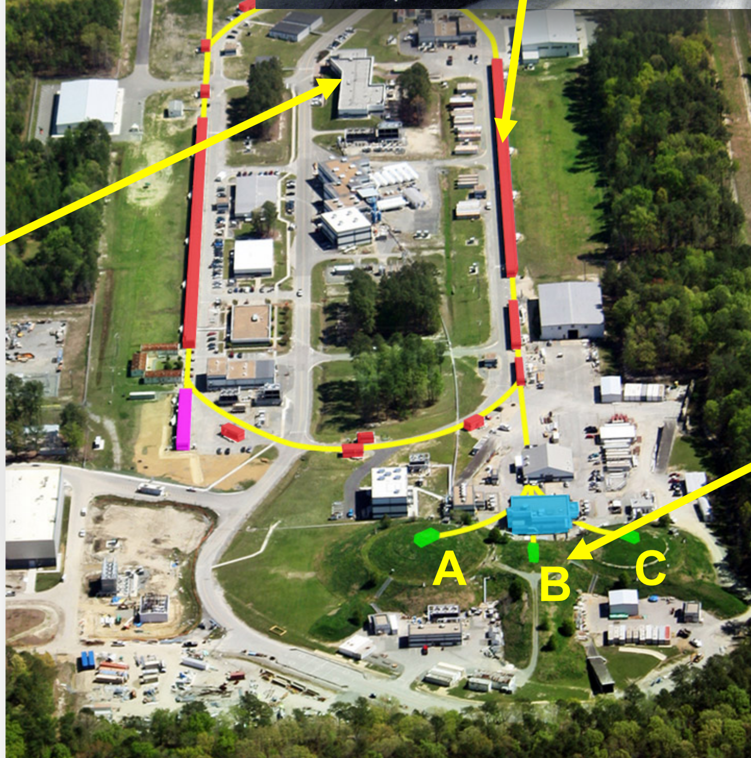
Cryomodules in the accelerator tunnel

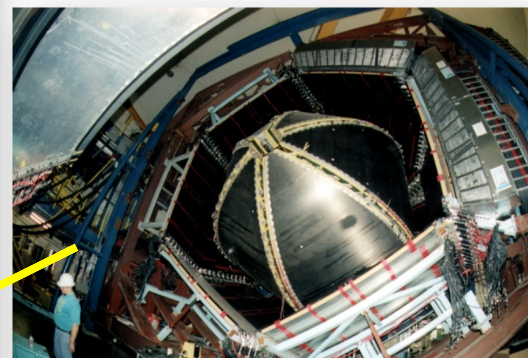


Superconducting radiofrequency (SRF) cavities



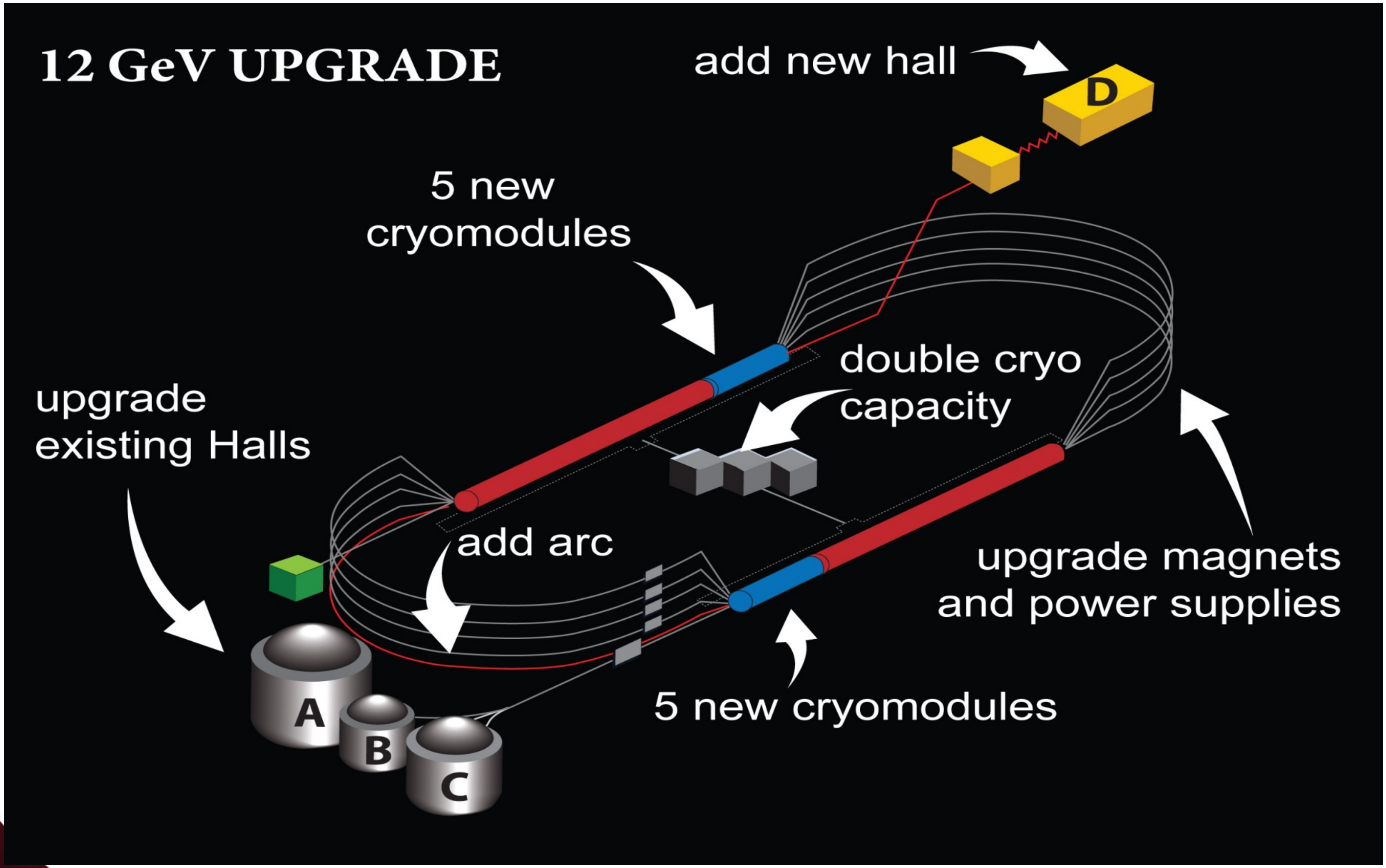
Free Electron Laser (FEL)





CEBAF Large Acceptance Spectrometer (CLAS) in Hall B

CEBAF energy upgrade



Introduction

- Bulk RRR – Residual Resistance Ratio ~ $R_{300}/R_{4.2}$

  Has no influence on superconducting rf properties (thermal stabilization)

- Important Interstitials H, C, N and O that contribute to RRR significantly
  - and tantalum, substitutional impurity does not significantly contribute to RRR
- Quality Factor $Q_0 = G/R_s$, where G is the geometry factor and it is independent of the cavity frequency & $R_s = R_0 + R_{BCS}(f,T,l_{en})$, ideal ~ 2×10^{11} @ 2 K
- Figure of merit of Nb ~ $Q_0 * E_{quench}$

Historical Example of Ingot Niobium

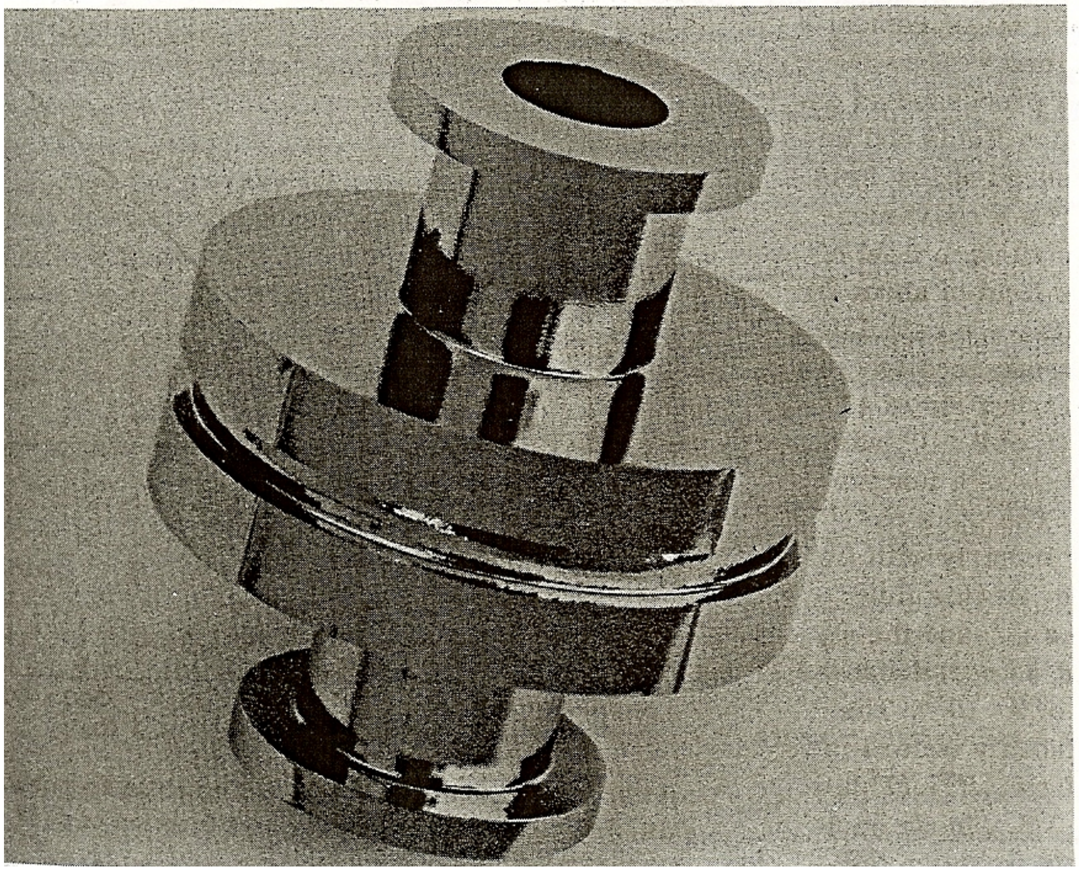


FIG. 1. An electron-beam welded TM_{010} mode Nb cavity. The cavity is resonant at 8.6 GHz and is 3.6 cm in over-all length.

H_{pk}~ 108 mT with BCP

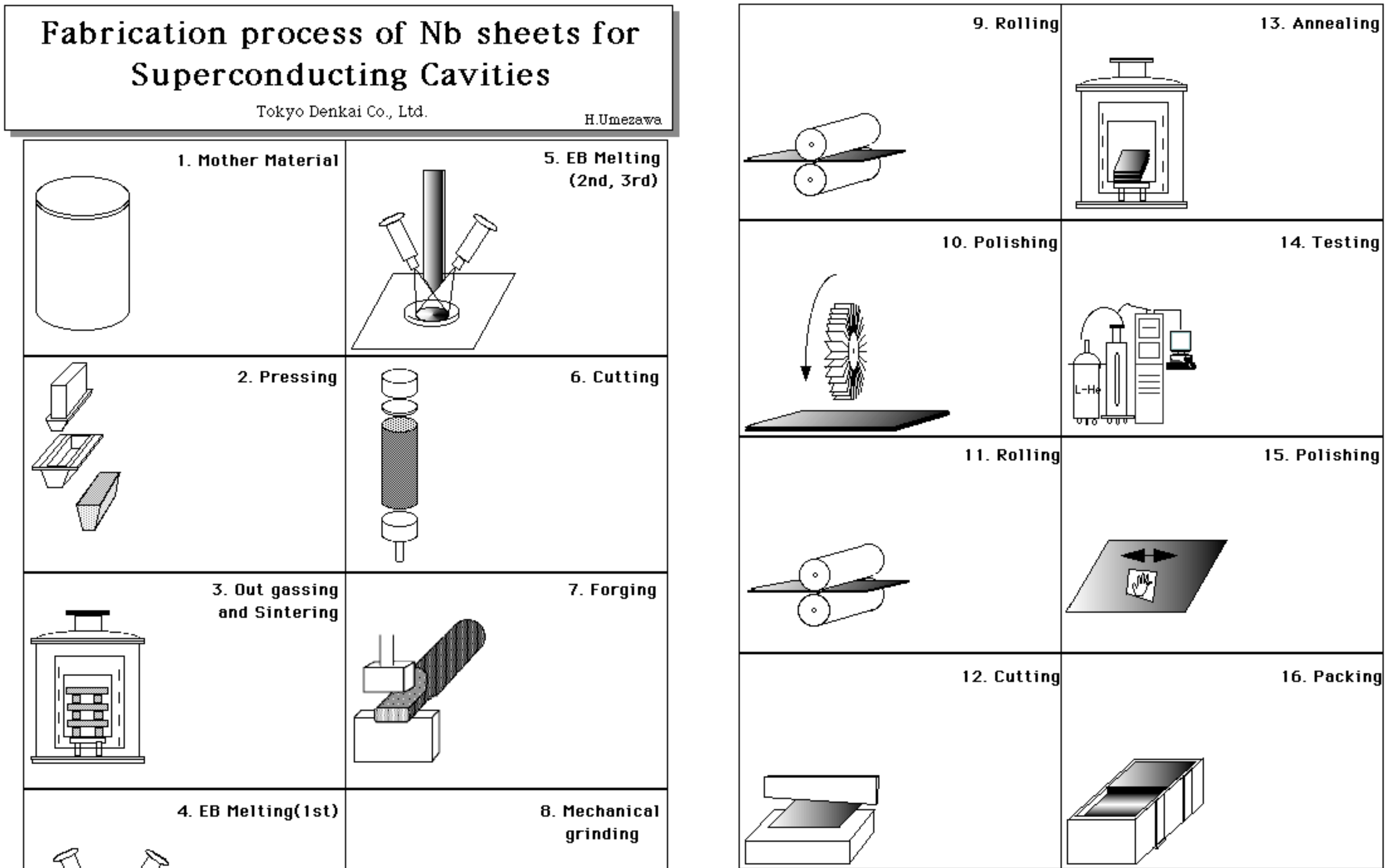
Stanford solid niobium cavity 1970

Niobium Specifications

- **Polycrystalline Niobium with ASTM #5 Grain Size or finer ~ 50 micro meters & fully recrystallized**
- **Percentage of elongation > 25**
- **Yield Strength > 10.7 KSI (~75 MPa) (~40 Mpa)**
- **RRR > 250 (>300)**
- **Tantalum < 1000 wt ppm (<500 wt ppm)**

