# **THERMODYNAMIC PROPERTIES OF SRF NIOBIUM**<sup>∗</sup>

P. Dhakal† , Thomas Jefferson National Accelerator Facility, Newport News, VA, USA S. Li, Quantum Design, San Diego, CA, USA

# *Abstract*

Bulk and thin films of niobium are the materials of choice in fabricating superconducting radio frequency (SRF) cavities for modern particle accelerators and quantum computing applications. The thermodynamic properties of Nb are of particular interest in heat management in cryogenic environments. Here, we report the results of measurements of the thermodynamic properties of niobium used in the fabrication of SRF cavities. The temperature and magnetic field dependence of thermal conductivity, Seebeck coefficient, and specific heat capacity was measured on bulk niobium samples.

# **INTRODUCTION**

Modern particle accelerators rely on superconducting radio-frequency (SRF) cavities to accelerate the charged particles as these SRF cavities can store energy with little dissipation [1]. Primarily made of bulk niobium (Nb), the state-of-the art processing techniques pushed the ultimate performance of these SRF cavities towards the theoretical limit on accelerating gradient set by the superconducting super-heating critical field. In addition, recent advances in the cavity processing technique of surface engineering with material doping led to exceptionally high quality factor resulting in very low RF power dissipation on cavity walls [2–6]. Ideally, the quality factor of SRF cavities is independent of the accelerating gradient until breakdown occurs when the cavity quenches. The quench of SRF cavities refers to the loss of the superconducting state by warming up a part of cavity above the critical temperature or by exceeding the superconducting critical field.

Quench event occurs at weak superconducting regions or at normal conducting defects on the cavity surface. The efficient transfer of the dissipated power to the helium bath is critical in restoring the cavity to the superconducting state and reliable operation of SRF-based accelerators. The thermodynamic properties such as resistivity, thermal conductivity, and thermal diffusivity are material dependent and the characterization of these properties is necessary to understand the performance limitation [7]. In this contribution, we have measured resistivity, thermal conductivity, specific heat cavity, and Seebeck coefficient of SRF Nb. The temperature and magnetic field dependence of thermal conductivity and heat capacity is also measured.

# **SAMPLE PREPARATIONS AND EXPERIMENTAL SETUP**

Rectangular bar sample of  $8 \times 1 \times 1$  mm<sup>3</sup> for resistivity, thermal conductivity, and Seebeck coefficient and a sample of ∼ 3×3×1 mm<sup>3</sup> weighing 82.4 mg are cut by a wire electrodischarge machining from a large grain niobium ingot sheet. The samples were treated with buffer chemical polishing (BCP) to remove  $~50 \mu m$  from the surface. The measurements were carried out at the Quantum Design facility using the physical properties measurement system [8].

## **EXPERIMENTAL RESULTS**

#### *Resistivity*

The dc resistivity of the sample was measured using the 4 probe method with a constant current of 1.5 mA. Figure 1(a) shows the temperature dependence resistivity with transition temperature  $~\sim$  9.3 K. The room temperature resistivity is measured to be 153 n $\Omega$ -m with a residual resistivity ratio (RRR) of 154.



Figure 1: Temperature dependence of (a) resistivity, (b) Seebeck coefficient, and (c) thermal conductivity.

This manuscript has been authored by Jefferson Science Associates, LLC under U.S. DOE Contract No. DE-AC05-06OR23177.

<sup>†</sup> dhakal@jlab.org

## *Seebeck Coefficient*

Figure 1 (b) shows the temperature dependence of the Seebeck coefficient of Nb which is the measure of induced thermometric voltage as a result of the temperature gradient across the metal. The Seebeck coefficient increases with the increase in temperature with peak values ∼ 75 K. The measured values are in agreement with previously reported SRF grade niobium [9]. The minimization of thermal current induced during the SRF cavity is needed to minimize the trapping of flux generated by the thermal current, mostly in bimetallic cavities such as Nb/Cu  $[10, 11]$ , Nb<sub>3</sub>Sn/Nb  $[12]$ ,  $Nb<sub>3</sub>Sn/Nb/Cu$  [13] and also in the helium tank and Nb creating a bimetallic junction [14]. The thermometric voltage is proportional to the difference in the Seebeck coefficient of bimetallic structure.

