1. **DOE award DE-SC0004222 Final Report**

2. **Foundations for quantitative microstructural models to track evolution of the metallurgical state during high purity Nb cavity fabrication**

3. **17 March 2014, for period starting 6/15/2010 to 6/14/2014**

4. **Brief Description of Accomplishments**

The goal of the Materials Science SRF Cavity Group of Michigan State University and the National Superconducting Cyclotron has been (and continues to be) to understand quantitatively the effects of process history on functional properties. These relationships were assessed via studies on Nb samples and cavity parts, which had various combinations of forming processes, welding, heat treatments, and surface preparation. Characterization of the microstructural state were performed at each stage, tracking the evolution and providing data for future modeling. Collaborations with personnel at the Thomas Jefferson National Accelerator Facility led to provision of interesting samples, and some joint efforts. Primary accomplishments are summarized and enumerated in the several paths of study that were followed, which are more or less in order of time in vested.

1. Thermal conductivity analysis of large grain single and bicrystal samples with different annealing and deformation histories show varying effects of heat treating, deformation, and hydrogen infusion. This allowed effects of grain boundaries to be separated from intrinsic properties of Nb. These results show that changes in thermal conductivity by as much as a factor of five are sensitive to crystal orientation, grain boundary misorientation, and operation of specific dislocation systems during deformation. Overall the quantitative results show that the phonon peak is degraded or removed due to the presence of deformation (high dislocation density) and the presence of hydrogen, in reversible ways, i.e. vacuum heat treatments can restore the phonon peak that was once present. The most effective heat treatment temperature appears to be about 1000°C for two hours. Lower heat treatment temperatures result in retention of some defects, and higher heat treatment temperatures may leave excess vacancies in the sample, which are retained (and may condense as dislocation loops) during cool down. This is an important issue for large grain material since vacancy sinks, such as grain boundaries, may be absent.

   While these overall trends are consistent with the experience of the community, the presence of significant outlying cases shows that there are unexplained physical phenomena that inhibit restoration of the phonon peak. In some cases, the phonon peak that was once present could not be regained after deformation or exposure to hydrogen. The number of these outliers is in the minority, but understanding the reasons for these outliers may be important as this may be related to the variability in cavity performance that is often observed when processing history is otherwise the same. Most of these results have been published, or presented at meetings with archived presentations, so the outcomes are available for the community to examine. Also, most of these samples have been characterized using synchrotron x-ray diffraction methods at the Advanced Photon Source, resulting in big quantities of data that are only partially examined, but are still available for future study.

   Overall, this identifies the importance for developing the ability to predict dislocation substructure prior to and after deforming, so that subsequent heat treatment can be most effectively chosen. This work was the focus of Ph.D. student Saravan Chandrasekaran, and he completed his Ph.D toward the end of this research contract.

2. A series of single crystal samples extracted from a high purity slice from an ingot were deformed and analyzed, leading to many curious effects. The samples were carefully oriented to favor different combinations of slip on 110 vs. 112 planes, and with different relative amounts of slip activity in similar or different 111 directions. The analysis of these results was quite challenging, and comparisons with prior single crystal experiments showed that there is a significant presence of dislocations in the ingots, and that slip on 112 planes may be the controlling factor for dislocation generation leading to hardening, even when slip on 110 planes is favored. This is consistent with prior work to some extent, but much of the prior work on Nb used either much higher or much lower purity Nb. The presence of O and H has a significant influence on the relative activity of slip systems. This work is the first systematic study on intermediate purity samples, which are technically relevant for cavity fabrication. Derek Baars completed his Ph.D. dissertation focusing on these samples.

   This work on as-extracted samples was very challenging to interpret, and it led to the need to anneal companion samples to examine how a standard 800 °C anneal affected the results. This work just started as the funding ended, and the results obtained since then indicate a more regular kind of deformation that is easier to interpret, which also provides a path for more meaningful interpretation of the deformation behavior of samples extracted from the as-received ingot. Perhaps the most important outcome is that the deformation path is significantly altered by annealing, in that slip on 110 planes is favored, whereas the pre-existence of dislocations biases slip to occur preferentially on 112 planes. Consistent with the presence of significant outliers from the main trend observed in the thermal conductivity experiments, some of the annealed samples show strong preference for slip on 112 planes. It is not yet known why.

