Niobium is the material of choice for superconducting radio frequency (SRF) cavity programs in different particle accelerator laboratories because of its mechanical properties favorable for formability, machinability and also high $T_C$ and high first flux penetration field $H_{C1}$. SRF cavity performances have been continually improved for past three decades to achieve reproducible quality factor of $10^{10}$ and accelerating fields ($E_{acc}$) of 30-35MV/m. The present approach for the fabrication of superconducting radio frequency (SRF) cavities is to roll and deep draw sheets of polycrystalline high-purity niobium. Jefferson Laboratory pioneered the use of large-grain/single-crystal Nb directly sliced from an ingot for the fabrication of single-crystal high-purity Nb SRF cavities [1]. The large grain/single crystal niobium has several potential advantages over the polycrystalline niobium as discussed in Ref. [2] and has become a viable alternative to the standard fine grain (ASTM grain size>6 µm), high purity (RRR $\geq$ 250) niobium for the fabrication of high-performance SRF cavities for particle accelerators. The other alternative approach of cavity fabrication is the niobium – coated copper cavities pioneered by CERN [3]. In many laboratories this path of coating technology is pursued as an alternative to bulk niobium technology [4].

Niobium metal superconductivity in SRF accelerators is a nanoscale, near surface phenomena because of small RF penetration depth of the order of 10 – 100 nm. The cavity performance improvement over the past 3-4 decades strongly indicates the topography and the chemistry of the inner surface impacts the final cavity performance [5].

Present day cavity preparation chemistry for the inner surface follows two different routes namely buffered chemical polishing (BCP) and electropolishing (EP). The BCP solution is
prepared with 1:1:1 or 1:1:2 (volume) mixture of HNO₃(69%), HF(48%) and H₃PO₄ (85%) at 15-20 °C. The large grain or single crystal material after BCP produces very smooth and shiny surfaces; the measured surface roughness was 50 times better then the BCP’d polycrystalline niobium [6]. In the present thesis the studies are carried out with BCP surface treatment and EP route is not explored.

It has been known for quite some time that the low temperature baking (LTB) (100 – 140 °C) is a necessary final preparation stage to achieve high accelerating gradients in niobium RF cavity [7, 8]. To date there exists different models to understand the LTB effect on niobium RF cavity, although none of them is complete in a sense that they can explain all the experimental observations. At present there is an oxygen diffusion model [9] which takes into account the oxygen diffusion with LTB resulting in dilution of oxygen pollution over the penetration depth. Over the time as research progressed it has been found that there are other possibilities which can explain Q-drop and baking effect namely the mechanisms based on hot-spot [10], dislocations [11], interface tunnel exchange [12] and magnetic impurities [13]. A present study of B. Visentin [14] on niobium samples with positron annihilation spectroscopy has shown that there is an increase of vacancy site with the LTB in a length scale of about 120 nm from the surface and as explained this might be due to the dissociation of hydrogen-vacancy complexes by baking followed by hydrogen diffusion.

An attempt has been made to study the material and superconducting properties of large grain niobium of different RRR value. In the present study the large grain samples are treated with BCP followed by different post purifications such as 600°C annealing, low temperature baking (LTB) in the range of 100 – 140 °C. To understand the low temperature baking improvement of cavity performance experimental studies of surface magnetization are carried out.
Also the study of positron annihilation spectroscopy shows a correlation of defect density dependence before and after annealing.

The performance of SRF cavity is mostly described by the dependence of the unloaded quality factor $Q_0$ with accelerating electric field $E_{acc}$. Several past experiments had shown that there is a continuous degradation of $Q_0$ with $E_{acc}$ (or peak magnetic field $B_p$) in the range of 20 – 100 mT. This phenomenon is termed as ‘medium field Q-slope’. The present study includes the prototype single cell low beta cavity design, fabrication, EB welding and low temperature RF test at 2K. In this study the medium field Q-Slope has been analyzed with the help of an added non linear term in Heabel’s analytical model [15] and an additional linear term for the BCS surface resistance $R_s$ [16].