Original CEBAF was built with 90% pyrochlore CBMM niobium

Process steps - fine grain Niobium



Note:

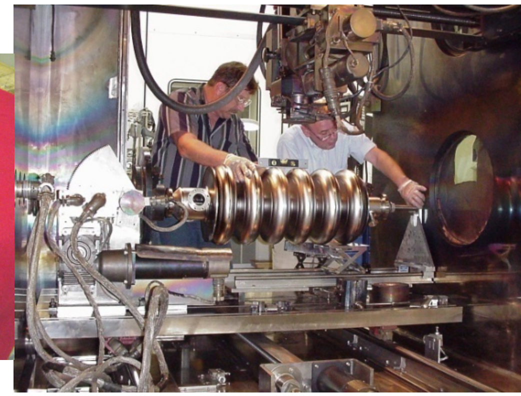
During this process foreign materials can be embedded so QA is required

Multi cell cavity fabrication

Forming

Machining

Welding



Tuning

~90% of CEBAF cavities were made with CBMM Pyrochlore ore based niobium
In comparison to present day use of Tantalite/Columbite ore based niobium

Extrinsic contamination of cavity dominated until after CEBAF construction

- Surface contamination
    - Molecular and particulate

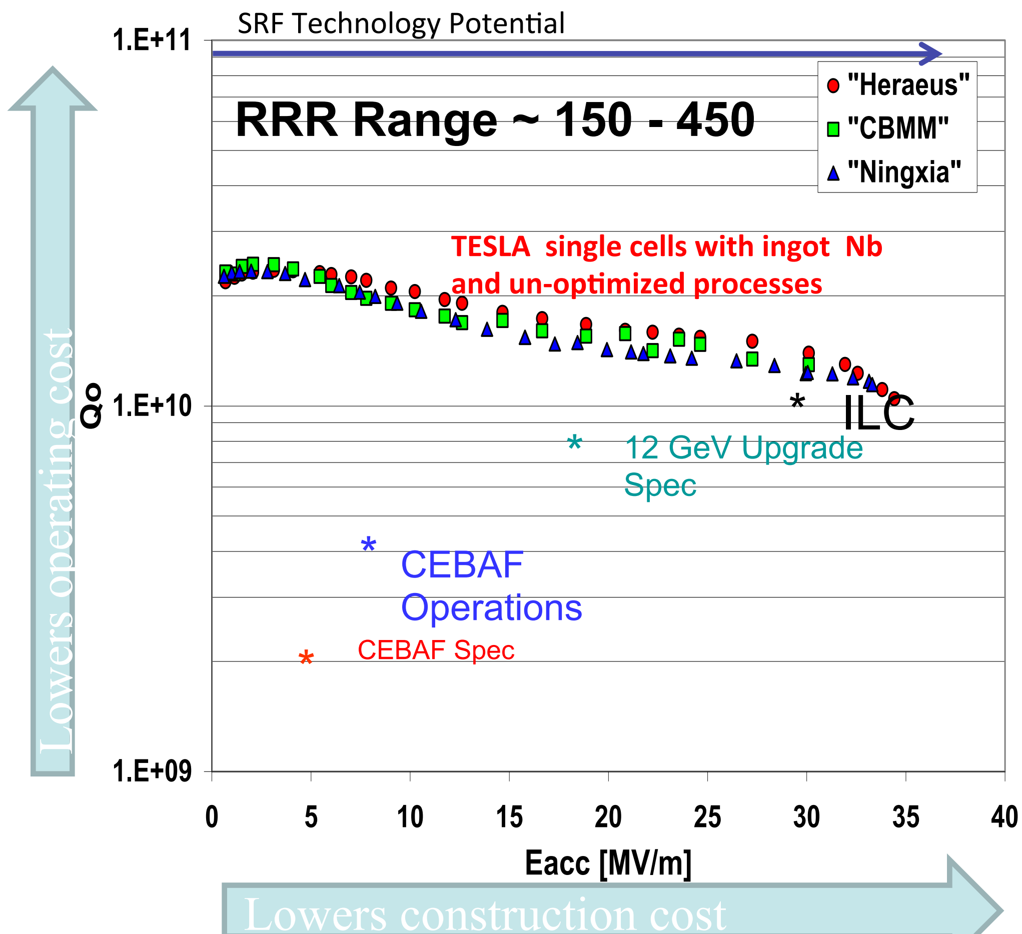
Vacuum Contamination Work Shop at JLab 1997

Minimizing organic and particulate recontamination addressed, several courses were held in subsequent years

Cavity processing steps

- Buffer chemical polishing (BCP) ~ 150 micro meters
- Electro polishing (EP) ~ 50 micro meters
- High pressure ultra pure water rinse
- ~ 600 – 900 °C heat treatment
- Light EP
- High pressure ultra pure water rinse
- Vacuum bake ~120 °C for up to 48 hours
- RF test

Niobium cavity – performance (CW)



RRR, type of niobium and vendor has no influence on cavity performance

Development of Large Grain/Single Crystal Niobium Cavity Technology at Jefferson Lab
P. Kneisel, G. Myneni, G. Ciovati, T. Carneiro Proc of Linac 2004

Niobium For SRF Cavities

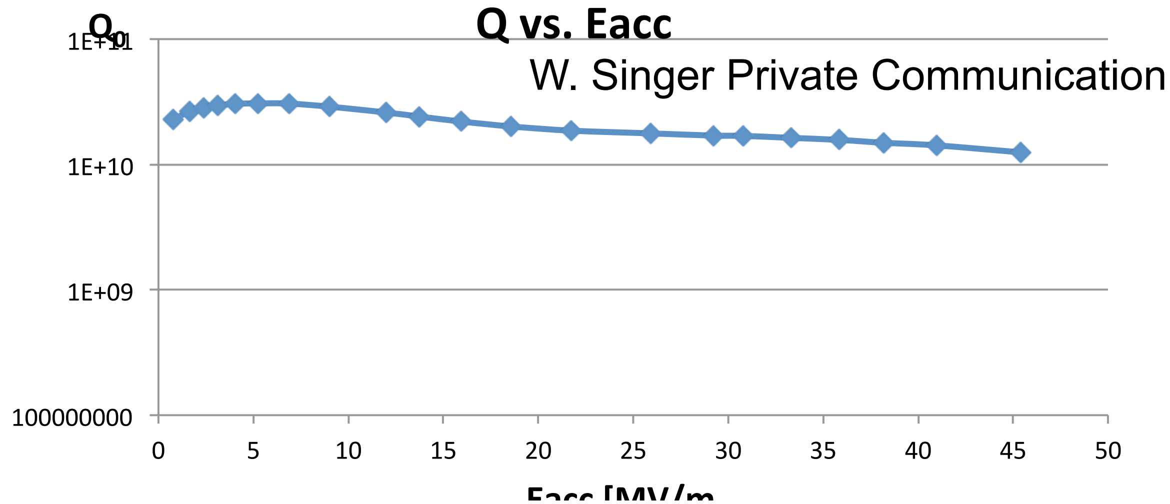
- **At present, Niobium for SRF cavities comes from Columbite/Tantalite ore**
  - **Niobium is present as "impurity"**
  - **Niobium is produced as a by-product of Tantalum**

- **Primary reason – the Tantalum content is lower**
  - **Tantalum is generally believed to negatively impact SRF properties of Niobium but there is no real data to back it up**
  - **JLab data shows reducing Tantalum content below 1000 ppm has no advantage for Superconducting RF cavities**
  - **Low Tantalum niobium is relatively expensive**

Ingot Niobium Technology
CBMM-JLab CRADA, August 2004

CBMM/JLab jointly applied for US Patent in April 2005 and the patent 8128765 B2 was granted on March 6, 2012. G. Myneni, P. Kneisel and T. Carneiro (CBMM) are the inventors

Many international institutions in Asia, Europe and America are developing ingot niobium cavities. DESY, KEK are among them., AES, Niowave and RI industries are involved.CBMM, Heraues, Ningxia, Tokyo Denkai and Wah Chang are able to provide ingot niobium



DESY sets E_{acc} world record with ingot niobium 9 cell TESLA cavities

Araxá Mine in Brazil & Pyrochlore Niobium
Original CEBAF 90% CBMM Nb

The CBMM open cast mine

Conveyor belt bringing the ore to concentration plant

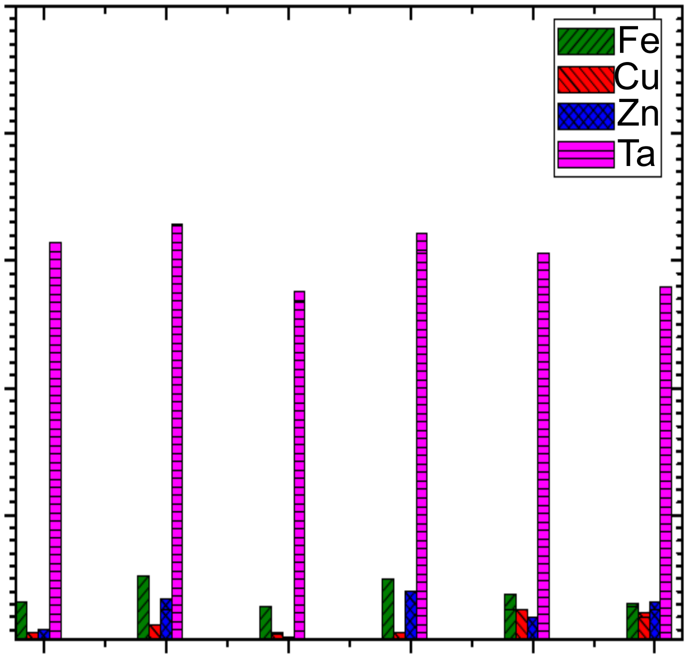
Electron beam furnace for the refinement of Niobium metal, producing 210 tons per annum

Finished Nb ingot from the Pyrochlore ore

Tantalum is an impurity and the amount depends on the location of the ore and can be up to ~ 1500 wt ppm and is uniformly distributed in niobium

Simplified niobium spec: ingot slices with Hv ~ 50

Uniform distribution of Ta has no effect on SC Parameters, BCP has large effect



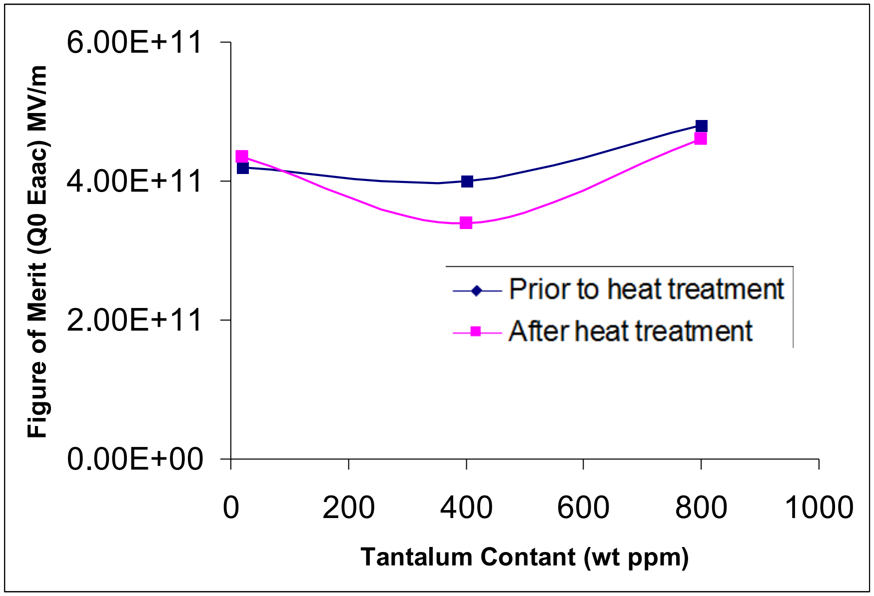
Sample	Ta-Content (ppm)	T_C at 100 Oe (K)	H_P (Oe)	H_{C2} (Oe)	H'_{C2} (Oe)
Technical-Niobium-1P	1339±36	9.18	1700	4150	7500
Technical-Niobium-2P	800 ±80	9.18	1600	4125	7500
Technical-Niobium-3P	243±10	9.2	1600	4090	7500
Technical-Niobium-1CT	1285±35	9.05	1150	3930	-
Technical-Niobium-2CT	684±54	9.05	1290	3735	-
Technical-Niobium-3CT	149±11	9.06	1350	3820	-