   As the data on the as-received condition can best be interpreted in the light of samples that were annealed, only a preliminary assessment has been published, though the work of Baars has laid a strong foundation for interpretation of these results. The analysis of the annealed samples is ongoing, and in about a year, a major paper that considers both sets should be submitted, as this work continues with the work of graduate student Di Kang.

3. In the interest in modeling the deformation process of large grain/single crystal samples, a crystal plasticity constitutive model for finite element simulation of deformation commenced. There has been some good progress, and this work continues. This is also challenging, due to the known non-Schmid nature of slip in body centered cubic metals that leads to significantly anisotropic yield surfaces. An important accomplishment in this effort is the development of a dynamic hardening rule that is necessary for single/large crystals, which recognizes the fact that when one slip system dominates the deformation process, dislocations will not accumulate because dislocations can enter and leave from the free surfaces. This model showed that the near-zero work hardening rate can be sustained until the crystal rotates to an orientation where slip on another slip system begins to be significant, but the increase in work hardening thereafter has not been sufficiently captured, and is the subject of continuing work (some progress in modeling this effect based upon physically meaningful processes has happened in the past couple months). The work on this topic commenced with graduate student Payam Darbandi, and continued with Aboozar Mapar, who made the progress described, notably on the dynamic hardening model, prior to the end of this grant. Mr. Mapar has since made

important progress, showing that the rotations are usually predicted correctly by the model, (there is work still to do on this issue), and the non-Schmid component has been installed in the model, and its refinement is still on the horizon.

4. Purchase and development of Laue camera facility: The grant provided funds to purchase and set up an area detector-based data acquisition system on our existing Laue camera, for characterization of grain orientations of large-grain materials and structures. This has enabled data acquisition in a manner similar to orientation imaging microscopy, but at a much larger scale needed to characterize the metallurgical state of large-grain Nb. This facility was used to characterize ingot slice orientations and their distribution. About 6 different ingots were characterized, which came from different suppliers, and the primary outcome of this study was to show that there is no pattern in preferred orientations or grain arrangements detected. This implies that production of sheet metal always starts with a random arrangement of large orientations. This important outcome is consistent with the observation that no two pieces of rolled sheet metal have the same microstructure/microtexture features, which probably contributes to the variability in cavity performance, at the least in indirect ways. On one ingot that was sliced like a board, milled, and then hand polished, the depth of surface damage was quantified by chemically removing the surface 100 µm, and the sharpness of diffraction patterns increased. This was documented in a semi-quantitative manner, and has been published. This work was completed by graduate student Di Kang.

5. Three collaborations with researchers at Jefferson Lab took place during this grant period, and two are ongoing: First, orientation imaging microscopy was analyzed and interpreted on samples with etch pits on the inner surface taken from a large grain cavity hot spot region in a collaboration with Gigi Ciovati and Xin Zhao. The surfaces were etched, revealing etch pits, which were analyzed with respect to possible dislocation slip system activity, and related to probable systems activated during forming. This work was published in PRST-AB in 2010, and this work commenced at the end of the grant prior to this one, and was completed at the beginning of this grant.

    Two efforts were made to metallurgically characterize single cell cavities at different stages of fabrication. In collaboration with Grigory Mendeleev, polycrystalline sheet was initially characterized in regions just outside of the equator and inside of the iris locations using material removed from the square sheet metal part. After forming, excess material was cut off using wire EDM, and surfaces close to the undeformed material were characterized, to determine how the texture changed. The measurements were made, but have yet to be deeply analyzed. The overall textures show that the gamma fiber was much more strongly developed in specific orientations in the iris than at the equator, consistent with the larger monotonic strain evolution in the iris.

