PROGRESS ON LARGE GRAIN AND SINGLE GRAIN NIOBIUM – INGOTS AND SHEET

AND

REVIEW OF PROGRESS ON LARGE GRAIN AND SINGLE GRAIN NIOBIUM CAVITIES*

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Abstract

Large grain and single crystal niobium has been proposed several years ago as an alternative material to poly-crystalline niobium for superconducting cavities, exhibiting potential advantages such as "stream-lined" procedures, reduced costs and better reproducibility in performance. Several major laboratories have investigated the use of large grain and single crystal material in the past years and the niobium producing industry has responded in providing ingot material with enlarged grain sizes. Besides a large number of single cell and multi-cell cavities from large grain niobium, several single crystal cavities have been fabricated and tested with good performances.

This contribution will review the progress since the SRF workshop in 2005 in material processing and handling and in cavity performances.

INTRODUCTION

High purity niobium sheets with RRR–values ≥ 250 are exclusively used for the fabrication of high performance cavities for SRF accelerator projects/future projects such as SNS, XFEL, ILC, RIA, ERL's or the CEBAF upgrade. These sheets are formed from a multiple electron beam melted ingot by an elaborate process of forging, annealing, rolling and chemical etching. Until now it has been believed that uniform and fine grain material (ASTM grain size > 6) is needed for deep drawing of cavity half-cells. The sheet manufacturing process bears the risk of foreign material inclusions in the material and stringent QA procedures such as eddy current or SQUID scanning of the sheets are administered to ensure defect-free material prior to the cavity manufacturing. On the other hand, the use of niobium cut from a large grain ingot could be an alternative and conceivably less expensive manufacturing process, if the mechanical properties and the formability of this material are comparable to sheet material.

Approximately three years ago a program was started at Jlab to pursue this idea, since large grain niobium had been made available by CBMM. At the 12th SRF workshop, held at Cornell University in 2005, the first encouraging results were reported. Two contributions discussed material properties of large grain and single crystal niobium and a contribution from Jlab reported about performances of cavities made from large grain and single crystal niobium. At that time two 2.3 GHz TM_{010} cavities of the scaled CEBAF High Gradient and Low Loss shapes had reached accelerating gradients of E_{acc} ~ 45 MV/m, corresponding to magnetic quench fields close to the critical field of niobium (~ 185 mT).

Many potential benefits were expected from the large grain material as listed below:

- Reduced costs for the material because the sheet fabrication step is eliminated.
- Comparable performance to fine grain material.
- Very smooth surfaces with buffered chemical polishing (BCP), electropolishing (EP) not necessary; this in particular might simplify the procedures for attachment of He-vessels after vertical qualification of multi-cell cavities and the assembly procedures for cavity strings.
- Less material QA such as eddy current or squid scanning.
- Possibly elimination or shortening of "in situ" baking because the onset value for "Q-drop" might be shifted to higher gradients.
- Higher thermal stability because of a "phonon peak" in the thermal conductivity.
- Possibly very low residual resistances (high Q-values), favoring lower operating temperature with less cryogenic losses.

Because of these potential advantages of large grain material, the interest in investigating it has grown between 2005 and 2007 and many laboratories have implemented their own programs for material studies and cavity fabrication/testing. Additionally, large grain niobium has become a subject of interest at every major accelerator conference and a two day workshop with 25 participants on " Single Crystal – Large Grain Niobium Technology" was organized in 2006 under the sponsorship of CBMM in Araxa, Brazil [1].

This small workshop had participants from 10 laboratories (FNAL, Jlab, DESY, KEK, INFN, MSU, FSU, NIST, NCU,PKU) and 8 industrial companies (AES, ACCEL, Tokyo Denkai, W.C. Heraeus, Starck, Plansee, CBMM, Wah Chang).

The workshop program consisted of overview talks on niobium technology and the planned International Linear Collider (ILC), reports of the different laboratories on

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Table 1: Summary of material studies on large grain and single crystal niobium.