**Chapter 1: Basics of Superconductivity and Superconducting Radio Frequency Cavity**

Superconductivity in mercury below $T_c = 4.2K$, was discovered in 1911 by Kammerlingh Onnes in Leiden. Niobium is the material of choice for SRF cavities as its properties are favorable for formability, machining and also high $T_C$ and high first flux penetration field $H_{C1}$. This chapter introduces the definitions of characteristic length scale of superconductivity, magnetization behavior, thermal conductivity dependence in superconducting state [17] and also discusses the phenomenological Ginzburg-Landau theory of superconductivity.

The next part of this chapter discusses the basics of SRF cavities and different structure types with different $\beta$ ($v/c$) values. Then the figure of merits for superconducting cavity such as accelerating gradient, quality factor $Q_0$, shunt impedance, peak electric and magnetic field are described. The deviations of ideal behavior of SRF cavity is attributed to different loss
mechanisms. The surface resistance is greater than the BCS resistance due to the residual resistance. The surface resistance variation with increasing accelerating gradient causes low field [18], medium field Q slopes and high field Q drop [19, 20, 21, 22] before it reaches the theoretical quench field. A detailed discussion of models of low and medium field Q-slope is presented, considering these two slopes are relevant for our studies of low $\beta$ cavities.

Chapter: 2 Motivation for the Present Research

Because of potential advantages of large grain material, the interest in investigating material properties with different surface treatments will help to understand the cavity performance behavior with the same surface preparation. This thesis presents results of investigations on following material properties and the cavity behavior,

- Thermal Properties – thermal conductivity, phonon peak and the effect of different processing and trapped vortices on the thermal conductivity of niobium ingot.
- Magnetic properties – $H_{c1}$, $H_{c2}$, $H_{c3}$ and changes in London penetration depth measured with different surface treatments.
- Positron annihilation studies to find defect density after different processing.
- Simulation of surface magnetization behavior before and after LTB.
- Mechanical properties – yield strength, tensile strength, and elongation that help to form the cavity.
- SRF cavity development with large grain niobium- design of cavity, EB-welding, surface treatments, room temperature RF characteristics and testing at 2K and medium field Q-slope analysis.
The electrons which have condensed into Cooper pairs do not contribute to any disorder or entropy transport anymore. The remaining fraction of electrons which contribute to heat transport decreases exponentially with decreasing temperature. Experimental thermal conductivity results are fitted with Bardeen-Rickayzen-Tewordt (BRT) theory. The important observation is that the superconducting gap energy does not change with the annealing of chemically polished large grain samples. But the decrease of lattice constant $a_0$ implies that hydrogen comes out from the bulk of the material on annealing, which is otherwise trapped in the tetrahedral positions increasing the lattice parameter. The measurements clearly show the presence of a phonon peak at around 2K. One important observation is that the phonon peak is eliminated by the presence of trapped vortices due to the strong scattering of phonons with vortex cores. When the vortices are trapped inside the sample, the fitting parameters indicate a reduction of the gap energy $\alpha$ due to the low energy excitations having very small energy gap $\sim \Delta_0^2/E_F$ close to the vortex core. Also the effective number of conduction electrons decreases due to the bound excitations in the vortex cores. The dependence of the thermal conductivity with the applied magnetic field for the samples with and without trapped vortices show the same $H_{C1}$ and $H_{C2}$ values as from the magnetization measurement. Finally when the temperature of the samples is cycled above $T_c$, the thermal conductivity measured for the sample in absence of an applied magnetic field is restored. The temperature dependence of the thermal conductivity at low temperature and low magnetic field agrees qualitatively with the model of Vinen et al [23]. In the vicinity of $H_{C2}$ the thermal conductivity agrees quite well with Houghton-Maki [24] theory for zero-field cooled samples. But if there is initial flux trapped within the sample, the measured
thermal conductivity deviates from the Houghton-Maki theory and observed an increase in thermal conductivity in the range of $0.2 \leq \mu \leq 0.6$. Where $\mu$ is the transport coefficient dependent on the reciprocal lattice vector $k_c$, Fermi velocity $v_F$ and the electron mean free path $l_e$.