    In a similar collaboration with Gigi Ciovati using large grain material, the orientation of crystals in the iris and equator regions were measured using the Laue camera, and the orientations were measured in selected parts of the material sliced off using EDM adjacent to the iris and equator. They were analyzed using orientation imaging microscopy, and synchrotron radiation experiments. These results show that the damaged layer is clearly in the range of 50-100 µm, but it varies with the crystal orientation. This supports the empirical practice of removing about 100 µm chemically from the inside of the cavity after assembly. There is clear evidence for much greater dislocation content adjacent to grain boundaries. Since the end of this grant, these pieces were annealed at 800 or 1000 °C, showing that there was little change in the equator and somewhat more change in the iris region, but no obvious recrystallization. This is surprising and indicates that recovery happened to a greater extent than recrystallization. This investigation is continuing. This result is also surprising in that recrystallization has been observed adjacent to equator welds in large grain cavities. Apparently the extremely rapid heating rate of welding facilitates recrystallization, whereas the furnace heating rate allows recovery to occur prior to reaching a recrystallization temperature. An outcome of this study (prior to the recent annealing work) led to annealing the cavity halves before welding, to determine if this helped cavity performance, but this has been inconclusive. This work has been done by Di Kang.

6. Collaboration with Jim Murphy at Univ. Nevada-Reno: Jim Murphy has been working on extremely high temperature conversion of polycrystalline tubes of Nb into single crystals. While there was some indication that this might work, the most recent tubes were large crystals, and not a single crystal. This was characterized using the Laue camera facility. This work was done by Di Kang.

7. Collaboration with K. Ted Hartwig and Shear Form, Inc.: There has been an ongoing effort to develop methods to homogenize the polycrystalline microstructure of Nb using equal channel angle processing at Shear Form, Inc., which has been funded via SBIR funds. They have provided some funding to MSU to characterize results of these studies, and Di Kang has done this characterization work. The outcomes are provided in the SBIR reports.

8. As there is ongoing interest in the possibility of hydroforming cavities from tubes, a tube hydroforming press that has been unused for several years was slowly brought back into operation. Several practice tubes of annealed aluminum and copper were bulged to demonstrate its ability to operate properly, and then a couple Nb tubes were tested. As expected, tubes that were merely welded from sheet metal, cracked near the weld after very little strain. An extruded tube provided by Shear Form also did not bulge very much, and this was due to the lack of uniform thickness. We have an ongoing collaboration with Shear Form, and will be testing tubes fabricated in a Phase II SBIR study in the future.

9. Initial assessment of dynamic mechanical analysis (DMA) on Nb single crystal samples shows promise for sensitive detection of defect state at temperatures below 250 °C. Using left over pieces between the single crystal tensile samples, preliminary studies of dynamic mechanical analysis were made with the assistance of Prof. Andre Lee at MSU. These studies show measureable changes in the internal defect content at about 120 °C, which may be correlated with important phenomena associated with the beneficial effects of the 120 °C bake. These results were shared with personnel at Fermi Lab, and they have focused on this phenomenon to a greater extent than we have.

5. Published Papers in which DOE support is acknowledged:

Nb Tubes for Seamless SRF Cavities, , S. Balachandran, R.C. Elwell, D. Kang, R.E. Barber, T.R. Bieler, K.T. Hartwig, IEEE Transactions on Applied Superconductivity 23(3), 7100904 (2013).

A. Mapar, T. R. Bieler, F. Pourboghrat, C. C. Compton, Crystal Plasticity Finite Element Modeling of Single Crystal Niobium Tensile Tests with Weighted Dynamic Hardening Rule, 2nd World Congress on Integrated

Computational Materials Engineering, pp. 255–258, TMS (The Minerals, Metals & Materials Society, 2013).

Hydrogen Saturation and the Thermal Conductivity of Superconducting Niobium, S.K. Chandrasekaran, T.R. Bieler, C. Compton, N.T. Wright, paper TUP067, Proceedings of SRF 2013, in press.

Quality Assurance and Acceptance Testing of Niobium Material for Use in the Construction of the Facility for Rare Isotope Beams (FRIB) at Michigan State University (MSU), Chris Compton, Doug Miller, Saravan Kumar Chandrasekaran, Neil T. Wright, Thomas R. Bieler, Di Kang, paper MOP033, Proceedings of SRF 2013, in press.