Subject	Comment	Reference
Mechanical properties	Yield strength, tensile strength, elongation, bulging, residual stress	[2, 18, 19]
Thermal properties	Thermal conductivity, phonon peak, effect of strain	[2, 3, 20]
Magnetic/electrical properties	H_{c1}, H_{c2}, H_{c3} for different crystal orientations, temperature dependence	[5]
Crystal orientation, recrystallization	Grain orientation in different material, etch rate, dislocation density	[3, 20, 21]
Forming/welding	Increase large grain to any size single crystal, fabrication issues	[9, 10, 20]
Flux penetration	Influence of grain boundaries on flux penetration	[4, 22]
Oxidation	Oxide composition for different crystal orientations	[6, 23, 24]
Field emission	Emitter density, cleaning, grain boundary segregation	[12]

their activities and reports from industry about niobium production and cavity fabrication. The conclusion from this workshop can be summarized as:

- it will be quite difficult to produce single crystal ingots of large diameter.
- For large grain application there is no real "show stopper".
- For large scale application, more experience and confidence in the material is needed, which can be gained by production of a cryomodule consisting of e.g eight 9 cell cavities.

In the following a review of the worldwide efforts between 2005 and 2007 will be given and it will be attempted to evaluate and compare the anticipated benefits with reality.

MATERIAL STUDIES ON SHEET MATERIAL

Table 1 lists the various kinds of material studies, which have been conducted in different laboratories between 2005 and 2007.

Mechanical Properties

Tensile tests performed with a single crystal from the center of an ingot with RRR ~ 280 and a Ta content of ~ 800 ppm supplied by CBMM showed far superior behaviour in ductility to fine grain material and an elongation of 100 % was achieved [2]. In addition, no yielding was observed in the elastic region of the stress-strain curve, a favorable property of the material in comparison to fine grain niobium (Fig. 1).

However, the mechanical properties show a significant anisotropy and a dependence on crystal orientation [3]: in the [100] orientation, the measured elongation exceeded 100%, whereas crystals grown in the [110] and [111] orientation broke at approximately half that value.

A 2-dimensional hydraulic bulging test on a large grain sample showed a significantly lower elongation after fracture - < 15% - compared to the uni-axial tensile test; the fracture occurred on a grain boundary close to the center of the disc used for the biaxial deformation.

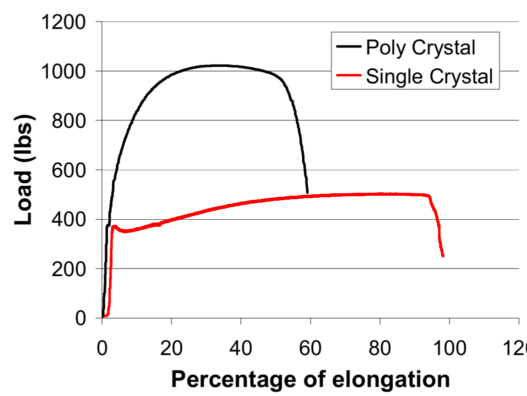

Figure 1: Load-elongation curves for the single and polycrystalline niobium samples.

Thermal Properties

Unstressed large grain or single crystal niobium shows a phonon peak around 2 K, which disappears, when stress is applied to the material, as shown in Fig. 2.

Magnetic Properties/Flux penetration

Magneto-optical imaging was used to observe flux penetration at grain boundaries and into single crystals of different orientations.

Grain boundaries are – as expected – a weak area for flux penetration. However, this weakness is very sensitive to the orientation of a grain boundary with respect to the applied external magnetic field. Only if a grain boundary is close to being parallel to the applied field is this weakness revealed. For randomly oriented sheets in an RF cavity, the larger the grain size the greater is the distance between these weak grain boundary locations [4].

