Single Crystal and Large Grain Niobium Research at Michigan State University

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Abstract. As Superconducting Radio Frequency (SRF) technology is used in more accelerator designs, research has focused on increasing the efficiency of these accelerators by pushing gradients and investigating cost reduction options. Today, most SRF structures are fabricated from high purity niobium. Over years of research, a material specification has been derived that defines a uniaxial, fine grain structure for SRF cavity fabrication. Most recently a push has been made to investigate the merits of using single or large grain niobium as a possible alternative to fine grain niobium. Michigan State University (MSU), in collaboration with Fermi National Accelerator Laboratory (FNAL) and Thomas Jefferson National Accelerator Facility (JLAB), is researching large grain niobium via cavity fabrication processes and testing, as well as exploring materials science issues associated with recrystallization and heat transfer. Single-cell 1.3 GHz ($\beta=0.081$) cavities made from both fine and large grain niobium were compared both in terms of fabrication procedures and performance. Two 7-cell cavities are currently being fabricated.

Keywords: Niobium, recrystallization, thermal conductivity, residual resistance ratio, SRF cavity

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LARGE GRAIN CAVITY DEVELOPMENT

In 2005, a research collaboration commenced between Fermi National Accelerator Laboratory (FNAL) and Michigan State University (MSU) to design and prototype SRF cavities to be used in a proton linac. The project of interest was the proposed Proton Driver [1]. The cavity design is similar to the high- β cavity for the Spallation Neutron Source (SNS), with an operating frequency of 1.3 GHz and a reduced β of 0.81. MSU completed the electro-magnetic modeling, produced fabrication drawings, fabricated the cavities (including tooling and fixtures) and performed RF testing (2 K). It was decided to fabricate (6) single-cell cavities, followed by (2) 7-cell cavities. The single-cell prototypes have the same shape as the end cell of the multi-cell cavity. In light of advancements made at Thomas Jefferson National Accelerator Laboratory (JLAB) using large grain niobium [2], a study was made using both "standard" fine grain RRR niobium and large grain RRR niobium to compare fabrication procedures and test results for the two types of materials. The fine grain material supplied by FNAL was procured from Wah Chang as 2.8 mm sheet. The fine grain material was specified with a 50 μ m grain microstructure and RRR >260. The large grain niobium was supplied by JLAB and was procured from CBMM as an ingot. Disks were then EDM (Electrical Discharge Machining) machined from the ingot.

On average, each 10 inch (254 mm) disk contained ~6 grains with a RRR of ~280 (Ta ~800 ppm), as shown in Figure 1a.

The fabrication of the 7-cell cavities is in progress. One cavity will be constructed from the Wah Chang fine grain niobium, with the second using large grain niobium from CBMM.

FIGURE 1. (a) Large grain niobium disk as EDM machined from ingot. (b) Large grain half-cell showing "earring" at the equator diameter.

Fabrication

Fabrication of all single-cells followed the same technique, with slight differences between the two niobium materials. Both materials were water-jet cut into disks 248 mm (9.975 inches) in diameter and 2.8 mm thick. Before stamping, the large grain disks received a light sanding to remove ridges left from the EDM machining (depending on cutting speed, the EDM wire's oscillations can leave ridges across the cutting surface). The disks were deep drawn into half-cells using a 200 ton hydraulic press. The fine grain half-cells were stamped using 200 tons, while the large grain half-cells reached 50 tons. The half-cells received additional coining at the iris at 100 and 40 tons, respectively. The final spring-back was measured across all the stamped half-cells. The averages were 0.25 mm (0.010 inches) for the fine grain half-cells and 0.12 mm (0.005 inches) for the large grain.

The surface of the half-cells appeared very different after deep drawing. The fine grain material remained macroscopically smooth, with some "orange peeling" at the iris. The large grain material was rough after the stamping, with macroscopic ridges development along grain boundaries, very easily felt with fingers. The large grain half-cells also showed "earring" at the equator, as shown in Figure 1b.

The cells were all machined on a CNC vertical mill using tooling fixtures as indication references. Both materials machined similarly, with the large grain niobium being slightly gummier. Visible spring-back was observed when the large grain half-cells were removed from the machining fixtures. The large grain microstructure eases movement and allows relief of stress from the machining, causing the half-cells to become slightly oval. Inter-locking step weld preps were used on all half-cell welds (iris and equator). Before welding, all parts were cleaned in a cleanroom environment using degreasers and ultrasonic cleaners. The half-cells then received a light 1:1:2 BCP (Buffer Chemical Polish) etch of ~20 μ m.

End flanges were first electron-beam welded to the beam tubes, and then machined. The beam tubes were then electron-beam welded onto the half-cells, completing the iris weld. All beam tubes were extruded tubes by Heraeus. The flanges were machined from Nb-55Ti material

using conflat knife-edges for vacuum sealing. The iris was welded from both the inside (RF surface) and the outside.