#### *Thermal Conductivity*

The thermal conductivity of SRF niobium has been extensively studied in the past to understand the effect of material and processing steps on the performance of SRF cavities [15, 16]. Figure 1 (c) shows the temperature dependence of thermal conductivity measured in our current study. The thermal conductivity of niobium depends on the grain size, grain boundaries, deformations, dislocations, and impurities. As shown in Fig. 1(c) inset, the thermal conductivity increases with the increase in temperature due to the increase in quasi-particles. The thermal conductivity below 4.0 K strongly depends on the metallurgical state of the sample. Generally, the sample with large grain and single crystal with low dislocations density shows a peak around 2.0 K before it decreases [17–19]. No phonon peak was observed in this measurement although the sample was machined from a large grain ingot without any high temperature annealing. The absence of a phonon peak is likely due to the large dislocation density present in the ingot sheet and hydrogen loading during the machining [18].

Figure 2 shows the thermal conductivity as a function of the applied dc magnetic field parallel to the length of the Nb sample at two different temperatures 2.0 and 5.0 K. The thermal conductivity remains constant at a low magnetic field up to the field of first flux penetration  $(H_{ffp})$  and it increases to the normal state value as the magnetic field approaches to upper the critical field ( $H_{c2}$ ). At 2.0 K, the  $H_{ffp}$  and  $H_{c2}$ was measured to be 185 and 390 mT, respectively which is in agreement with the values reported earlier in magnetization measurements [17]. In literature,  $\kappa(H)$  showed a dip when the H >  $H_{ffp}$ , as a result of additional scattering of fluxoids with the quasi-particles [20]. However, no noticeable dip near  $H_{ffp}$  was observed in this study.

### *Heat Capacity*

Figure 3 shows the heat capacity as a function of temperature at different external applied magnetic field. The magnetic field was applied perpendicular the sample surface. The heat capacity increase with temperature and the drop in

# **SRF Technology**

**Nb material**



Figure 2: Thermal conductivity as a function of dc magnetic field at temperatures 2.0 K and 5.0 K.

heat capacity corresponds to the temperature at which the sample transitions to a normal state.



Figure 3: Heat capacity as a function of temperature at different dc magnetic fields.

Figure 4 shows the heat capacity as a function of the external applied magnetic field at 2.05 K. The heat capacity in the Meissner state remains constant and increases when the field starts to penetrate and remains constant when the samples transition to normal conducting state above  $H_{c2} \sim$ 390 mT. In a type-II superconductor, the magnetic field penetrates in the form of vortices, and the contribution to the heat capacity comes from the finite density of states within the the vortex cores and the outside core of the vortices in the regions of  $\lambda > r > \xi$ , where r is the radius of vortex core.

### **DISCUSSION**

The resistivity, thermal conductivity, Seebeck coefficient, and heat capacity were measured on Nb samples used in SRF cavity fabrications. The results are in consistent with the previously measured thermodynamics properties of Nb.

Content from this work may be used under the terms of the CC BY 4.0 licence (© 2023). Any distribution of this work must maintain attribution to the author(s), title of the work, publisher, and DO

ቴ terms £

ŝ

്മ

from this work may

Content

CC BY 4.0 licence (© 2023). Any distribution of this work must maintain attribution to the author(s), title of the work, publisher, and DOI



Figure 4: Heat capacity as a function of external dc magnetic field at 2.05 K.

The present study is extended to magnetic field dependence of thermal conductivity and heat capacity, which has a particular interest in SRF cavities when the performance of the SRF cavities is limited by quench due to magneto-thermal instability.

For a long time, researchers have been using high purity Nb in order to fabricate SRF cavities, which is measured by the RRR of the material. One of the reasons for using high RRR Nb is to achieve high thermal conductivity at operating temperature. The study showed that the thermal conductivity of Nb at low temperature  $(< 4 K)$  is dominated by phonons which are mainly scattered by grain boundaries and dislocation networks. Thus, using larger grain Nb (large phonon mean free path) over fine grain Nb is being explored in fabrication of SRF cavities [21, 22]. In recent years, the magnetic field dependence of thermal conductivity is being considered as a switching technique in low temperature electronic superconducting circuits and quantum sensors [23].