In single crystals of different orientation the flux penetration is fastest for the [111] orientation, slower for the [110] orientation and slowest for crystals grown in [100] direction [5].

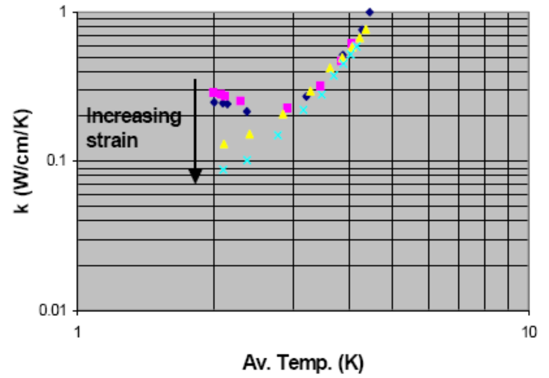

Figure 2: Thermal conductivity of large grain, high purity niobium. The phonon peak at 2 K disappears, when the material is stressed [3].

Oxidation

Single crystal niobium specimens of different crystal orientations ([100],[110] and [111]) were analyzed using TEM and SIMS and using Focussed Ion Beam (FIB) preparation techniques for the TEM samples. Uniform niobium oxide thicknesses were found for all three samples after buffered chemical polishing, ranging from 4.9 nm for the [100] crystal to 8.3 nm for the [110] orientation. Figure 3 shows the cross section oxide on the [111] crystal.

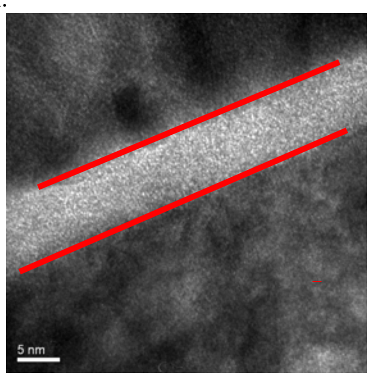

Figure 3: Cross section TEM micrograph of [111] as received sample showing niobium oxide thickness of 7.5 nm

SIMS analysis for all three orientations after heat treatments at 90C, 600C and 1250C showed high concentration of hydrogen between the niobium and the oxide layer, but a relatively low level in the oxide. No high oxygen concentration was found below the oxide layer.

Fabrication Issues

Because of the mechanical anisotropy, large grain niobium sheets show some behavior during deep drawing, which does not occur in poly-crystalline niobium such as slippage of grains at grain boundaries, thinning in some areas of large deformation or tearing, ellipticity of half cells due to spring back and "earing" as shown in Fig. 4.

Figure 4: Forming of half cells from sheet, showing "earing" and grain slippage [7, 8].

However, none of these problems are "show stopper" in fabrication of cavities from large grain sheet material. It has been stated by one of the manufacturers that, even though the assembly of half cells for electron beam welding is somewhat more difficult at the equators, there is no increase in production time and production costs for large grain material [8].

The effects of strain and dislocations in the material after deep drawing are important for re-crystallization processes during electron beam welding and new crystals can form. However, under appropriate annealing conditions even highly deformed single crystals can keep their orientation and the dislocation structure can recover without formation of sub-grains [9, 10]. This result was applied in the fabrication of a single crystal 1300 MHz cavity made by enlarging a smaller single crystal by several rolling and annealing steps and then matching the orientations of both single crystals during electron beam welding [11]. Figure 5 shows the electron beam welding connection for unmatched and matched orientations of two single crystals.

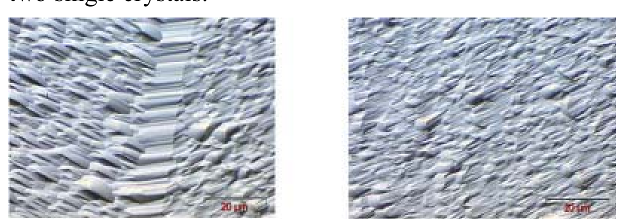

Figure 5: Electron Beam Welding connection for two single crystals: unmatched (left) and matched orientation (right) [9].