S. B. Roy et al Supercond. Sci. Technol. 25 (2012) 115020

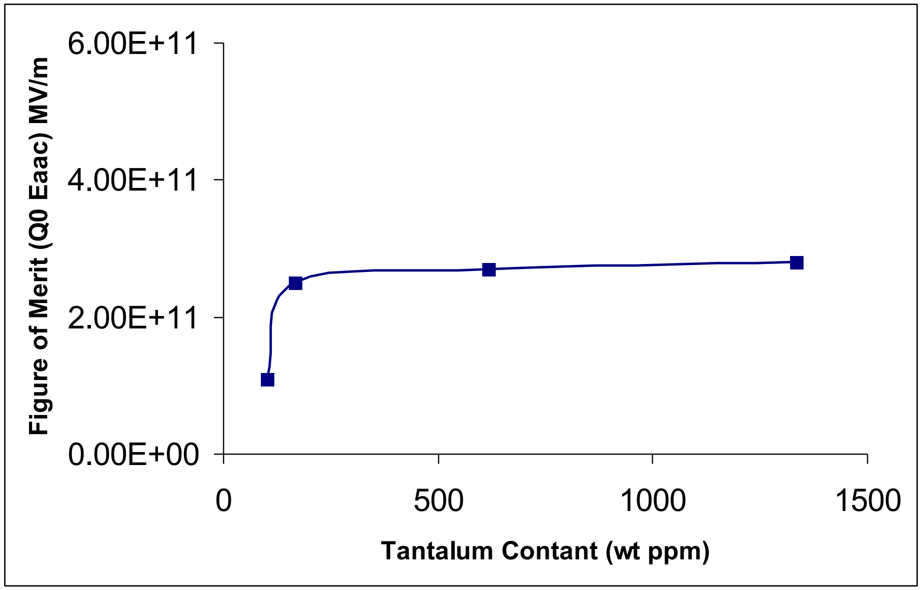
Ingot niobium has superior performance

Tantalum does not affect the performance

Large grain ingot niobium



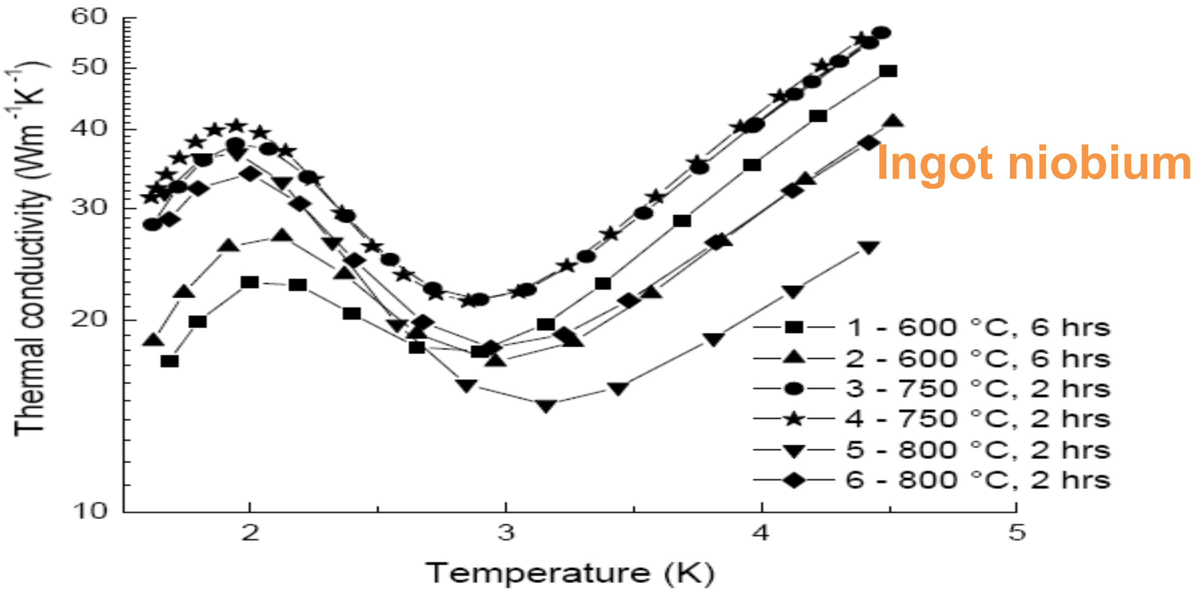
Polycrystalline niobium



Empirically FOM ~ 1.0 x10^{13}

Influence of Ta content in high purity niobium on cavity performance
P. Kneisel, G. Myneni, G. Ciovati, D. Proch, W. Singer, T. Carneiro et al in Proceedings of PAC 2005

Tantalum and RRR have minimal influence on phonon peak



Specimen	Estimated RRR	Tantalum content (ppm) [3]	Heat Treatment	Titanium getter
1	191	1275	600 °C, 6 hrs	No
2	131	668	600 °C, 6 hrs	No
3	190	756	750 °C, 2 hrs	Yes
4	196	756	750 °C, 2 hrs	Yes
5	104	1322	800 °C, 2 hrs	No
6	143	523	800 °C, 2 hrs	No

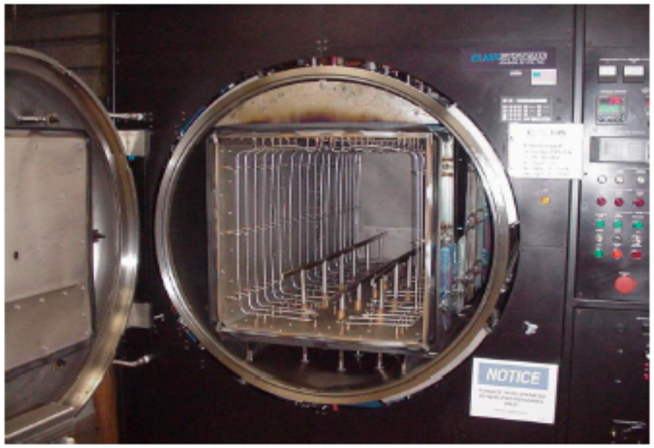
Saravan
PhD student at MSU

Hydrogen absorption with BCP and EP

- **Very high equilibrium hydrogen activities (fugacity) have been estimated when Nb metal is in contact with water or BCP solution**
- **Hydrogen is readily absorbed into Nb when the protective oxide layer is removed**
- **Lower H fugacity's are obtained due to an anodic polarization of Nb during EP and hence lower hydrogen absorption**

R.E. Ricker, G. R. Myneni, J. Res. Natl. Inst. Stand. Technol. 115, 353-371 (2010)

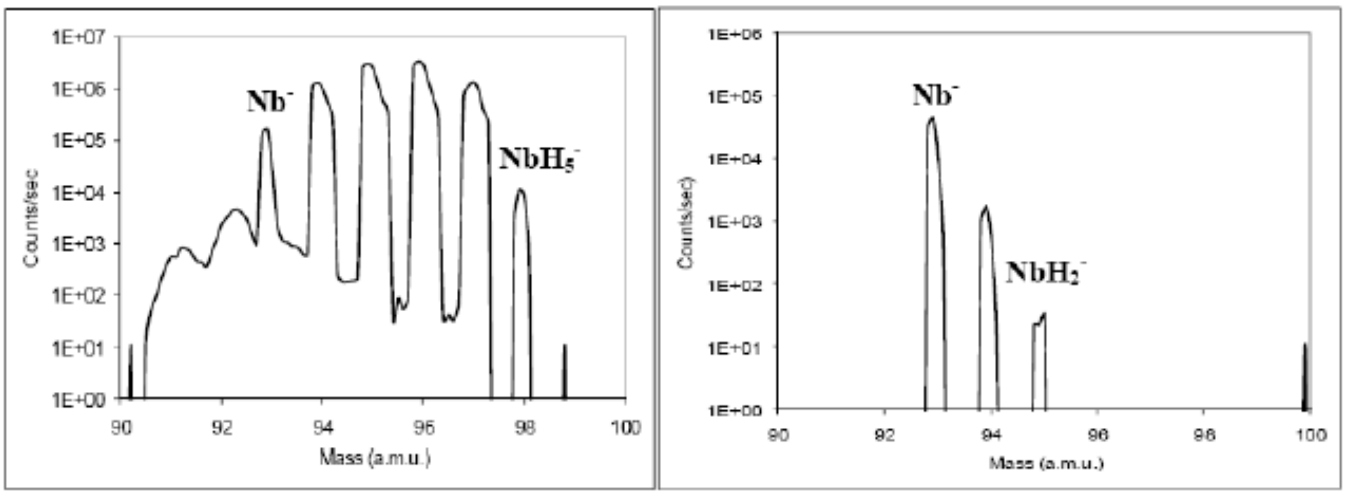
Heat treatment to remove hydrogen



- 800°C/3h, pressure ~ 10^{-6} mbar
- **No chemical etching afterwards!**
- Nb samples were treated with the cavities and depth profiling of the impurities was done at NCSU

~ 2 orders of magnitude lower hydrogen content after HT

Currently used furnaces contaminate the cavity surfaces, chemical re-etching reintroduces H



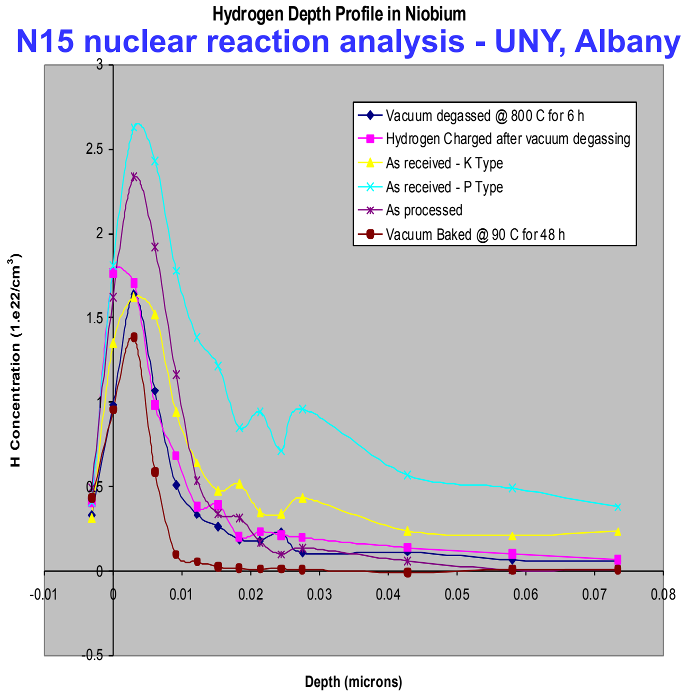
(a) (b)

FIGURE 1. SIMS mass spectra showing difference in H between (a) non-heat treated and (b) heat treated sample.

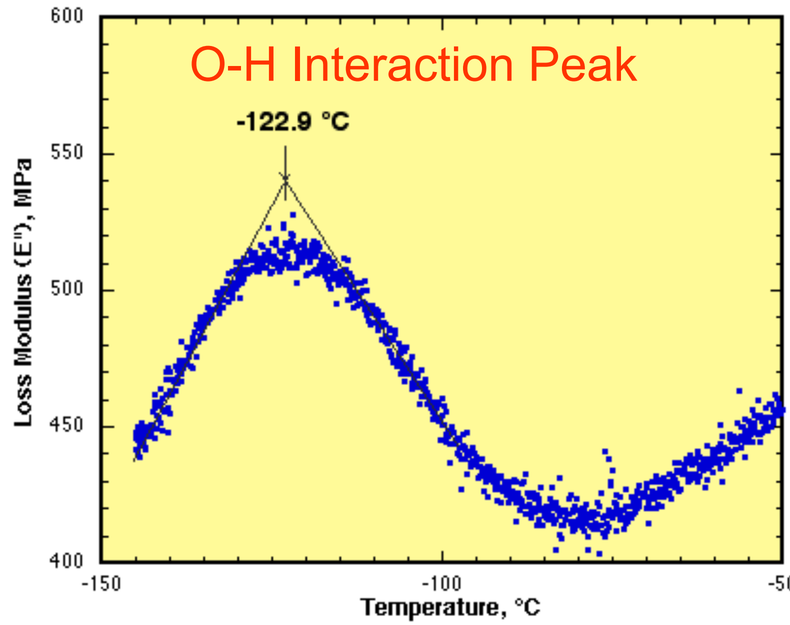
G. Ciovati, G. Myneni, F. Stevie, P. Maheshwari, and D. Griffis, Phys. Rev. ST Accel. Beams 13, 022002 (2010).