Phonon scattering in the thermal conductivity of large-grain superconducting niobium as a function of heat treatment temperature, S.K. Chandrasekaran, T.R. Bieler, C.C. Compton, N.T. Wright, AIP Conf Proc 1434, pp 976--982 (2012).

Characterization of Large Grain Nb Ingot Microstructure Using OIM and Laue Methods, D. Kang, T.R. Bieler, D.C. Baars, C. Compton, G. Ciovati, T.L. Grimm, A. Kolka, Paper THPO067, Proceedings of SRF2011, Chicago, IL USA (JACOW web site: http://accelconf.web.cern.ch/accelconf/index.html), p. 890, 2012.

Nb Tubes for Seamless SRF Cavities, , S. Balachandran, R.C. Elwell, D. Kang, R.E. Barber, T.R. Bieler, K.T. Hartwig, IEEE Transactions on Applied Superconductivity 23(3), 7100904 (2013).

Effect of Heat Treatment Temperature on the Thermal Conductivity of Large Grain Supercondcutng Niobium, S.K. Chandrasekaran , N.T. Wright, T.R. Bieler, C.C. Compton, Paper WEIOA06, Proceedings of SRF2011, Chicago, IL USA (JACOW web site: http://accelconf.web.cern.ch/accelconf/index.html), p. 593, 2012.

Comparison of the role of moderate heat treatment temperatures on the thermal conductivity of ingot niobium, S.K. Chandrasekaran, T.R. Bieler, C.C. Compton, W. Hartung and N.T. Wright, Symposium on the Superconducting Science and Technology of Ingot Niobium (SSTIN10), eds. G. R. Myneni, G. Ciovati, M. Stuart, American Institute of Physics, conference proceedings v. 1352, (2011), pp. 131-41.

Characterization of large grain Nb ingot microstructures using EBSP mapping and Laue camera methods, D. Kang, D.C. Baars, T.R. Bieler, C. Compton, Symposium on the Superconducting Science and Technology of Ingot Niobium (SSTIN10), eds. G. R. Myneni, G. Ciovati, M. Stuart, American Institute of Physics, conference proceedings v. 1352, (2011), pp. 90-99.

Comparison of the role of moderate heat treatment temperatures on the thermal conductivity of ingot niobium, S.K. Chandrasekaran, T.R. Bieler, C.C. Compton, W. Hartung and N.T. Wright, Symposium on the Superconducting Science and Technology of Ingot Niobium (SSTIN10), eds. G. R. Myneni, G. Ciovati, M. Stuart, American Institute of Physics, conference proceedings v. 1352, (2011), pp. 131-41.

Physical and mechanical metallurgy of high purity Nb for accelerator cavities, T.R. Bieler, N.T. Wright, F. Pourboghrat, C. Compton, K.T. Hartwig, D. Baars, A. Zamiri, S. Chandrasekaran, P. Darbandi, H. Jiang, E.

Skoug, S. Balachandran, G. E. Ice, and W. Liu, Physical Review Special Topics - Accelerators and Beams 13, 031002 (2010).

Characterization of Etch Pits Found on a Large-Grain Bulk Niobium Superconducting Radio-Frequency Resonant Cavity, Xin Zhao, G. Ciovati, T.R. Bieler, Phys. Rev. ST Accel. Beams 13, 124701 (2010) [11 pages].

Posters presented at SRF-13 in Paris (these will be published papers on JACOW site) (https://oraweb.cern.ch/pls/srf2013/search.html):

Surface Damage and Effects of Heat Treatment on Large Grain Nb Cavities, D. Kang, T.R. Bieler, C. Compton, Paper TUP016

Study of Slip in High Purity Single Crystal Nb for Accelerator Cavities, D. Kang, D.C. Baars, T.R. Bieler, C. Compton, Paper TUP017.

Dynamic Hardening Rule; A Generalization of the Classical Hardening Rule for Crystal Plasticity, A. Mapar, D. Kang, T. R. Bieler, F. Pourboghrat, C. C. Compton, paper TUP037.