Field Emission on Single Crystal Niobium

DC field emission studies using a field emission scanning microscope have been conducted at the University of Wuppertal since many years. Recent studies – the thesis project of A. Dangwal – focused on the emitter density on large grain and single crystal niobium samples, prepared by buffered chemical polishing and finally cleaned by either high pressure water rinsing or Dry Ice Cleaning (DIC). The results of these studies can be summarized as following [12]:

- Mirror like surfaces – surface roughness ~ 7.5 nm – have been achieved on single crystals after 100 µm of buffered chemical polishing.
- Cleanest surfaces have been obtained after DIC.
- On an emitter-free surface after DIC fields up to 1000 MV/m were measured, which allowed to determine the work-function for two different crystal orientations, namely 3.8 eV for [111] and 4 eV for [100] crystal orientation.
- After "in situ" heat treatment at 150 °C more emitters were found on large grain niobium, indicating grain boundary segregation, whereas on a single crystal sample no new emitters appeared.
- Large grain/single crystal niobium samples show less emitters after 100 µm buffered chemical polishing and subsequent high pressure water rinsing (HPR) than samples prepared by electropolishing and HPR.

CAVITY RESULTS

Single cell and multi-cell cavities from large grain and single crystal niobium from different manufacturers have been fabricated and tested at Cornell University, DESY, Jefferson Lab, KEK, Michigan State University (MSU) and Peking University (PKU). Large grain material is now available from CBMM, Ningxia, W.C. Heraeus, Wah Chang (limited) and in the near future from Tokyo Denkai [13]. ACCEL is presently the only company with experience in manufacturing single and multi-cell cavities from large grain material.

The following sections will review the cavity work done since the SRF workshop in 2005.

Cornell University

At Cornell University a study was conducted on fine grain and large grain single cell cavities comparing temperature maps with respect to " hot spots" and their appearance near grain boundaries (see Fig. 6) [14].

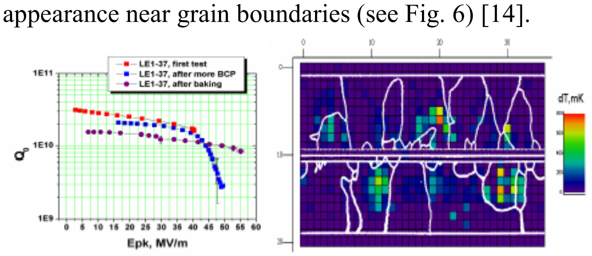

Figure 6: T-Map for the large grain cavity performance indicating the location of grain boundaries and "hot spots" [14].

The investigation showed that grain boundaries are not necessarily the areas of flux penetration.

Peking University

At Peking University efforts are underway to develop the technology of niobium cavity fabrication in collaboration with Chinese industry; both single cell and multi-cell cavities are being manufactured.

A single cell TESLA shaped cavity from large grain Ningxia RRR niobium was – in collaboration with Jlab – surface treated and tested at Jlab. The best performance of the cavity after post-purification is shown in Fig. 7 before and after "in situ" baking at 120 °C for 12 hrs.

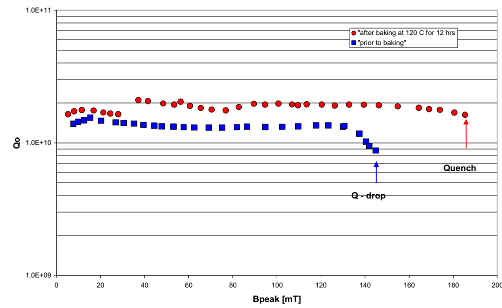

Figure 7: Performance of the first PKU produced Single cell cavity, made from Ningxia large grain niobium.