Hydrogen Interactions

N15 nuclear reaction analysis - UNY, Albany

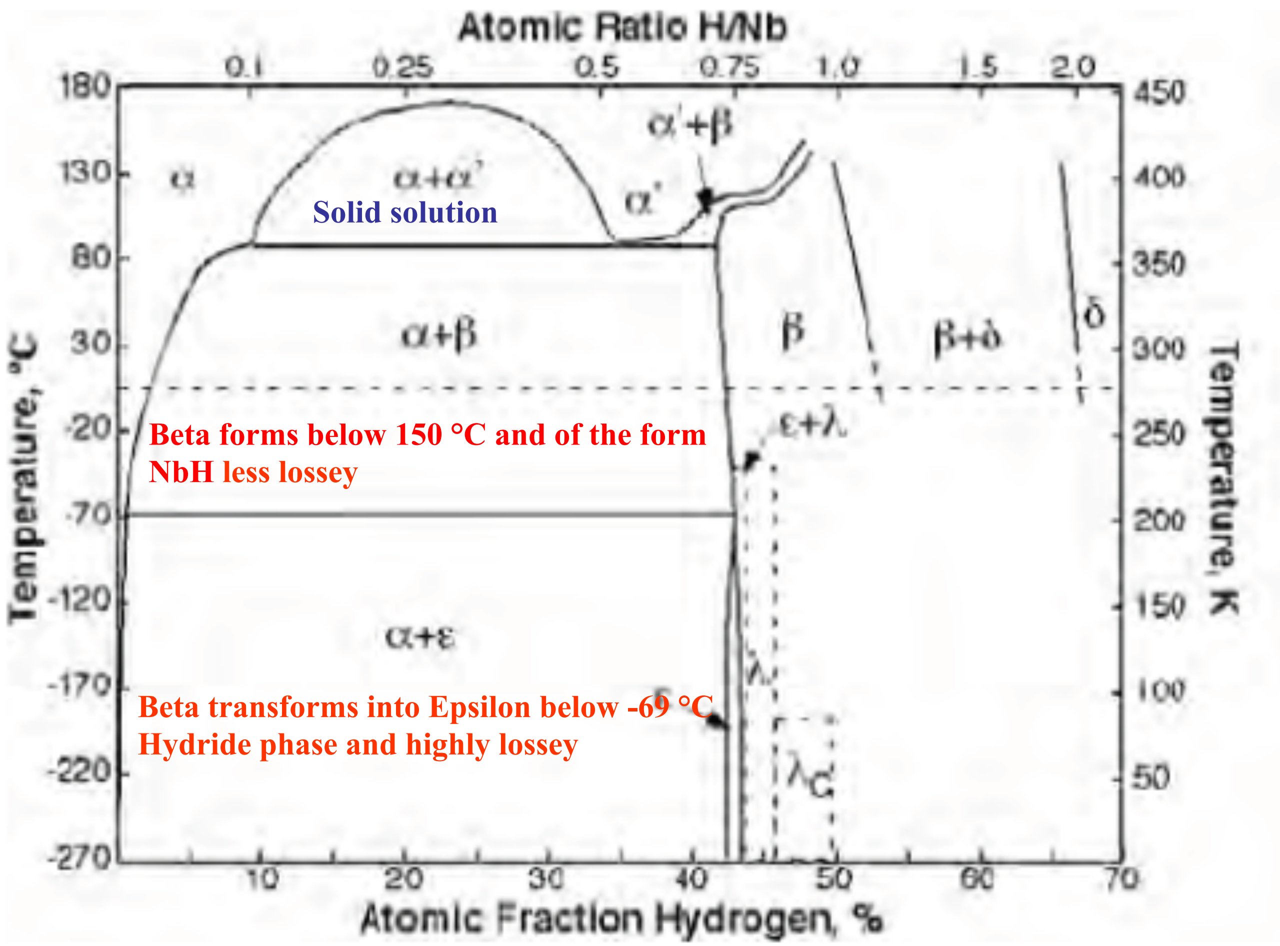


Internal friction data - NIST, Gaithersburg

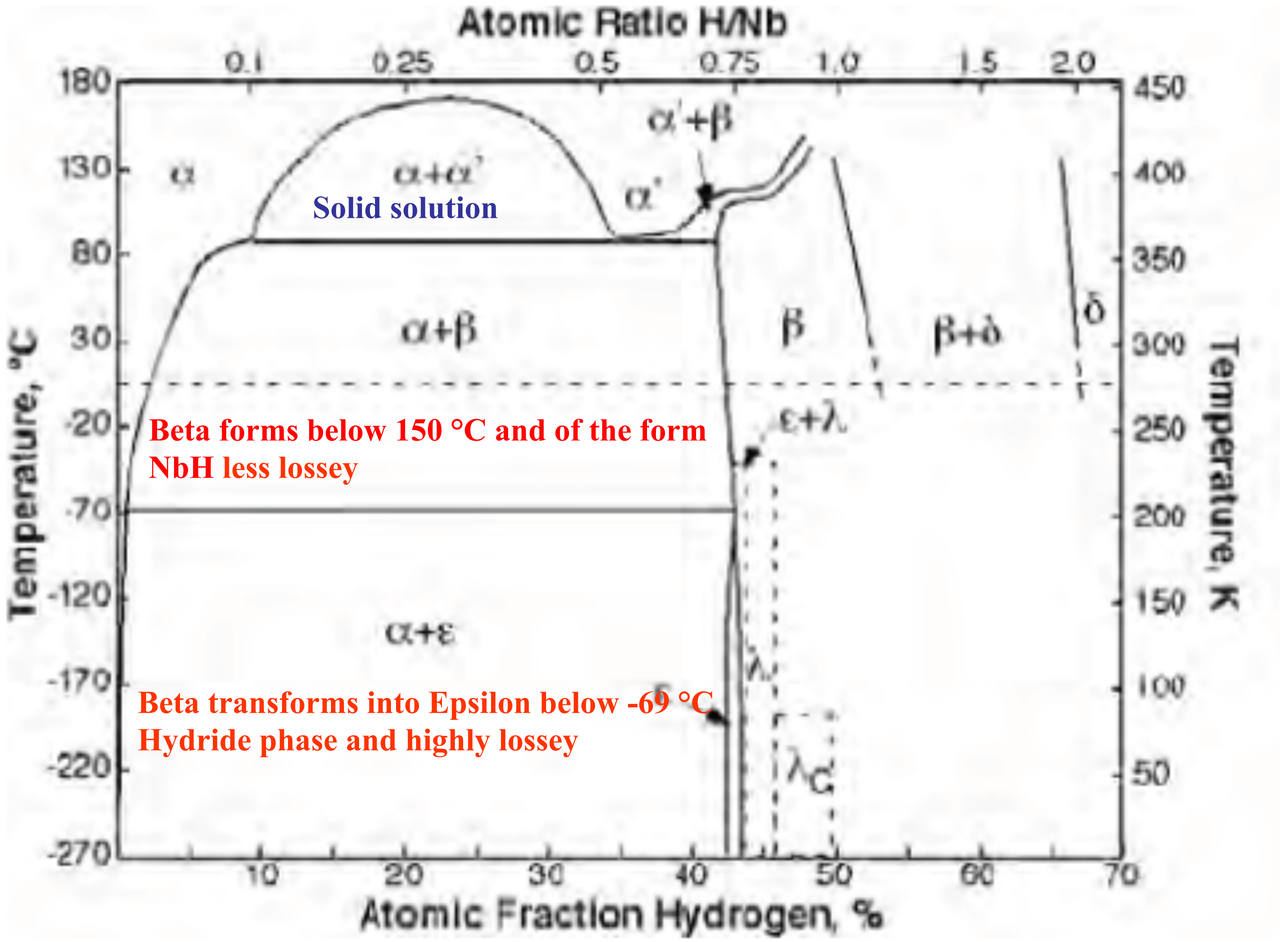


Hydrogen is most mobile at room temperature, interacts with defects, interstitials and is influenced by residual stress concentration gradients and affects the magnetic properties