Hydrogen Saturation and the Thermal Conductivity of Superconducting Niobium, S.K. Chandrasekaran, T.R. Bieler, C. Compton, N.T. Wright, paper TUP067.

Quality Assurance and Acceptance Testing of Niobium Material for Use in the Construction of the Facility for Rare Isotope Beams (FRIB) at Michigan State University (MSU), Chris Compton, Doug Miller, Saravan Kumar Chandrasekaran, Neil T. Wright, Thomas R. Bieler, Di Kang, paper MOP033.

Presentations (non-archived)

Predicting Deformation of Single Crystal Niobium Using Crystal Plasticity Finite Element Method, Aboozar Mapar, Thomas Bieler, Farhang Pourboghrat, Christopher Compton, TMS Annual Meeting, San Antonio, TX, March 4-7, 2013.

Combined Experimental and Computational Study on the Activity of Slip Systems in Single-Joint Tensile Deformation, Payam Darbandi, Farhang Pourboghrat, Thomas Bieler, Tae-Kyu Lee, TMS Annual Meeting, San Antonio, TX, March 4-7, 2013.

Predicting Deformation of Single Crystal Niobium Using Crystal Plasticity Finite Element Method, Aboozar Mapar, Thomas Bieler, Farhang Pourboghrat, Christopher Compton, TMS Annual Meeting, San Antonio, TX, March 4-7, 2013.

Characterizing Slip System Behavior in High Purity Nb for Accelerator Cavities, Di Kang, Derek Baars, Thomas Bieler, Chris Compton, TMS Annual Meeting, San Antonio, TX, March 4-7, 2013.

Microstructure Development in Seamless Nb Tube, Shreyas Balachandran, Roston Elwell, Di Kang, Thomas Bieler, Karl Hartwig, TMS Annual Meeting, San Antonio, TX, March 4-7, 2013.

Characterizing Dislocation Substructure in Large Grain Niobium for Accelerator Cavities, Di Kang, Derek Baars, Gianluigi Ciovati, Thomas Bieler, Chris Compton, MS&T Meeting, Pittsburgh, PA, 9 Oct 2012.

Mechanical and Physical Metallurgy of High Purity Niobium used for Superconducting Radio Frequency Cavities for Particle Accelerators, **Invited,** Thomas Bieler, Di Kang, Saravan Chandrasekaran, Aboozar Mapar, Gianluigi Ciovati, Pashupati Dhakal, Ganapati Myneni, Neil Wright, Farhang Pourboghrat, Chris Compton, MS&T Meeting, Pittsburgh, PA, 9 Oct 2012.

Crystal Plasticity Finite Element Modeling of Deformation of Single Crystal Niobium, Aboozar Mapar, Thomas Bieler, Farhang Pourboghrat, Christopher Compton, MS&T Meeting, Pittsburgh, PA, 9 Oct 2012.

Refining Crystal Plasticity Finite Element Model for Deformation of Single Crystal Niobium, A. Mapar, T.R. Bieler, F. Pourboghrat, C.C. Compton, Jefferson Lab, SRF Materials Workshop, Newport News, VA, July 16-17, 2012.

Recovery of Phonon Peak in Annealed Niobium as a Function of Initial Strain and Hydrogen Concentration S.K. Chandrasekaran, T.R. Bieler, C.C. Compton, N.T. Wright, SRF Materials Workshop, Jefferson Lab, Newport News, VA, July 16-17, 2012.

Examination of surface damage layer on SRF cavities using EBSD, Kang Di, G. Ciovati, T.R. Bieler, C. Compton, SRF Materials Workshop, Jefferson Lab, Newport News, VA, July 16-17, 2012.

The Effect of Hydrogen Saturation on the Phonon Peak in Thermal Conductivity of Superconducting Large Grain Niobium, Saravan K. Chandrasekaran and Neil T. Wright, Thomas R. Bieler, Christopher Compton, 18th Symposium on Thermophysical Properties, June 24-29, Boulder CO.

The Role of Heat Treatment Temperature on the Thermal Conductivity of Superconducting Niobium, S. K. Chandrasekaran, T. R. Bieler, C. C. Compton, and N. T. Wright, Cryogenic Engineering Conference & International Cryogenic Materials Conference (CEC/ICMC) Spokane, WA, June 13-17, 2011.