After the iris welds were completed, the cavity's inner surfaces were inspected for imperfections. The large grain half-cells were mechanically polished on the inside surface to smooth the raised grain boundaries, following the procedure used at JLAB. The half-cell assemblies were chucked into jaws of a machining lathe. Prior to spinning, the grain boundaries were ground using an alumina pad mounted to a pneumatic tool. The remaining inner surface was ground lightly while spinning, to feather out the residual surface irregularities at the boundaries. Using 3M microfinishing film with the cavity assembly spinning, the whole inner surface was polished, working down in grit size ($60\text{ }\mu\text{m} \rightarrow 40\text{ }\mu\text{m} \rightarrow 30\text{ }\mu\text{m} \rightarrow 20\text{ }\mu\text{m}$). The fine grain half-cell assemblies received little polishing, done only to touch up questionable surface imperfections (weld splatter, scratches and dings). After inspection and polishing, all cells again received a light 1:1:2 BCP etch ($\sim 10\text{ }\mu\text{m}$).

The half-cells were joined at the equator using a full penetration electron-beam weld. The same weld parameters were used for both materials. Because of large mismatches in weld prep alignment, an additional weld fixture was required for the large grain half-cells. This fixture was used to draw the equator joint together, supplying equal force around the equator. Use of this fixture required an additional chamber pump down to remove the fixture prior to completing the weld. The equator welds were completed using a 50 kV, 46 mA beam at 12 inches/min on weld preps with 2 mm penetration depth. All electron-beam welds were completed at Sciaky, Inc (Chicago, IL).

Testing

Four single-cell cavities were completed and tested, two fine grain and two large grain. The fine grain cavities were tested at MSU. The cavities were cleaned in a cleanroom environment and prepared for chemistry. Using 1:1:2 BCP, $\sim 180\text{ }\mu\text{m}$ of niobium was removed. The cavities were then high-pressure rinsed (HPR) for 60 minutes and allowed to dry in Class 10 air. The cavities were assembled onto a vertical test insert, evacuated, and tested in a vertical Dewar.

The large grain single-cell cavities were sent to JLAB for heat treatment and performance testing. The cavities were tested four times each, after the various preparation steps given in Table 1.

TABLE 1. Preparation of large grain cavities at JLAB.

	Heat Treatment	Etching (BCP)	HPR
Test 1	600°C for 10 hours	50 μm before, 50 μm after	60 min.
Test 2	120°C for 12 hours		
Test 3	1250°C for 2 hours*	50 μm after	60 min.
Test 4	120°C for 12 hours		

* Postpurification heat treatment, using titanium as getter material.

A summary of the test results is shown in Figure 2. Full details on the cavity testing and results were presented at Linac 2006 [3]. The test results show better performance in the large grain single-cells. Note however that neither of the fine grain cavities was annealed. The present plan is for the fine grain single-cells to be heat treated and retested at JLAB.

FIGURE 2. Test results for single-cell cavities: fine grain after BCP and HPR processing, and large grain after heat treatment, BCP, and HPR processing.

Multi-cell Cavity Fabrication

One 7-cell cavity is being constructed from fine grain niobium and the other from large grain niobium. The half-cell fabrication and iris welding is finished, as shown in Figure 3b. The completed “dumbbells” are undergoing checks to determine the amount of material to be removed at the equator for weld prep. Fabrication is expected to be complete in early 2007.

FIGURE 3. (a) Design drawing of the 1.3 GHz, $\beta=0.081$ 7-cell cavity. (b) Completed large grain dumbbell assemblies for fabrication of 7-cell cavity.

MATERIALS SCIENCE

The MSU/FNAL collaboration has been engaged in a materials science program with the focus on niobium, to better understand the relationships between cavity preparation, mechanical properties, thermal properties, and cavity performance. The effect of strain in single grain and large grain niobium and its role in recrystallization and heat transfer properties are being examined.

Recrystallization

Recrystallization plays a key role in the mechanical and thermal properties of niobium. During cavity fabrication, strain is introduced into the niobium during the deep drawing, rolling, and machining steps. If this strain is not removed/recovered from the material prior to electron-beam welding, the heat from the welding can generate recrystallization. New crystal orientations may occur by solidification of new large grain poly-crystalline structures in the weld melt zone and recrystallization and/or grain growth of the niobium in the heat affected zone.

When fine grain niobium is welded, solidifying grains have orientations similar to those present in the parent material, indicating that solidification occurs epitaxially on existing grains, which grow significantly as the temperature increases with the arrival of the weld pool, as shown in the heat affected zone in Figure 4a. The grain size in the heat affected zone is close to the 1-2 mm size observed in the melt zone. Electron-beam welding of large crystal niobium was also studied. A sample with two grain orientations was sliced into halves with EDM machining and the lower half was rotated 180° about a vertical axis to produce a weld sample where each of the two crystals orientations would form a bi-crystal joint, with two triple grain locations, shown in Figure 4b. Orientation Imaging Microscopy (OIM) after welding showed no new crystal orientations in areas where the two crystals were joined. However, in areas where the three crystals were joined, additional crystal orientations formed, as shown in Figure 4c and 4d, which correspond to the two middle inset boxes in Figure 4b. This indicates that triple junctions alter the stability of the solidification process in the moving weld pool. Further details of this research were presented at ASC 2006 [4].