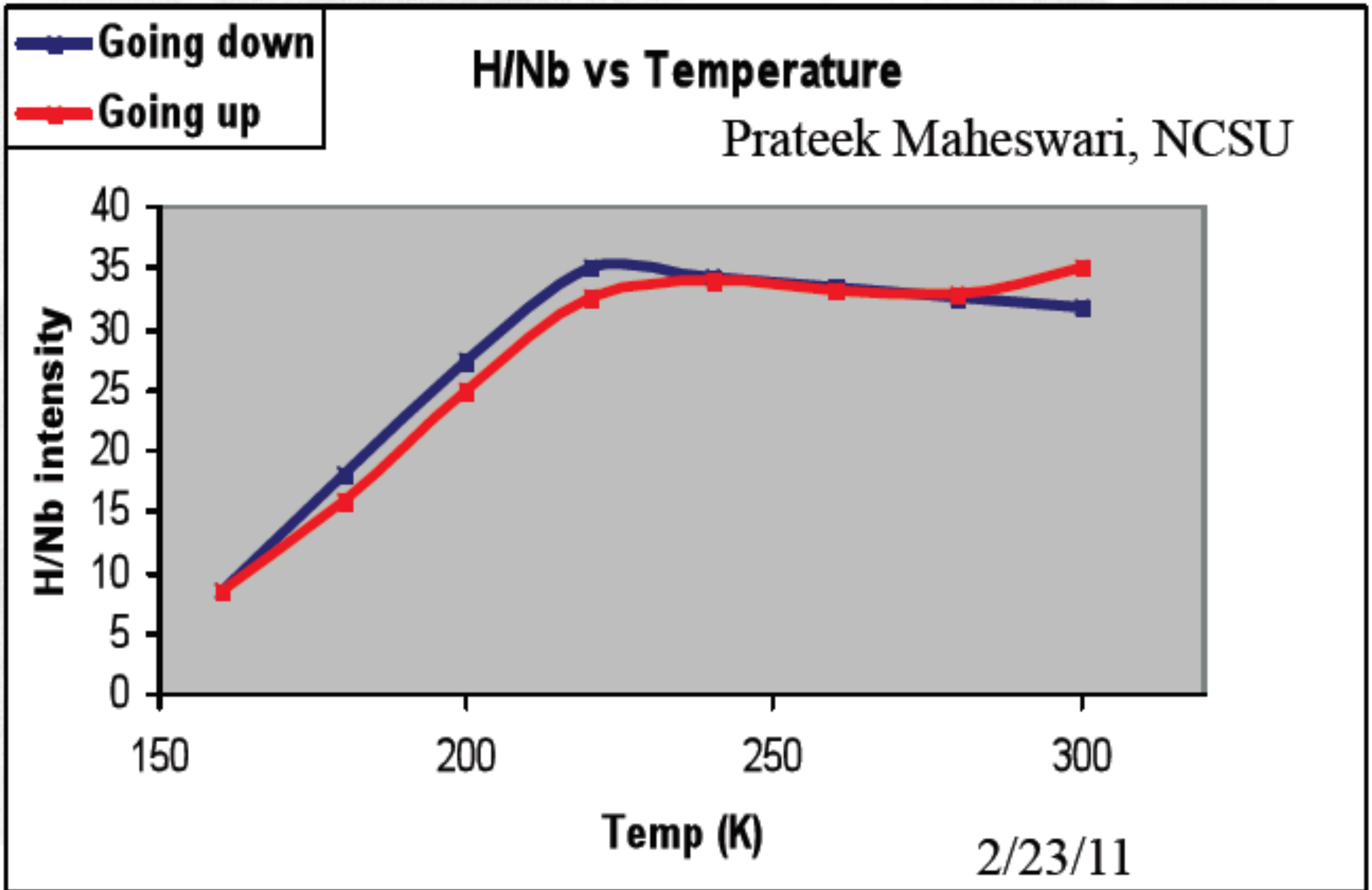
Niobium – hydrogen phase diagram



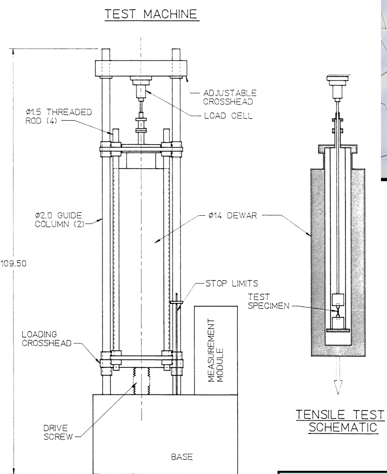
Niobium – hydrogen phase diagram

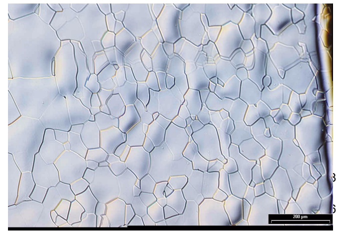


Hydrogen phase change

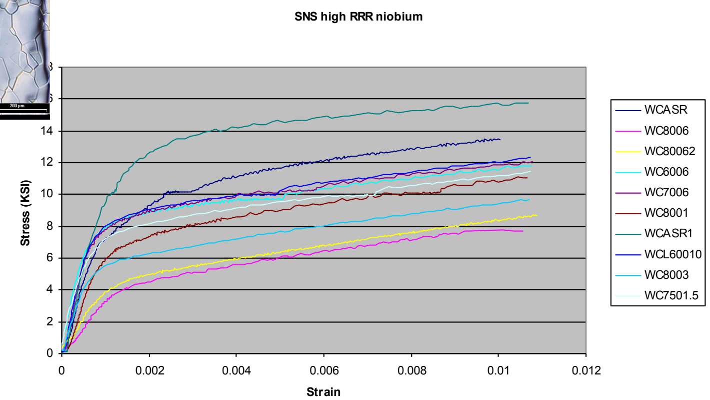


Microyielding Issue

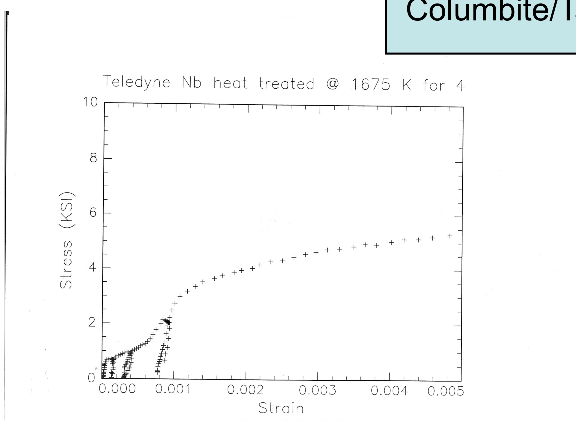


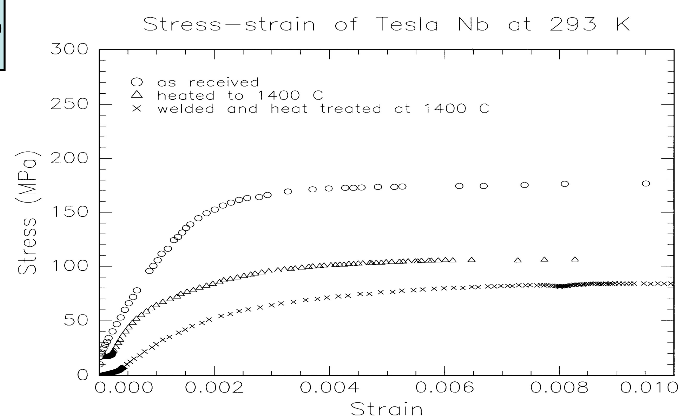


Ta ~ 200 wt ppm



Columbite/Tantlite Nb

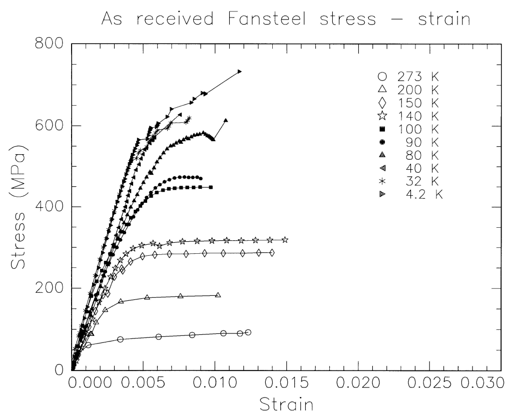


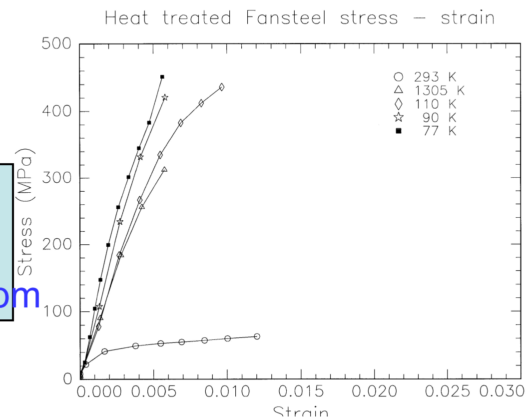


Heat Treatments and Welds Lead to Micro Yielding

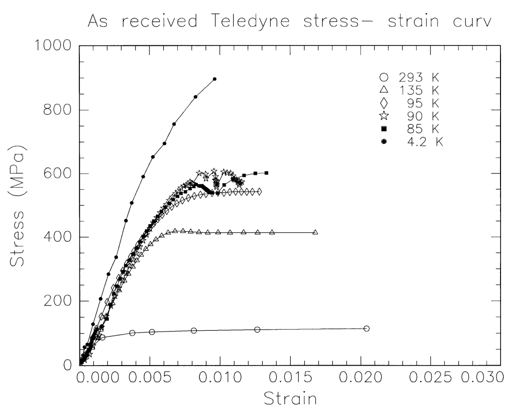
Jefferson Lab

Pyrochlore Vs Columbite Nb

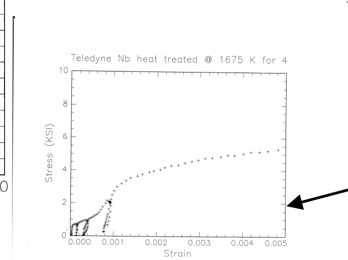


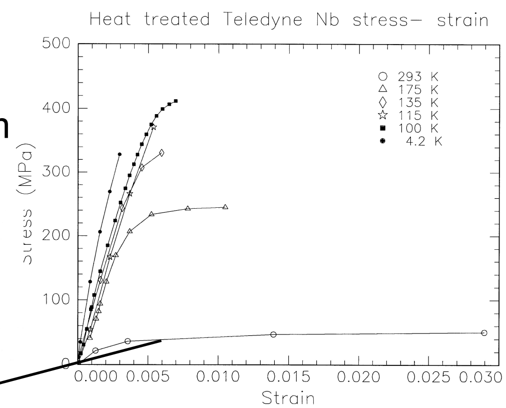


Pyrochlore
CBMM
Ta < 1000 wt ppm

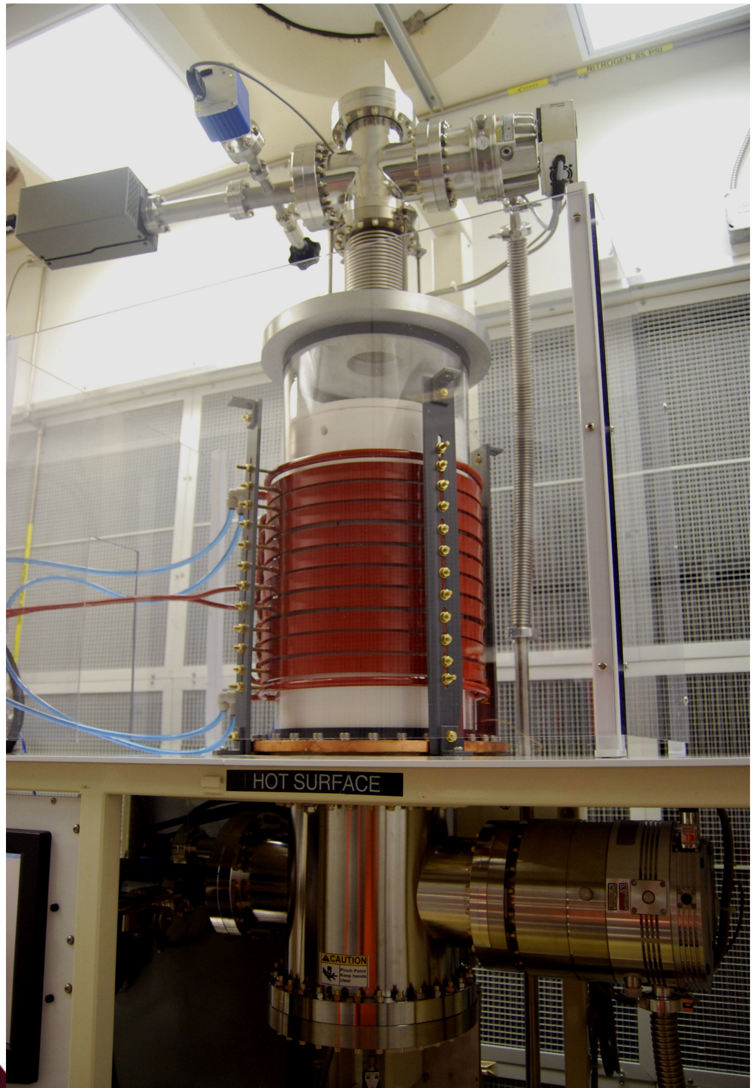


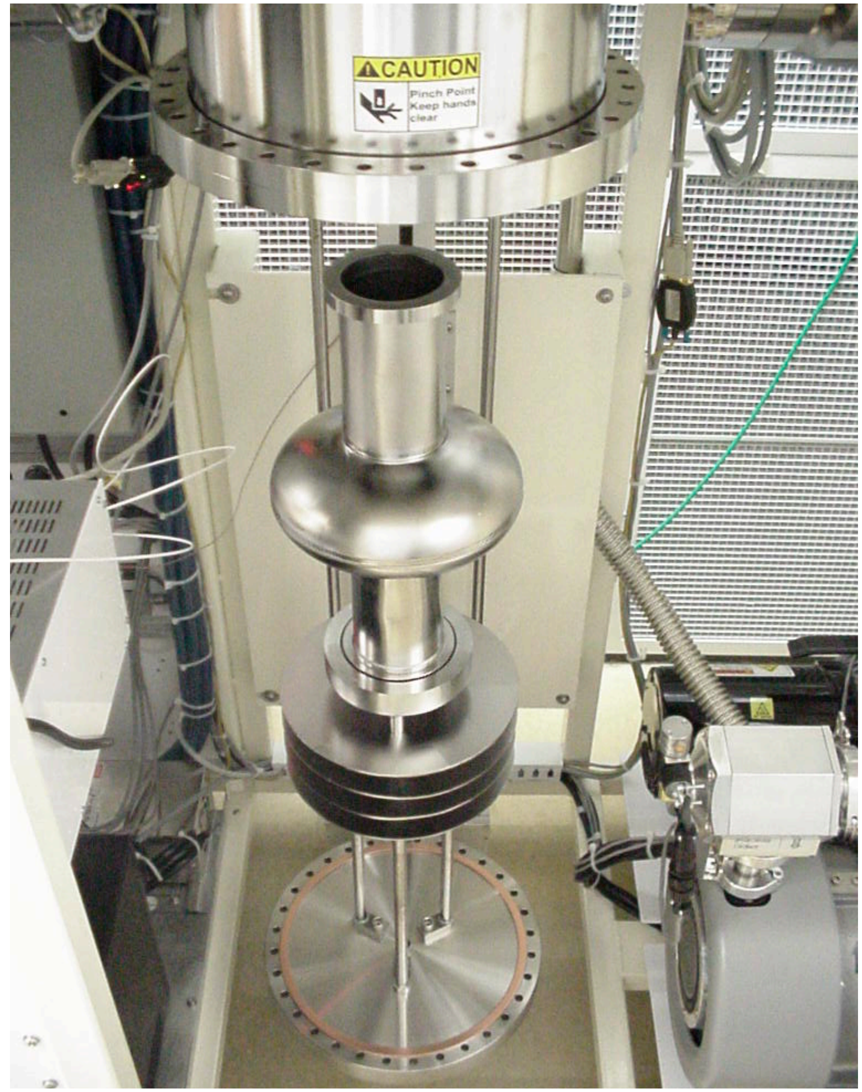
Columbite
Ta ~ 200 wt ppm





Clean UHV furnace - patents applied for



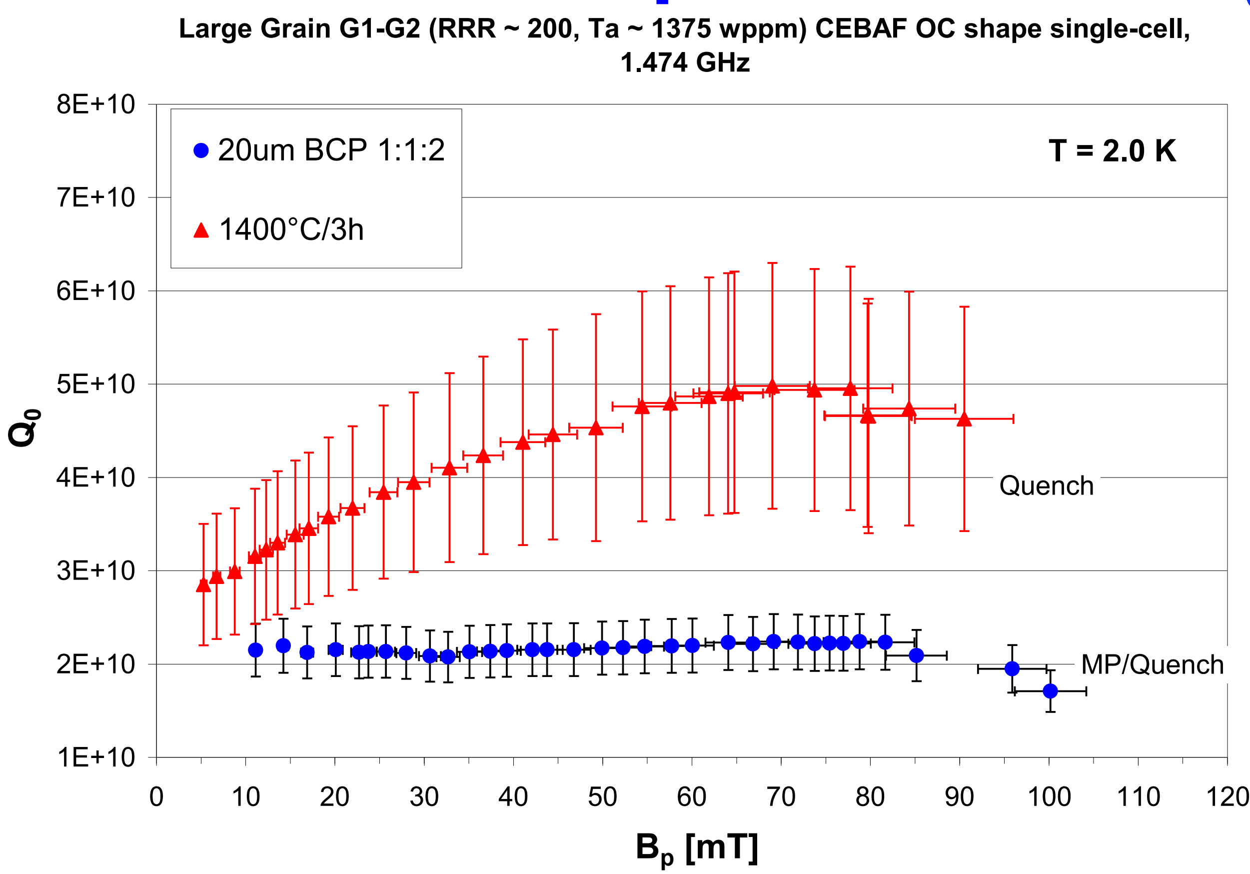


Design and Performance of a new Induction Furnace for Heat Treatment of Superconducting Radiofrequency Niobium Cavities, P. Dhakal, G. Ciovati, W. Rigby, J. Wallace, G. Myneni
Review of Scientific Instruments 83, 065105 (2012)

Cavity material and preparation

- CBMM ingot niobium, RRR ~ 200 (>350), Ta ~ 1350 (<500) wt ppm, inexpensive 50% to 60% less than conventional Nb
- Barrel polishing 73µm, BCP 65µm , a total of 138 µm removal and high pressure UHP water (~250 µm for fine grain)