The magnetic quench field of H~185 mT, which is close to the fundamental limit of niobium, corresponds to an accelerating gradient of E_{acc} = 43.5 MV/m, one of the highest fields obtained in a single cell cavity of this shape [15].

KEK

At KEK, three Ichiro – type single cell cavities were fabricated from large grain Ningxia niobium and tested after different amounts of material removal by centrifugal barrel polishing (CBP), BCP and electropolishing (EP) [16]. Cavities # 1 and #2 reached very high gradients of E_{acc}=47.9 MV/m (H_{peak}~ 170 mT) and E_{acc}= 43 MV/m (H_{peak}~ 155 mT), respectively, after 90 µm CBP, 10 µm and 80 µm EP, whereas the third cavity performed mediocre. In all cases low residual resistances were measured; however in contrast to the BCP'd large grain cavities a baking time of 48 hrs (12 hrs for BCP'd cavities) was necessary to remove the high field Q-drop.

DESY

Besides Jlab, DESY is the laboratory with the largest interest in evaluating large grain niobium as an alternative to polycrystalline material. The efforts are directed towards material studies and cavity performance studies, based both on BCP and EP as final surface treatments.

Several single cell cavities have been fabricated by industry or "in house" from large grain W.C. Heraeus niobium and these cavities reached very high gradients in the vicinity of E_{acc}~40 MV/m after EP. BCP degraded the performance. More recently, a single crystal single cell cavity was fabricated from a Heraeus large grain disc with a single crystal in the center by enlarging the single crystal as mentioned above [17]. This cavity was surface treated by BCP and tested at Jlab numerous times with successive amounts of material removal and as a best result achieved a gradient of E_{acc} = 38.5 MV/m (H_{peak} ~ 164 mT). Figure 9 shows the "history" of the cavity performance.

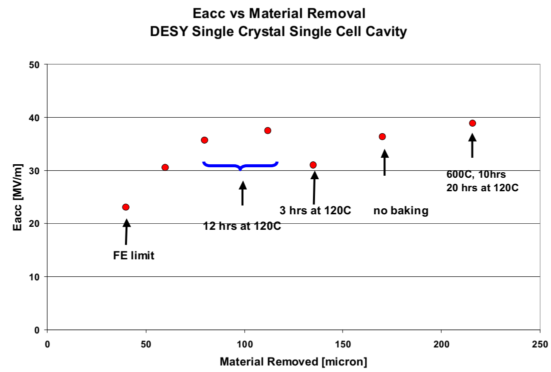

Figure 9: Performance "history" of DESY single crystal cavity.

In addition, three 9-cell TESLA cavities were fabricated by ACCEL for DESY from large grain Heraeus niobium. These cavities performed very well with small spread in data as shown in Table 2 [11].

Table 2: Performance of 9-cell TESLA cavities, fabricated from large grain Heraeus niobium.

Name	**Treatment**	**E_{acc}** (MV /m)	**Q_0 at E_{acc}**	**Limit**
AC112	130 µm BCP, 800C, HPR	30.5	2×10^{10}	FE
AC113	130 µm BCP, 800C, HPR	27.4	2×10^{10}	Quench
AC114	130 µm BCP, 800C, HPR	28.7	2.1×10^{10}	Quench

Jefferson Lab

Between SRF 2005 and the end of 2006 we have fabricated 17 single cell cavities and 3 multi-cell cavities of different shapes and frequencies from four different niobium manufacturers (CBMM, Ningxia,W.C.Heraeus and Wah Chang), using material from 9 different ingots and different cutting methods for the sheets (saw cutting, wire EDM and wire saw cutting). Whereas the multi-cell cavities did not perform very well because of manufacturing issues, the single cell cavities gave encouraging results [7].

A statistical "snapshot" of 51 tests on the single cell cavities is shown in Fig. 10. Among those tests we conducted a study comparing the performance of one cavity each from CBMM, Ningxia and Heraeus material after applying the same surface treatment procedures. Despite a significant cost difference in raw material the cavities performed very similar as shown in Fig. 11.