FIGURE 4. a) OIM scan of the heat affected zone of fine grain niobium after electron-beam welding. (b) Single-crystal weld sample preparation. (c) OIM scan of bi-crystal interface after e-b welding. (d) OIM scan of tri-crystal interface after e-b welding. Weld direction was from left to right.

A study is underway to measure recrystallization behavior in heat affected zones of single-crystal niobium welds, which occur in locations where large plastic strains developed upon forming. Sections of the outer ring of niobium used to make half-cells were used as tensile

specimens (with no additional processing steps). A single crystal section was deformed in tension to induce strains similar to those present in the equator and iris regions of half-cells. OIM scans were made on the deformed samples to verify that they are still truly single crystal, and to determine their crystal orientation. The samples were then welded on one end, to simulate the thermal history that would occur under standard niobium electron-beam welding. The samples were scanned again to see whether the single crystal orientation had been maintained. The study is still in its early stages, but a preliminary sample showed that substantial recrystallization occurred, as illustrated in Figure 5. The crystal orientation is indicated by the color, so the smooth gradation in color in the sample before welding indicates gradual changes in crystal orientation that arise from plastic deformation. After welding, the boundaries with highly different colors indicate formation of crystals with very different orientations. In addition, observations on completed large grain single-cell and multi-cell cavity welds also show evidence of recrystallization occurring in both the iris and equator heat affected zones.

FIGURE 5. (a) Niobium sample used in recrystallization studies. (b) OIM scan of niobium sample after applying strain, before heating. (c) OIM scan of niobium sample showing recrystallization caused by heating introduced by welding (the speckled green regions after welding are an artifact of incomplete removal of surface oxide).

Heat Transfer

A program is underway to characterize the heat transfer and Kapitza resistance properties of niobium. For temperatures below 3 K, heat transfer is dominated by phonon movement in the crystal lattice. Residual strain and grain boundaries in the material can slow down phonons and consequently the rate of heat transfer. Single-crystal and bi-crystal samples were fabricated to measure their heat transfer characteristics, and compared with fine grain niobium measurements. The single crystal samples were EDM machined from large grain disks, with no prior forming. The initial data showed the phonon peak to be almost absent in both samples, matching very closely with fine grain niobium with known residual strain levels, see Figure 6a. These measurements suggest that the single crystal niobium has residual strain trapped in it from ingot/purifying processes, or that other mechanisms are inhibiting phonon movement. One alternative mechanism could be trapped hydrogen in the material. Single and large crystal niobium is known to have high hydrogen content due to EDM machining. The samples will be heat treated and retested. Annealing will reduce the strain and the hydrogen content, which is expected to cause the phonon peak to increase, as observed in fine grain niobium (see Figure 6b). Further details on heat transfer studies were presented at ASC 2006 [5].

FIGURE 6. (a) Thermal conductivity measurement for single-crystal and fine grain samples. (b) Phonon peak behavior in annealed fine grain niobium with increasing strain; the phonon peak diminishes as the residual strain increases.

CONCLUSIONS AND FUTURE WORK

Single-cell cavities were fabricated from fine and large grain niobium and tested. The fabrication was similar for both materials, but some additional work was required for the large grain material. Early test results show a performance advantage for the large grain cavities, which reached higher gradients with no electro-polishing. The fine grain single-cell cavities will undergo retesting with additional heat treatment to provide a better basis for comparison. Fine grain and large grain 7-cell prototype cavities are being fabricated, with test results expected in the summer of 2007.

Electron-beam welding of single grain niobium can be completed while maintaining the single crystal microstructure, but recrystallization is likely in regions near welds where large plastic strains from cavity forming occurred. In welds between large grain materials with no plastic strain, additional crystal orientations developed during solidification of the weld pool in the vicinity of grain boundaries. Systematic studies of recrystallization behavior in single and large grain niobium are in progress. If recrystallization becomes a fabrication concern, methods to recover stored strain without recrystallization in a pre-welding heat treatment will be explored to determine whether the single or large crystal character can be maintained after forming. The influence of initial crystal orientation on forming processes and its influence on spring-back also need to be assessed for alignment issues during the cavity fabrication.

Initial heat transfer measurements of single crystal niobium, with no prior forming, show evidence that internal defects are inhibiting phonon movement. The single- and bi-crystal heat transfer samples will be given heat treatments and retested. Removal of strain should allow the phonon peak to recover, as seen in fine grain samples. Combined with recrystallization studies, heat treatment procedures will be sought that to ease SRF large grain niobium cavity production.

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