RF field dependence of Q_0



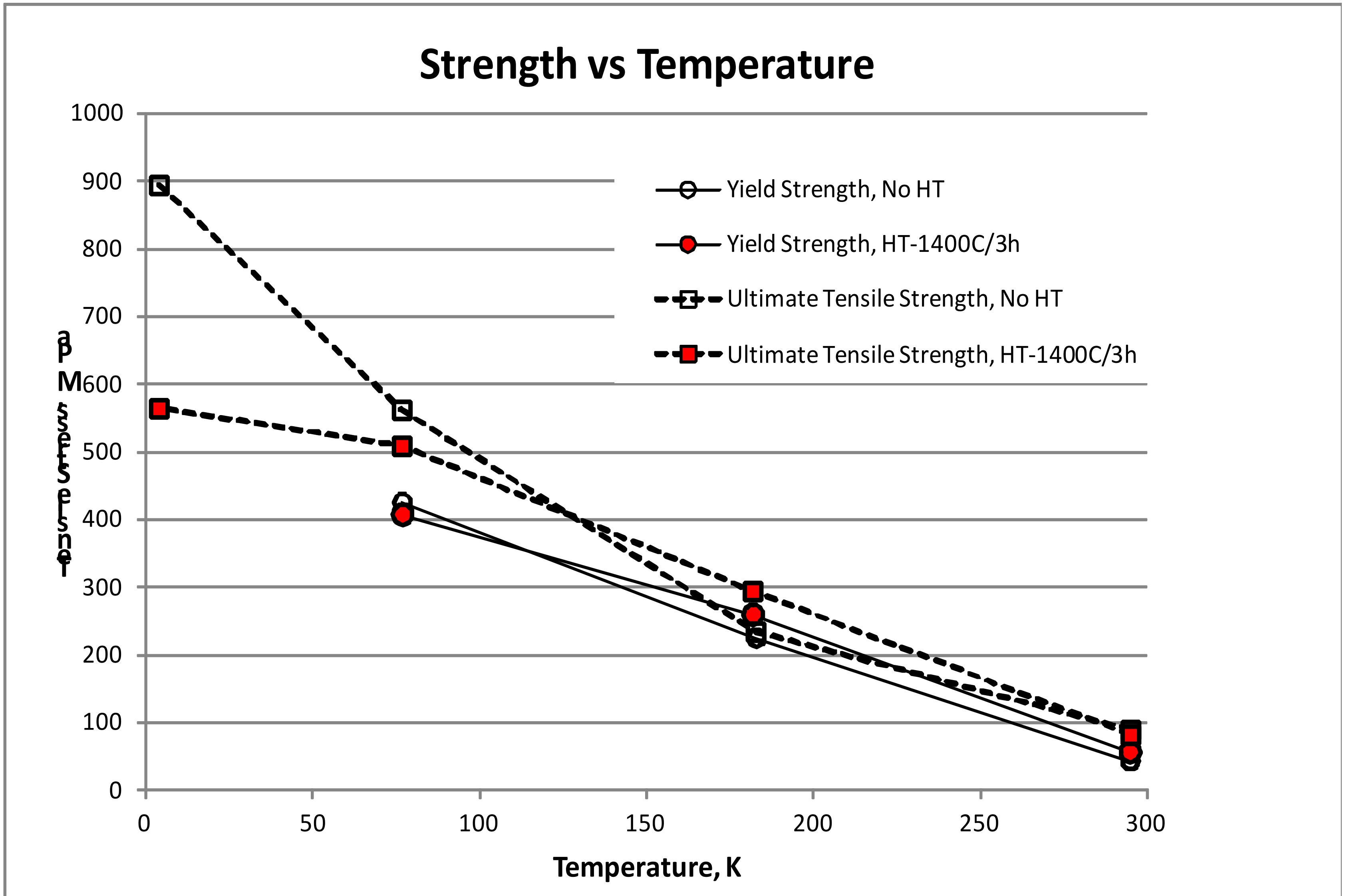
- **Low-field Q-increase extending up to ~70 mT**
- **Q_0 is ~4× higher than the average of CEBAF Upgrade cavities**
- **Q_0(1.5 K) ~ 2×10^{11}**

Dhakal, Ciovati and Myneni Proceedings of IPAC 2012

Intrinsic contamination of Nb & proton-dislocation interaction appear to determine the performance of the cavities

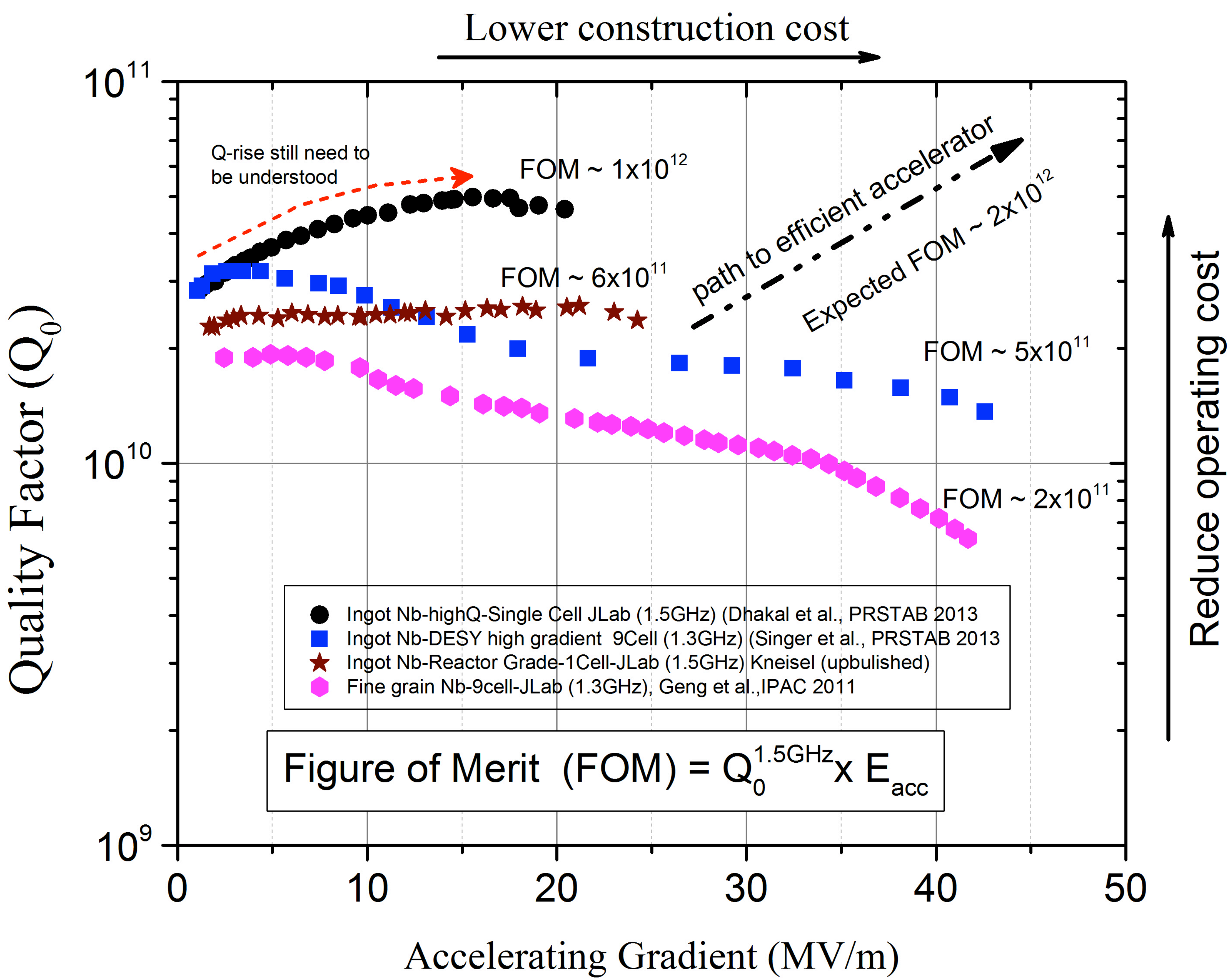
- **Niobium is a prolific hydrogen absorber in the absence of the natural surface oxide**
  - **Hydride formation**
  - **Dislocations-proton interaction**

Improved mechanical properties with 1400 C HT



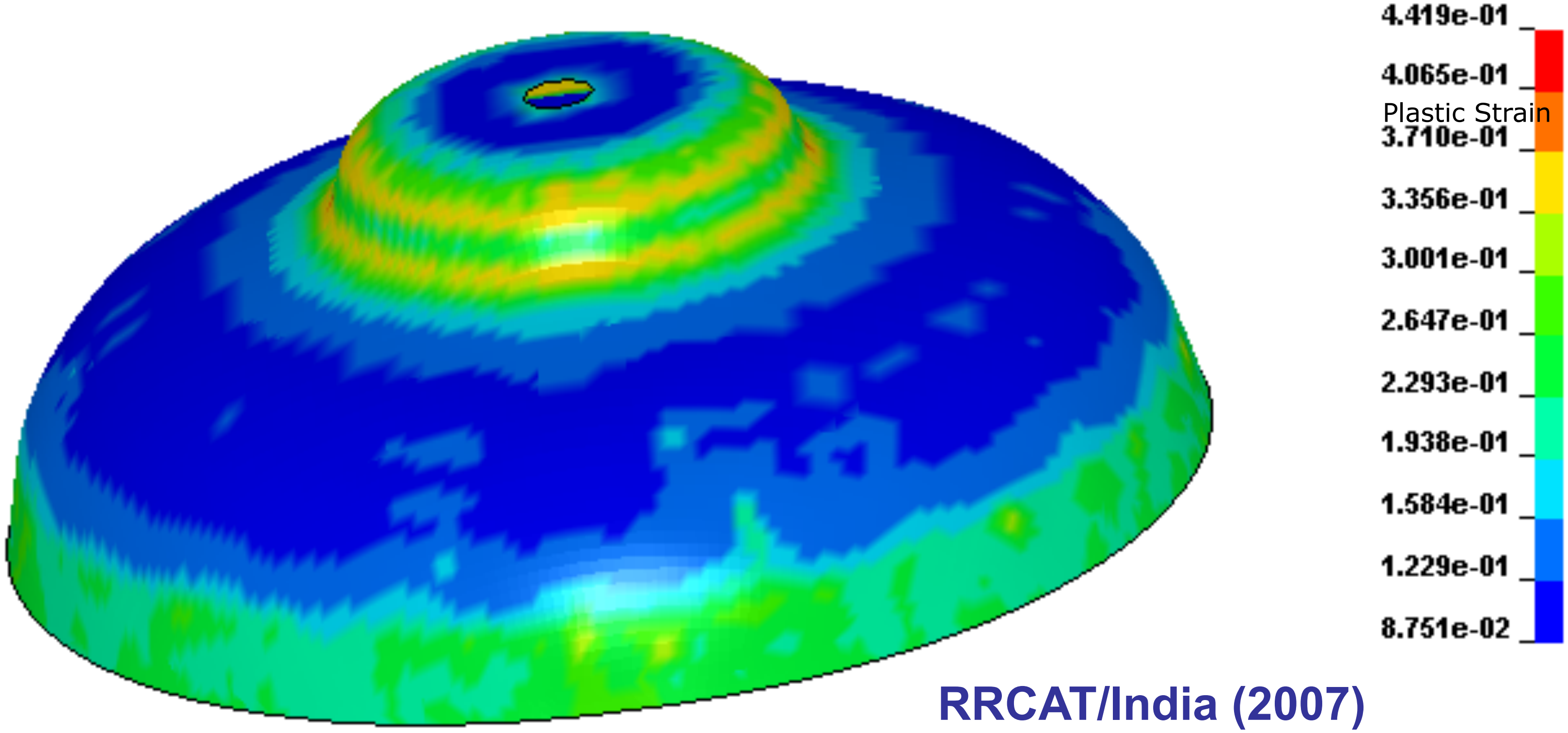
R.P Walsh and D. M. McRae NHMFL 2012

Jefferson Lab



Lower construction cost
Q-rise still need to be understood
FOM ~ 1x10^12
FOM ~ 6x10^11
path to efficient accelerator
Expected FOM ~ 2x10^12
FOM ~ 5x10^11
FOM ~ 2x10^11
Reduce operating cost
Quality Factor (Q0)
Accelerating Gradient (MV/m)
Ingot Nb-highQ-Single Cell JLab (1.5GHz) (Dhakal et al., PRSTAB 2013
Ingot Nb-DESY high gradient 9Cell (1.3GHz) (Singer et al., PRSTAB 2013
Ingot Nb-Reactor Grade-1Cell-JLab (1.5GHz) Kneisel (upbulished)
Fine grain Nb-9cell-JLab (1.3GHz), Geng et al.,IPAC 2011
Figure of Merit (FOM) = Q0^1.5GHz x Eacc