Thermal diffusivity of the material is an important parameter in heat transfer when rf power is dissipated on the cavity surface operating in the GHz frequency range. Thermal diffusivity measures the rate of heat transfer from hot to cold, whereas thermal conductivity is the measure of materials ability to transfer heat from hot to cold. Figure 5 shows the thermal diffusivity of niobium in the current study and compared with previous work. Also plotted the thermal diffusivity of the superconducting  $Nb<sub>3</sub>Sn$  and some common metals, copper and tungsten [25]. The thermal diffusivity of Nb in superconducting state is lower than most common metals but higher than superconducting  $Nb<sub>3</sub>Sn$ .

As mentioned, the Seebeck coefficient allows us to characterize the effect of thermal current-induced magnetic field when bi-metallic structures are involved in SRF cavity fabrications. On some occasions, the thermal current is generated in Nb cavities during cavity quench [26]. The thermo-current induced magnetic field is trapped in the form of vortices resulting in a higher rf loss.



Figure 5: Temperature dependence of thermal diffusivity of Nb along with  $Nb<sub>3</sub>Sn$  (taken from Ref. [24] and reference therein), high purity copper, and tungsten [25].

# **SUMMARY**

We presented the thermodynamic properties of SRF Nb subjected to chemical polishing. The thermodynamic properties influence the RF performance of SRF cavities at cryogenic temperature. Future studies will be focused on investigation of such properties with respect to other processing techniques applied to SRF cavities.

## **ACKNOWLEDGEMENTS**

We would like to acknowledge Dr. Aashish Chourey at Quantum Design for his assistance. We would like to acknowledge Dr. Gianluigi Ciovati and Dr. Ganapati Rao Myneni for several discussions on the research topic.

#### **REFERENCES**

- [1] H. Padamsee,J. Knobloch, T. Hays, *RF Superconductivity for Accelerators*, New York, NY, USA: J. Wiley & Sons, 1998.
- [2] P. Dhakal *et al*., "Effect of high temperature heat treatments on the quality factor of a large-grain superconducting radiofrequency niobium cavity", *Phys. Rev. Spec. Top. Accel. Beams*, vol. 16, p. 042001, 2013. doi:10.1103/PhysRevSTAB.16.042001
- [3] P. Dhakal, "Nitrogen doping and infusion in SRF cavities: A review", *Phys. Open*, vol. 5, p. 100034, 2020. doi:10.1016/j.physo.2020.100034
- [4] A. Grassellino *et al.*, "Unprecedented quality factors at accelerating gradients up to  $45 \,\mathrm{MVm^{-1}}$  in niobium superconducting resonators via low temperature nitrogen infusion", *Supercond. Sci. Technol.*, vol. 30, p. 094004, 2017. doi:10.1088/1361-6668/aa7afe
- [5] P. Dhakal, S. Chetri, S. Balachandran, P. J. Lee, and G. Ciovati, "Effect of low temperature baking in nitrogen on the performance of a niobium superconducting radio frequency cavity", *Phys. Rev. Accel. Beams*, vol. 21, p. 032001, Mar. 2018.