**Chapter: 4 Low Temperature Baking Effect on Bulk Magnetization, Penetration Depth and Surface Magnetization**

The measurement of DC magnetization provides the first flux penetration value $H_{C1}$, the upper critical magnetic field $H_{C2}$ and from M-H curve the thermodynamic critical field $H_C$ is calculated. Pinning of vortices leads to irreversible magnetization curves. By connecting a small pick-up coil around the sample rod as part of a L-C oscillator, it is possible to measure the changes of the penetration depth as a function of the applied DC magnetic field by measuring the changes of the oscillator’s resonant frequency $f_0$ (the base frequency is 270 kHz, sampling up to a depth $\sim 10 \mu m$) while slowly ramping up and down the magnetic field above $H_{C3}$. This method, which was applied for Nb surface studies, provides information about surface pinning and allows measuring the surface critical field $B_{c3}$. The irreversibility of the curve between $B_{c1}$ and $B_{c2}$ is an indication of surface pinning. The result shows that with the increased baking temperature the contaminated layer thickness increases to an average of 5.8 nm, 9.5 nm and 19.6 nm at 100 °C, 120 °C, and 140 °C baking temperature respectively. But at 140 °C both the contaminated layer thickness and bulk $H_{C2}$ increases which may be due to the partial dissociation of the Nb$_2$O$_5$ layer as explained in the oxygen diffusion model. The oxygen diffusion model corresponds to a diffusion depth of 7.6 nm, 19 nm and 40 nm for a baking at 100 °C, 120 °C, and 140 °C for 12 hour duration. Except for the 100 °C bake the experimentally derived surface layer thickness is about half of the oxygen diffusion depth.
calculated by the theoretical model. Also the first flux penetration i.e $B_{C1}$ increases with the 100 °C and 120 °C baking for 12 hr. in vacuum. But it falls to 100 mT at 140 °C baking. The data has been analyzed with the Schmidt’s model [25] taking into account the low temperature correction to $H_{c3}/H_{c2}$ ratio in the clean limit. The enhanced $H_{c3}/H_{c2}$ ratio has been interpreted as due to the increased contaminated surface layer thickness. This fact is further corroborated by the penetration depth measurement.

Chapter: 5  Defect Depth Profiling by Positron Annihilation Spectroscopy

Defect depth profiles were studied by slow positron implantation spectroscopy using a 50 mCi, 22Na positron source. In the defect depth profiling study the positron energy E is varied from 0.2 to 20.2keV. The line shape parameter or the S-parameter dependence on positron energy is fitted by the VEFIT software package. Large grain niobium specimen (square shaped, 1cm by 1 cm, thickness 3mm) was first chemically polished with a mixture of HF, HNO$_3$, H$_3$PO$_4$ in a volumetric ratio of 1:1:1. About 180 μm is removed from the outer surface of the specimen. Then we performed the defect depth profile study with the positron beam. In the second phase of the experiment the sample was heat treated at 600 °C for 10 hour in a vacuum of the order of 10$^{-5}$ Torr. This was done to degas the hydrogen from the sample. Then we carried out the positron implantation spectroscopy measurements following this treatment. It has been observed that after annealing the defect density increases at the surface layer. This might be due to the hydrogen degassing which when comes out of the specimen leaves open volume defect within the sample. The three layer fitting of the chemically polished sample showed a surface layer of 79 nm thick has the diffusion length of 65 nm, from 79 nm to 100nm layer has a diffusion length of 0.5 nm and the bulk of the sample have a diffusion depth of 7.4 nm. After annealing it shows a surface
layer of 10 nm with diffusion length 39.4 and the bulk is having a diffusion length of 139 nm. The important observation is that after annealing the defect density at the surface increases whereas in the bulk the defect density decreases.

Chapter: 6  Design, Fabrication and RF Test of $\beta = 0.49$, $f = 1050$ MHz
Elliptical Cavity

The first part of the chapter presents the experimental setup and mechanical measurements of large grain niobium samples. Mechanical properties such as yield strength, tensile strength and elongation were measured with different post-purification of as received material.