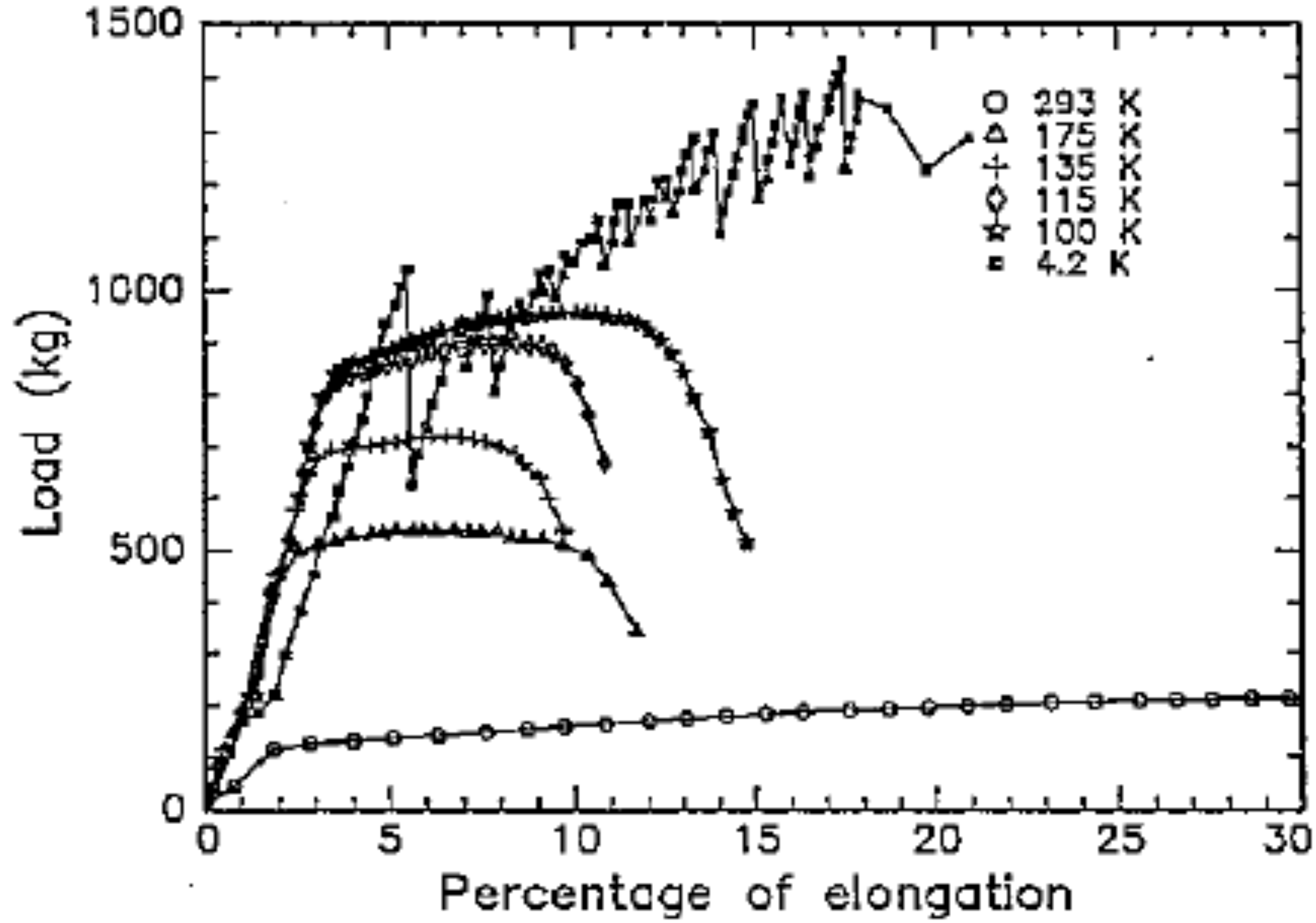
Plastic Strain



RRCAT/India (2007)

Residual stress is largest near the equator and Iris

Peak surface magnetic filed is highest near the equator, where quenches originate, etch pits seen and is the bed for hydrogen-dislocation interactions



Hydrogen-dislocation interaction

From 1993 CEC/ICMC JLab publication

What has been demonstrated

- High RRR (~>300) is not essential

  Fourth melt ingot Nb is OK

- Tantalum up to 1300 wt ppm is fine

- Ingot niobium technology demonstrated by DESY with conventional processes

- Simplified JLab new processes provide a factor of ~ 4 Q_0 improvement

Jefferson Lab

JSA

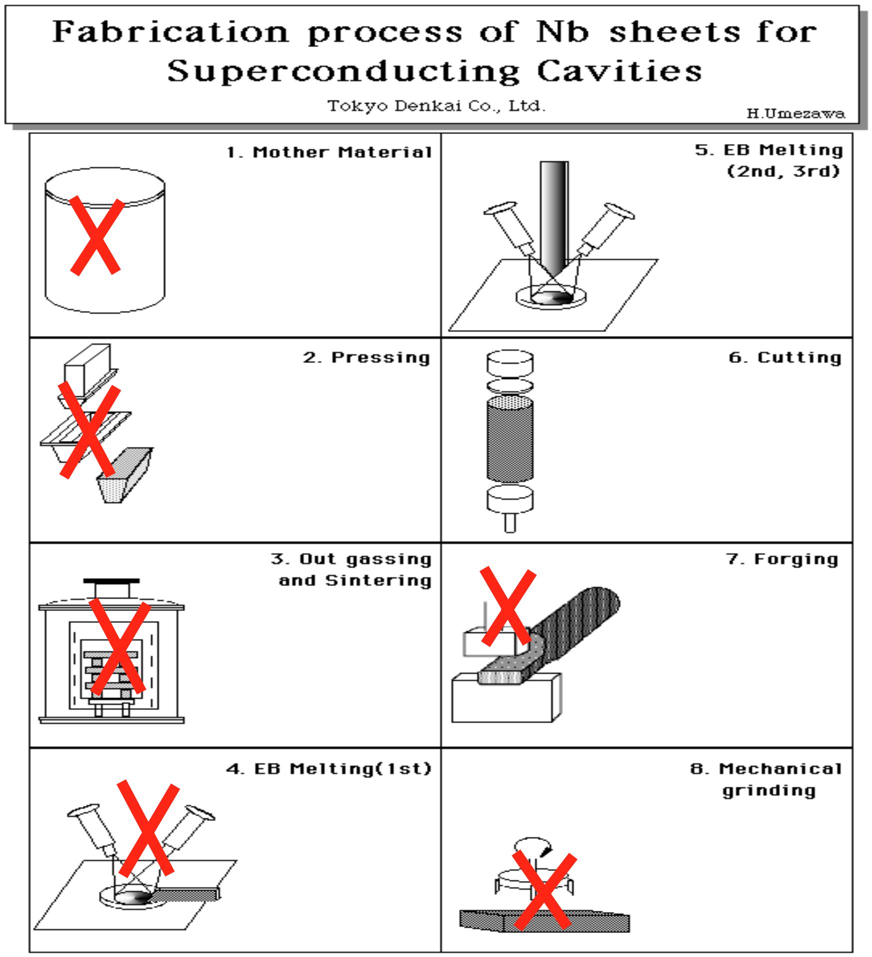
Cryogenic Refrigeration Cost Reduction with improved Qo (~factor of 3) CW SRF Cavities

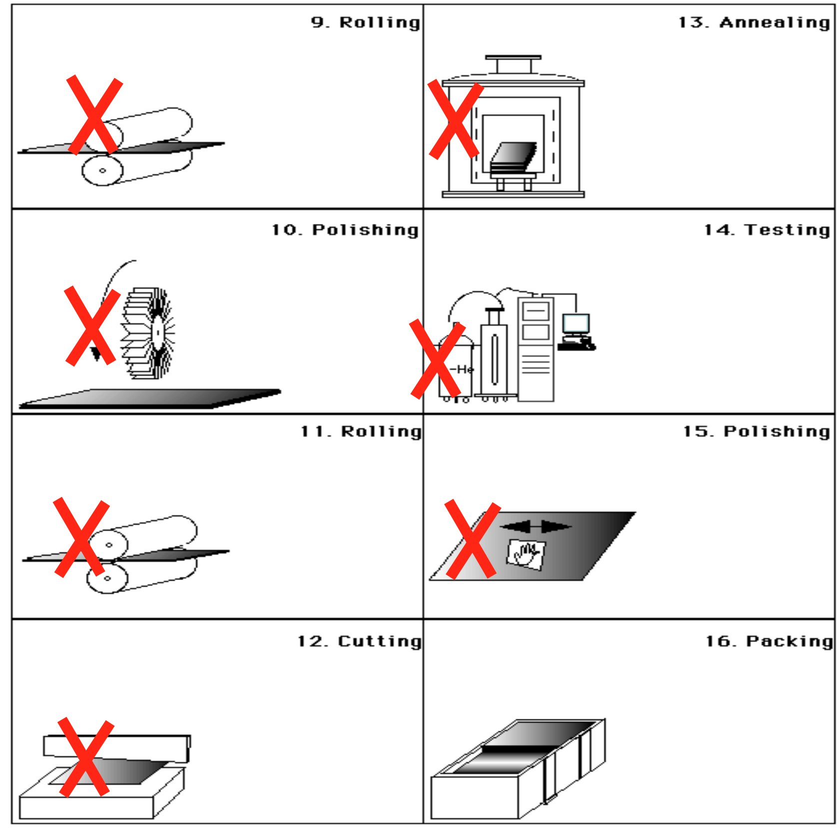
- **A 10 kW 2 K refrigerator costs ~ 100 M$**

- **A factor of 3 improvement in Qo will lower this to ~ 45 M$**

- **The power consumption and hence the operating costs will be reduced by a third**

Economic, efficient and sustainable path for CW applications





Note:
Cost of the ingot sliced Nb sheets anticipated to be less than a third of polycrystalline Nb & no QA

Expected material cost savings at least 50%

Simplified process steps

- Minimize the process steps
  - Centrifugal barrel polishing ~ 100 micro meters
  - High pressure ultra pure water rinse
  - > 1400 C heat treatment
  - High pressure ultra pure water rinse
  - RF test
- These simple steps are expected to minimize hydrogen absorption

Future Outlook

- **Ingot niobium technology (low RRR, high tantalum content) has proven to be ideal for CW SRF applications**

- **We expect that this technology will be the preferred choice for future superconducting CW linacs world-wide**

- **An international workshop on proton-hydrogen interactions will be organized to further develop the understanding of the last unresolved scientific issue**

Virginia Nuclear Medical Isotope Development Facility

S. Gobalkrishnan[1], S. M. Bilbao y Leon[1], G. Myneni[2], D. Wells[3], J. Zweit[1]
[1] Virginia Commonwealth University, Richmond, VA, USA
[2] Jefferson Laboratory, Newport News, VA, USA
[3] South Dakota School of Mines and Technology, Rapid City, SD, USA

- The US FDA approved the use of the radioisotope ^{67}Cu, for cancer treatment and imaging trials in humans, about 25 years ago. However, despite its attractive decay and imaging properties, ^{67}Cu is not yet available in sufficient quantities from spallation reactions using high energy cyclotrons. In order to accelerate its clinical use in therapeutic and imaging applications, an alternative approach to its production is needed. Superconducting 50 MeV electron linacs are expected to remedy this situation very effectively. Virginia Commonwealth University Schools of Medicine (including Center for Molecular Imaging), and Engineering, Jefferson Lab and South Dakota School of Mines and Technology have come together under the recently established Virginia Nuclear Energy Consortium umbrella to jointly develop the Virginia Nuclear Medical Isotope Development Facility (VNMIDF). The partner institutions are planning to develop new approaches to provide a "step change" in the production of copper radioisotopes, in particular ^{67}Cu for multi-modality biomedical imaging and multi-therapeutic applications. The strategy is extensible and amenable to the production of other radioisotopes spanning a variety of applications including scientific, medical and industrial uses. The VNMIDF will not only aid in the development of critically needed medical radioisotopes but will also provide huge opportunities in materials research, including theranostic (therapeutic and diagnostic) nanoparticles and complementary research in novel approaches in non-invasive image guided technologies in surgical, radiation, drug therapeutics and training of future generation of scientists and engineers.

Accapp'13 abstract

International Symposium On Hydrogen In Matter (ISOHIM) Publications

Hydrogen in Materials and Vacuum Systems AIP CP 671
http://www.virtualjournals.org/dbt/dbt.jsp?KEY=APCPCS&Volume=671&Issue=1

Hydrogen in Matter AIP CP 837
http://www.virtualjournals.org/dbt/dbt.jsp?KEY=APCPCS&Volume=837&Issue=1

Single Crystal Large Grain Niobium AIP CP 927
http://www.virtualjournals.org/dbt/dbt.jsp?KEY=APCPCS&Volume=927&Issue=1

Superconducting Science and Technology of Ingot Niobium AIP CP 1352
http://scitation.aip.org/dbt/dbt.jsp?KEY=APCPCS&Volume=1352&Issue=1

JLab's worldwide network of collaborators

Tadeu Carneiro, Marcos Stuart – CBMM	ingot niobium technology
F. Stevie, P. Maheswari (grad student), D. Griffis – NCSU	niobium surface science
R. Ricker – NIST	hydrogen-niobium system
J. Wallace – Casting Analysis Corporation	co-PI DOE ONP ARRA Q_0 improvement program
Björgvin Hjörvarsson – Uppsala University	hydrogen-niobium system
B. Lanford – UNY, Albany	nuclear reaction analysis
R. Pike and summer student interns – W&M	XRD analysis of niobium
Hani Elsayed-Ali, Ashraf Hassan Farha (grad student) – ODU	niobium nitride
Asavari Dhavale & J. Mondal (grad students) – BARC/HBNI	ingot niobium properties
Sindhunil Roy – RRCAT	SC properties of niobium
Saravan Chandrasekaran – MSU	ingot niobium properties
International Symposium On Hydrogen In Matter (ISOHIM)	non profit organization for education/training

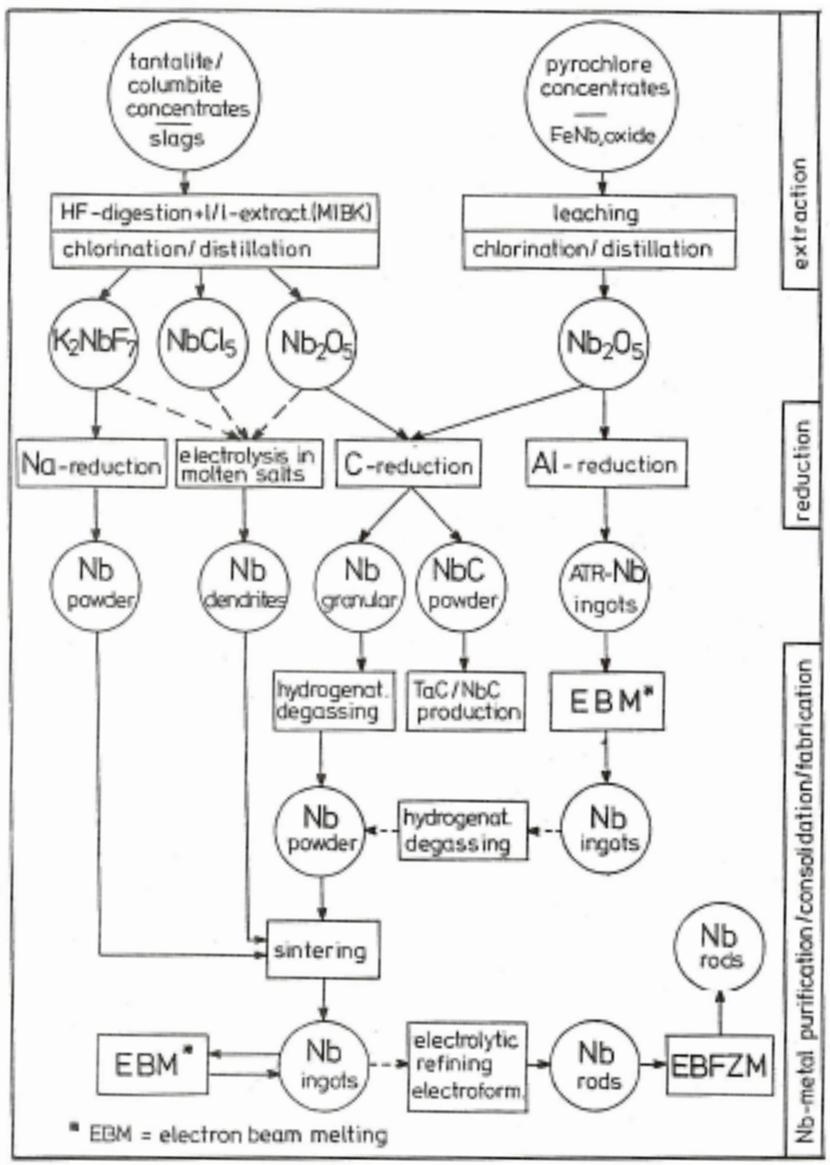
Acknowledgements to all colleagues at JLab