doi:10.1103/PhysRevAccelBeams.21.032001

**SRF Technology Nb material**

- [6] E. M. Lechner, J. W. Angle, F. A. Stevie, M. J. Kelley, C. E. Reece, and A. D. Palczewski, "RF surface resistance tuning of superconducting niobium via thermal diffusion of native oxide", *Appl. Phys. Lett.*, vol. 119, no. 8, p. 082601, Aug. 2021. doi:10.1063/5.0059464
- [7] P. Dhakal, G. Ciovati, and G. R. Myneni, "Role of thermal resistance on the performance of superconducting radio frequency cavities", *Phys. Rev. Accel. Beams*, vol. 20, p. 032003, 2017. doi:10.1103/PhysRevAccelBeams.20.032003
- [8] Quantum Design, Physical Property Measurement System, https://www.qdusa.com/products/ppms.html
- [9] J. -M. Vogt, O. Kugeler, and J. Knobloch, "High-Q operation of superconducting rf cavities: Potential impact of thermocurrents on the rf surface resistance", *Phys. Rev. Spec. Top. Accel. Beams*, vol. 18, p. 042001, 2015. doi:10.1103/PhysRevSTAB.18.042001
- [10] T. Junginger, "Field dependent surface resistance of niobium on copper cavities", *Phys. Rev. Spec. Top. Accel. Beams*, vol. 18, p. 072001, 2015. doi:10.1103/PhysRevSTAB.18.072001
- [11] P. Dhakal and G. Ciovati, "Effect of cooldown and residual magnetic field on the performance of niobium–copper clad superconducting radio-frequency cavity", *Supercond. Sci. Technol*, vol. 31, p. 015006, 2018. doi:10.1088/1361-6668/aa96f5
- [12] S. Posen and D. L. Hall, " $Nb<sub>3</sub>Sn$  superconducting radiofrequency cavities: fabrication, results, properties, and prospects", *Supercond. Sci. Technol.*, vol. 30, p. 033004, 2017. doi:10.1088/1361-6668/30/3/033004
- [13] G. Ciovati et al., "Development of a prototype superconducting radio-frequency cavity for conduction-cooled accelerators", *Phys. Rev. Accel. Beams*, vol. 26, no. 24, p. 044701, 2023. doi:10.1103/PhysRevAccelBeams.26.044701
- [14] J.-M. Köszegi and J. Knobloch, "Advanced study of the thermoelectrically generated magnetic field in a nine-cell cavity during the superconducting phase transition", *Phys. Rev. Accel. Beams*, vol. 22, p. 052001, 2019. doi:10.1103/PhysRevAccelBeams.22.052001
- [15] P. Xu, "Investigation of the Thermal Transport in Superconducting Niobium and Tantalum", Ph.D. thesis, Michigan State University, MI, USA, 2019.
- [16] S. K Chandrasekaran, "Role of metallurgy in the thermal conductivity of superconducting niobium", Ph.D. thesis, Michigan State University, MI, USA, 2013.
- [17] P. Dhakal, G. Ciovati, P. Kneisel, and R. Myneni, "Superconducting DC and RF Properties of Ingot Niobium", in *Proc. SRF'11*, Chicago, IL, USA, Jul. 2011, paper THPO057, pp. 856–861.
- [18] S. Balachandran, P. Xu, P. Dhakal, M. Carl, R. P. Walsh, and P. J. Lee, "Effect of Strain on the Resistivity and Thermal Conductivity of High Purity Niobium", *IEEE Trans. Appl. Supercond.*, vol. 33, pp. 1–5, 2023. doi:10.1109/TASC.2023.3262218
- [19] P. Kneisel *et al.*, "Review of ingot niobium as a material for superconducting radio frequency accelerating cavities", *Nucl. Instrum. Methods Phys. Res., Sect. A*, vol. 774, pp. 133–150, 2015. doi:10.1016/j.nima.2014.11.083
- [20] L. S. Sharath Chandra, M. K. Chattopadhyay, S. B. Roy, V. C. Sahni, and G. R. Myneni, "Magneto thermal conductivity of superconducting Nb with intermediate level of impurity", *Supercond. Sci. Technol.*, vol. 25, p. 035010, 2012. doi:10.1088/0953-2048/25/3/035010
- [21] G Ciovati, P Dhakal, G. R. Myneni, "Superconducting radiofrequency cavities made from medium and low-purity niobium ingots", *Supercond. Sci. Technol.*, vol. 29, p. 064002, 2016. doi:10.1088/0953-2048/29/6/064002
- [22] G. R. Myneni, *et al.*, "Medium grain niobium SRF cavity production technology for science frontiers and accelerator applications", *J. Instrum.*, vol. 18, p. T04005, 2023. doi:10.1088/1748-0221/18/04/T04005
- [23] M. Yoshida, Md. R. Kasem, A. Yamashita, K. Uchida, and Y. Mizuguch, "Magneto-thermal-switching properties of superconducting Nb", *Appl. Phys. Express*, vol. 16, p. 033002, 2023. doi:10.35848/1882-0786/acc3dd
- [24] A. Davies, "Material properties data for heat transfer modeling in Nb3Sn magnets", *Illinois Accelerator Institute*, vol. 17 pp. 1–53, 2011. https://uspas.fnal.gov/materials/ 21onlineSBU/Background/Further\%20reading\ %20-\%20Cryogenic\%20material\%20properties. pdf
- [25] J. E. Jensen, W. A. Tuttle, R. B. Stewart, H. Vrechna, and A. G. Prodell, "Cryogenic data notebook", Brookhaven National Laboratory, NY, USA, 1980, vol. 1, sections I-IX.
- [26] J. Knobloch and H. Padamsee, "Flux Trapping in Niobium Cavities During Breakdown Events", in *Proc. SRF'97*, Padova, Italy, Oct. 1997, paper SRF97B06, pp. 337–344.