The second part of the chapter presents the design, fabrication, electron beam welding and low temperature RF testing of $\beta = 0.49$, $f = 1050$ MHz fine grain and large grain cavities. Two single cell cavities of frequency 1050 MHz, $\beta =$0.49 have been designed, fabricated and tested at 2 K. One was made of polycrystalline niobium and the other one was made of large grain niobium. Both the cavities exhibited an unloaded Q value of $1 \times 10^{10}$ and reached the design value of 5 MV/m for the low $\beta$ section of the proton linac. The large grain cavity was quenching at 6 MV/m at 2K and several areas with many pits on the surface were visible. The experimental value of Lorentz force detuning coefficient was significantly high and suggests the need for a stiffening ring for a multi-cell structure. The medium field Q-slope analysis showed a significantly higher value of $\gamma$ in the large grain cavity compare to the fine grain cavity although both the cavities were subjected to similar surface treatments before the RF test. The factor $\gamma$ depends on RBCS, Kapitza raistamne Rk, thermal conductivity K and the wall thickness d. The higher value of $\gamma$ was most likely related to higher RF losses at the pit’s locations.
Chapter: 7  Summary and Conclusions

Based on the above studies the following conclusions have been made.

1. High purity niobium mechanical properties vary from batch to batch and are very sensitive to various treatments and handling.

2. Elastic behavior of high purity niobium remains unchanged with different heat treatments only a change in yield strength is observed after 1200°C heat treatment.

3. The thermal conductivity measurement of large grain niobium samples in the Meissner state is well described by the model of Ref. [15] within the experimental error of ±6%.

4. One important observation is that the phonon peak is eliminated by the presence of trapped vortices due to the strong scattering of phonons with vortex cores.

5. The dependence of the thermal conductivity with and without trapped vortices show the same $H_{C1}$ and $H_{C2}$ values in the magnetization measurement.

6. The temperature dependence of the thermal conductivity at low temperature and low magnetic field agrees qualitatively with the model of Vinen et al. In the vicinity of $H_{C2}$ the thermal conductivity agrees quite well with Houghton-Maki theory for the zero-field cooled samples. But if there is initial flux trapped within the sample, the measured thermal conductivity deviates from the Houghton-Maki theory and it was observed an increase in thermal conductivity in the range of $0.2 \leq \mu \leq 0.6$.

7. The bulk properties of the samples such as $T_C$, $B_C$, $H_{C1}$ and $H_{C2}$ were essentially unchanged with surface treatments such as BCP, 600°C annealing, LTB.
8. Surface pinning measurement shows that the $H_{C3}$ value increases with the increased LTB temperature. It also shows that the surface $H_{C1}$ is lower than the bulk $H_{C1}$ and that the highest surface $H_{C1}$ was obtained after baking at 120°C for 12 hour.

9. The irreversibility between $H_{C1}$ and $H_{C2}$ decreases significantly with the low temperature baking, thus a reduction in the surface pinning centers can be inferred.

10. The increase of $H_{C3}/H_{C2}$ ratio in the LTB regime has been analyzed with the Schmidt’s model taking into account the low temperature correction to $H_{C3}/H_{C2}$ ratio. The enhanced $H_{C3}/H_{C2}$ ratio has been interpreted as the increased contaminated surface layer thickness. This fact is further corroborated by the penetration depth measurement.

11. The important observation in the positron annihilation studies (PAS) is that the surface defect density increases after the annealing whereas in the bulk the defect density decreases. This is complimentary to the conclusion inferred with the magnetization measurement.

12. RF Properties of 1050 MHz, $\beta = 0.49$ single cell Elliptical cavity has been studied by means of 2D cavity tuning code SUPERFISH and 3D High Frequency Simulation code CST Microwave Studio. Electromagnetic properties of the optimized cavity shape carried out on 2D code SUPERFISH are compared with 3D code CST Microwave Studio. Numbers of mesh cells are optimized by the adaptive mesh solver technique. It has been observed that the mode frequency remains almost constant for more than $10^5$ mesh cells.

13. Two single cell cavities of frequency 1050 MHz, $\beta = 0.49$ have been designed, fabricated and tested at 2 K. One was made of polycrystalline niobium and the other one was of large grain niobium. Both the cavities exhibited an unloaded Q value of $1 \times 10^{10}$ and reached the design value of 5 MV/m for the low $\beta$ section of the proton linac.
14. The medium field Q-slope analysis showed a significantly higher value of $\gamma$ in the large grain cavity compare to the fine grain cavity although both the cavities were subjected to similar surface treatments before the RF test. The higher value of $\gamma$ is unusual for this type of material and was most likely related to higher RF losses at the pits’ locations.

REFERENCES


Publications during Ph.D work

**Journal Publications**


**Conference Publications**