This performance comparison is being further pursued: five cavities each have been fabricated from the different material of the three manufacturers and they all will be treated the same way, before and after post-purification.

Initial test results for Ningxia and Heraeus cavities are shown in Figs. 12 and 13.

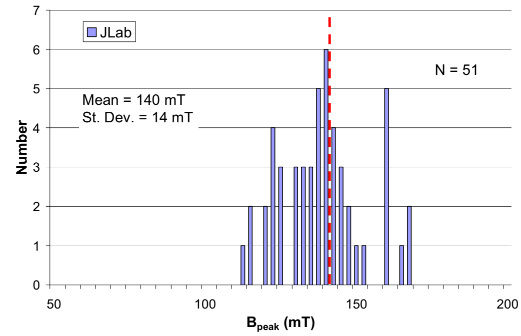

Figure 10: Statistics of large grain single cell cavity performance.

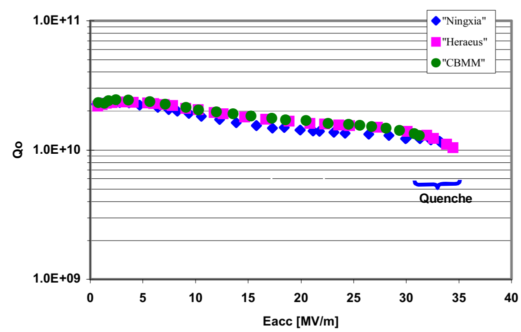

Figure 11: Performance comparison of single cell cavities fabricated from different niobium suppliers.

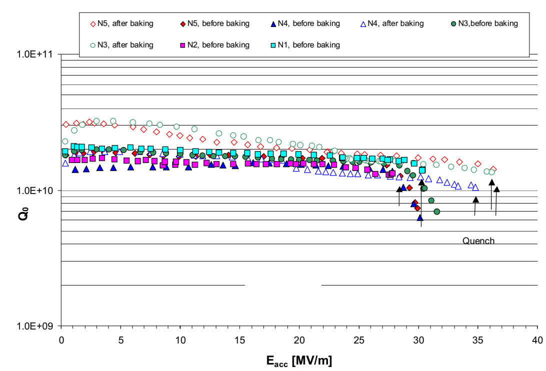

Figure 12: Results from "Ningxia" cavities (TESLA shape).

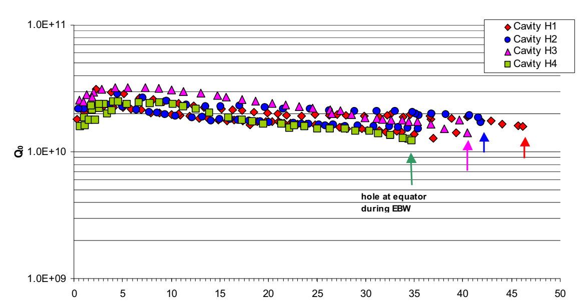

Figure 13: Initial results from "Heraeus" cavities (LL shape).

SUMMARY

Large grain material provides some challenges in fabrication of cavities, but is not a "show stopper". Single crystal sheets would be desirable for manufacturing, but no significant performance improvements over large grain niobium have been seen so far.

The performance of cavities made from large grain niobium is comparable with fine grain niobium and very smooth and high performing surfaces can be achieved with BCP, eliminating the need for electropolishing.

For projects such as the XFEL or cw applications, cavities from large grain niobium offer "streamlined" procedures: BCP, shorter "in situ" baking times and less QA such as eddy current scanning .

Potentially beneficial features such as reproducibility of performance after BCP treatment , cost advantages over polycrystalline niobium and high Q values need further investigations as well as the development of an effective. cutting method. Additional confidence in this material will be generated by the evaluation of more 9-cell cavities or a full cryomodule as presently pursued at DESY.

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