Back up slides

CEBAF niobium (Wah Chang and Fansteel)

ASTM ~5 fine grain
Nb sheets
Ta <1000 wt. ppm

10% form Cabot Columbite/ Tantalite Ore

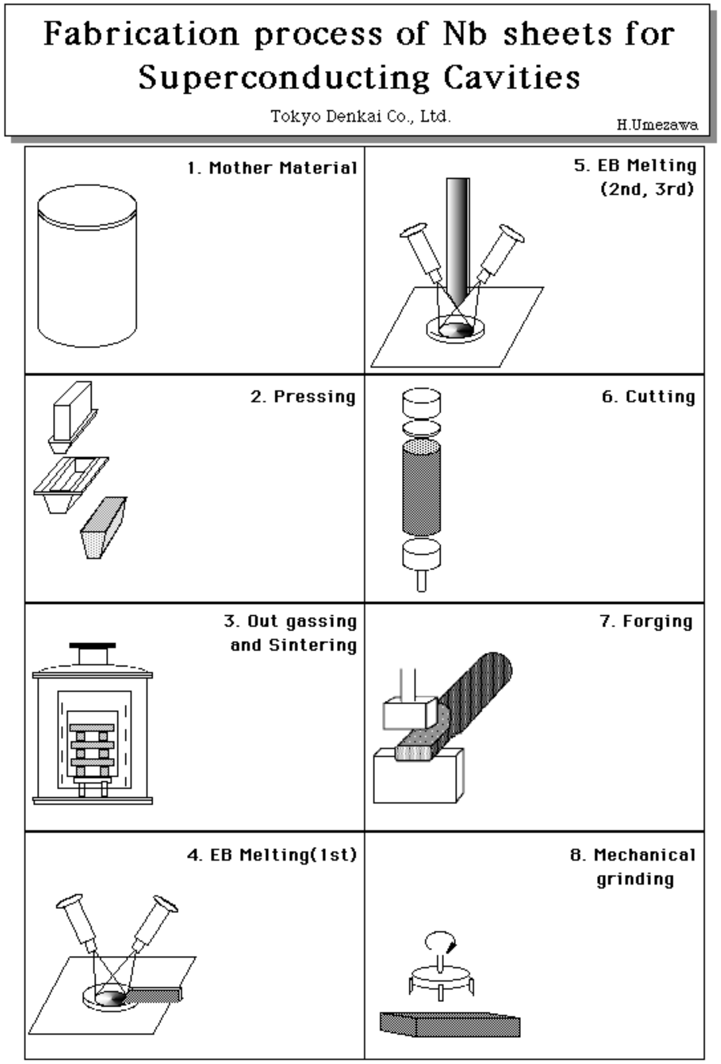


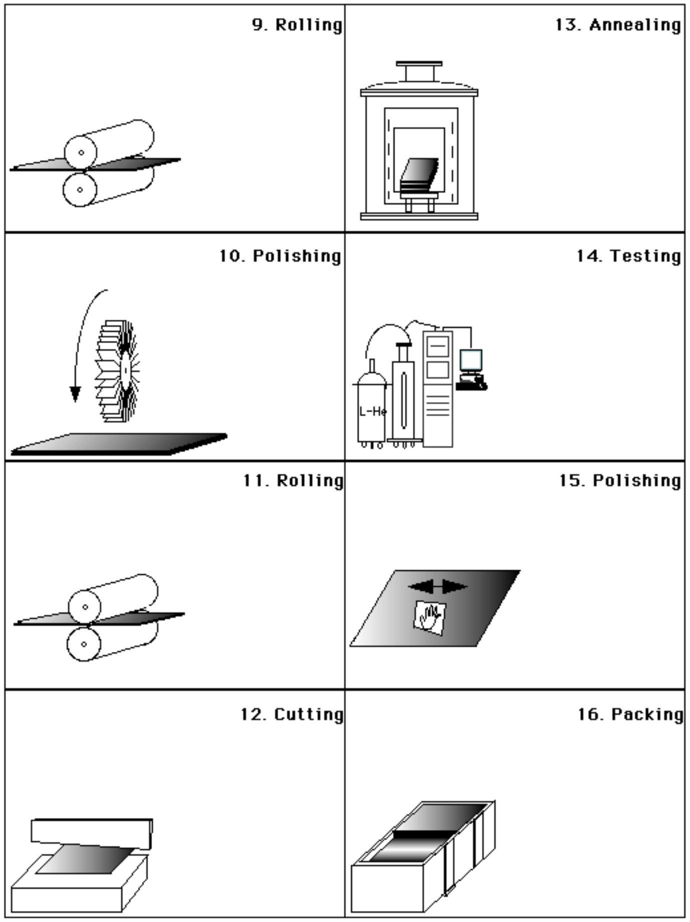
K. Schulze et al in Niobium 1981

90% from CBMM Pyrochlore Ore

Jefferson Lab

Process steps - fine grain Niobium





Note:
During this process foreign materials can be embedded so QA is required

Current niobium derived from Cm/Ta Ore

Ta < 500 wt. ppm

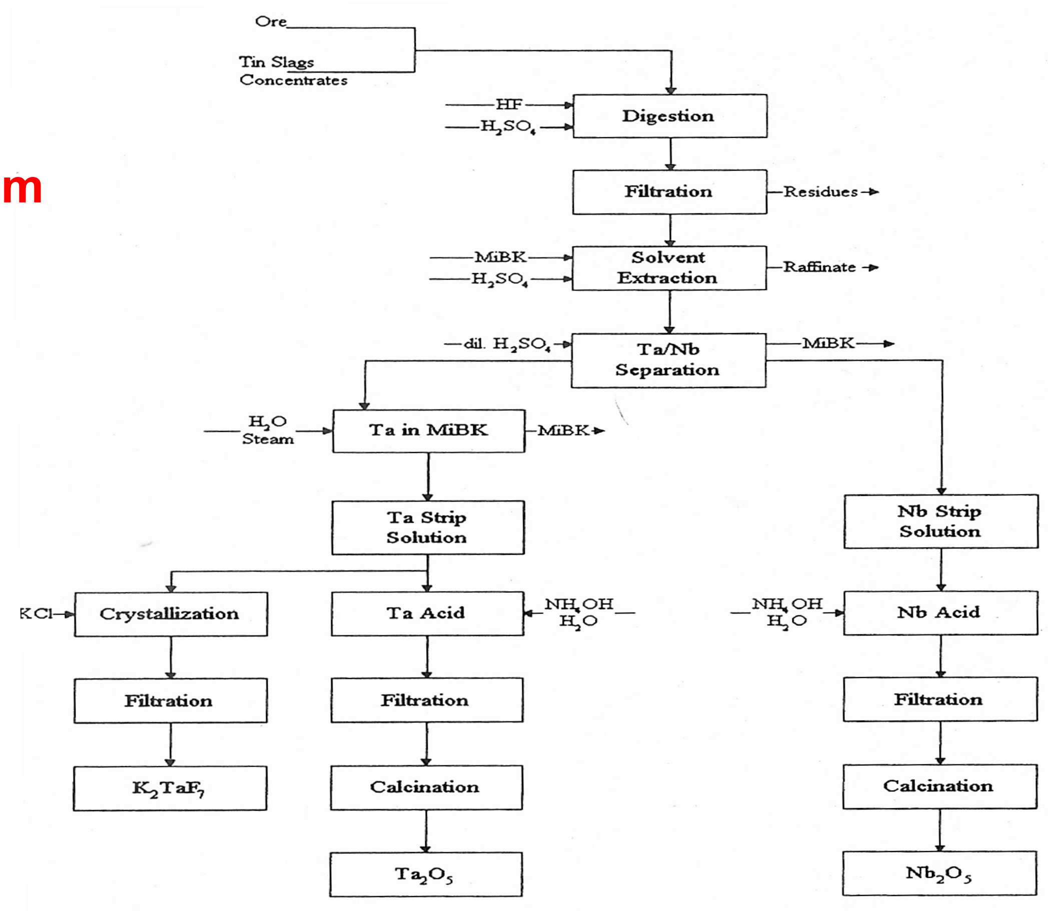


Figure 2: Hydrometallurgical Processing of Niobium and Tantalum.

Heraeus, Ninxia, Plansee, Tokyo Denkai and Wah Chang

Eckert et al in Niobium Science and Technology 2001

Ingot niobium from Pyrochlore Ore CBMM
Ta up to ~ 1500 wt. ppm

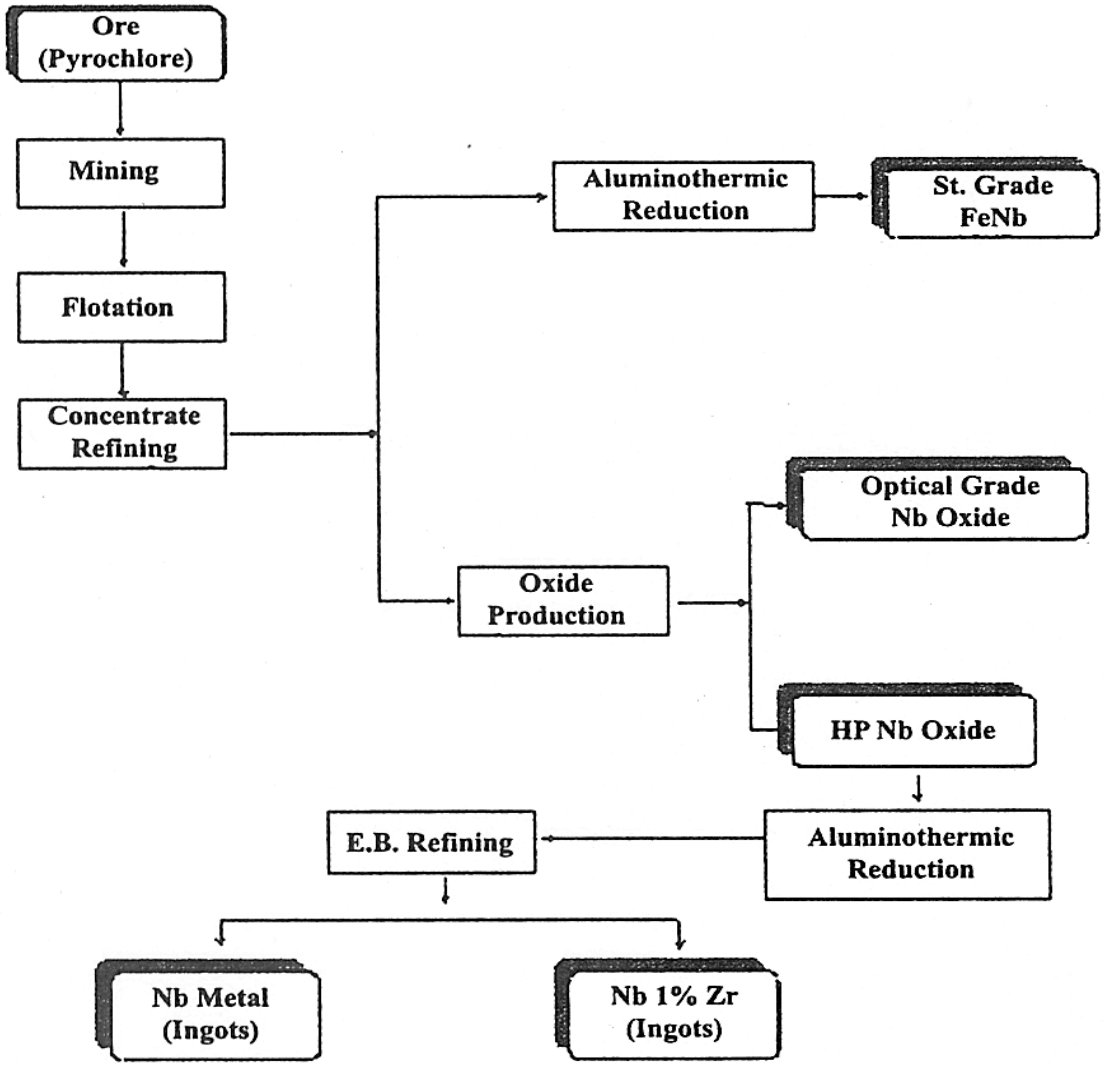


Fig. 3: Production flow chart at CBMM.

H. R. S. Moura in Niobium Science and Technology 2001