Stress-Strain Behavior of Nb Single Crystal Tensile Specimens with Different Grain Orientations, Di Kang, Derek Baars, Aboozar Mapar, Payam Darbandi, Thomas Bieler, Farhang Pourboghrat, Chris Compton, TMS Annual Meeting, Orlando, FL March 11-15, 2012.

From Ingot to Polycrystal Sheet; Origins of Heterogeneous Microstructure, Texture, and Deformation in Pure BCC Metals, D.C. Baars, D. Kang, A. Mapar, I. Jarvis, M.A. Crimp, F. Pourboghrat, C. Compton, T.L. Grimm, T.R. Bieler 18th International Symposium on Plasticity and its Current Applications, San Juan, Puerto Rico, Jan 3-8, 2012.

Single crystal defect properties and tensile behavior of high purity Nb from Nb ingot slice, D.C. Baars, P. Darbandi, D. Kang, F. Pourboghrat, T.R. Bieler, C. Compton, Symposium on the Superconducting Science and Technology of Ingot Niobium (SSTIN10), Thomas Jefferson National Accelerator Facility, Newport News, VA, September 22-24, 2010.

Thermal conductivity of ingot niobium - estimating with processing history, S.K. Chandrasekaran, T.R. Bieler, C.C. Compton, W. Hartung and N.T. Wright, Symposium on the Superconducting Science and Technology of Ingot Niobium (SSTIN10), Thomas Jefferson National Accelerator Facility, Newport News, VA, September 22-24, 2010.

Nondestructive measurement of grain orientations and orientation gradients in Nb ingot slices, Di Kang, T.R. Bieler, D.C. Baars, C. Compton, Symposium on the Superconducting Science and Technology of Ingot Niobium (SSTIN10), Thomas Jefferson National Accelerator Facility, Newport News, VA, September 22-24, 2010.

Needed material property studies to enable optimization of hydroforming, (Invited) T.R. Bieler and F. Pourboghrat, US Hydroforming Workshop, Fermilab, Batavia, IL, 1st Sep 2010.

Physical and Mechanical Metallurgy of High Purity Nb for Cavity Fabrication, (Invited) T.R. Bieler, D.C. Baars, N.T. Wright, C.C. Compton, Argonne National Laboratory, July 23, 2010.

Comparison of Optimized Finite Element Crystal Plasticity Model and Tensile Tests of Niobium Single Crystals, Derek Baars, Payam Darbandi, Chris Compton, Wenjun Liu, Rozaliya Barabash, Thomas Bieler, TMS Annual Meeting, Seattle, WA, Feb. 15, 2010.

Deformation Mechanism for Polycrystal Niobium at Cryogenic Temperature, Payam Darbandi, Derek Baars, Saravan Chandrasekaran, Farhang Pourboghrat, Tom Bieler, Chris Compton, TMS Annual Meeting, Seattle, WA, Feb. 15, 2010.

6. List of people working on this research activity

Saravan Chandrasekaran was fully supported on this project, focusing on thermal conductivity studies and developing a modeling paradigm for identifying the components that affect the thermal conductivity in Nb.

Di Kang, graduate student working with Thomas R. Bieler, is mostly supported on this project. He is also supported by a small subcontract with Shear Form, Inc. to characterize microstructures of samples that have been severely plastically deformed using methods that will hopefully enable reproducible fabrication of cavities via hydroforming. He has also been partially supported by FRIB to do characterization of incoming lots of niobium used to fabricate the FRIB accelerator.

Aboozar Mapar, graduate student jointly advised by Farhang Pourboghrat and Thomas R. Bieler was supported by this award. Prior to Aboozar's arrival, Payam Darbandi got the modeling process for BCC metals started.

Chris Compton provided organizational and coordination support to enable practical guidance and infrastructural support from the perspective of materials research needs for accelerator science.

Thomas R. Bieler, Neil T. Wright, and Farhang Pourboghrat provide guidance for the three graduate students working on this project.

7. **Unexpended funds:** All funds were spent.