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LOW TEMPERATURE THERMAL CONDUCTIVITY OF NIOBIUM AND MATERIALS FOR SRF CAVITIES

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Abstract

A test facility, allowing the test of 4 samples simultaneously during each run, was developed for measuring at low temperature (T= 1.5 K-60 K) the thermal conductivity k(T) of niobium and other materials used for the fabrication of SRF cavities. The measurements are performed using steady-state axial heat flow method with a careful control of heat leaks to the surrounding. Several samples of different materials (industrial Nb sheets, Ti…) were either tested as received or/and subjected to various Heat Treatment (H.T) prior to the experiment then tested. The resulting experimental data are presented and compared to the experimental results previously reported by other groups. As expected, H.T @ 1200°C with Ti gettering improves the Nb RRR by a factor of 3 and consequently k(T). Finally, the correlation between the Niobium RRR and the thermal conductivity at T=4.2 K is confirmed in good agreement with the Wiedemann-Franz law.

INTRODUCTION

SRF cavities accelerating structures are a very promising technology for various large scale applications such as ILC, facilities based on high power proton linac (ESS [1], MYRRHA) dedicated to various applications (spallation neutron sources, nuclear waste transmutation). Most of SRF cavities actually used worldwide since 3 decades or foreseen (e.g ILC) are made of high purity (i.e RRR≥300) bulk niobium (Nb) sheets (thickness : 3 mm- 4mm). Moreover, at large scale the maximum achieved accelerating field E_{acc} in SRF β=1 cavities for electrons are still below the ultimate intrinsic theoretical limit corresponding to the superheating magnetic field B_{sh} (i.e $E_{acc}^{\max} \sim 50$ MV/m corresponding to B_{sh}= 190 mT@ T = 2.0 K) of Nb. More precisely, the two fundamental RF parameters E_{acc} and unloaded quality factor Q_0 of SRF resonators are limited by two dissipative phenomena or anomalous RF losses at high gradient (i.e. $E_{acc} \geq 15$ mV/m for β=1 resonators): 1) Electron Field Emission (EFE) and/or Thermal Breakdown (TB) or quench. Thanks to the world-wide R&D effort two decade ago, the onset threshold of EFE was pushed towards accelerating gradients in the range 20 mV/m $\leq E_{acc} \leq 30$ mV/m. Briefly, the recipe used consists of eliminating and removing the main EFE sources (whiskers, foreign dust particles,…) from the cavity internal surface by careful cleaning of the RF surface, handling and by avoiding any further contamination during each step of cavity preparation. We should then focus on TB. According to experimental observations, quench is triggered by anomalous RF losses due to strong Joule heating of localized defects: these so-called defects (size: ~1µm-10µm) are normal conducting metallic inclusions in the Nb or/and weakly superconducting regions on the Nb RF surface. Hence, in order to achieve, at a large scale, the ultimate superconducting performances of bulk Nb one have to overcome and find remedies to quench in two ways: 1) Reduce defects size and density on the RF surface, 2) Thermal stabilization of the unavoidable remaining defects. Thanks to an important improvement of Nb sheets production and purification, high thermal conductivity $k_{Nb}(T)$ (i.e RRR≥300, thermal conductivity at T=4.2 K $k_{4.2} \geq 60$ W/m.K) Nb for SRF resonators production is nowadays industrially available. Moreover, the accelerating gradient of 33 mV/m needed for ILC is very challenging. As $k_{Nb}(T)$ at low temperature (i.e T ≤10 K) is the main thermos-physical parameter which controls cavity thermal quench limit ($E_{acc}^{\max} \sim (k_{Nb})^{1/n}$ with n≅ 2), it is important to improve and master the process for increasing k_{Nb} and to get accurate experimental data. Furthermore, it was recently demonstrated that nitrogen doping and nitrogen infusion allows reduction of dynamic RF losses in SRF cavities by a factor ~3. In the framework of this international R&D program, it is necessary to control, measure reliably and with a good precision the thermal conductivity of niobium and other materials commonly used in SRF cavities fabrication. The aim of this paper is to present, analyze and discuss the experimental thermal conductivity data obtained with a dedicated apparatus. The paper is organized in four sections. The experimental set-up and procedure are described in the first section. In the second section we will present and discuss thermal conductivity our experimental data which are analyzed and compared to with previously reported data. In the same section, we discuss also RRR data in correlation with thermal conductivity.

TEST-STAND DESCRIPTION

The principle of the method used is the so-called steady state axial heat flow technique operated in the low heat flux q regime (i.e. q~50W/m^2 and ΔT ~10 mK over a length l=10 mm) but with some refinements in order to insure a good reliability of the measurement and to improve the corresponding sensitivity and accuracy. The test-cell developed (Fig. 1) for precise measurement of low temperature (i.e. 1.5 K-60 K) thermal conductivity of different materials commonly used for the fabrication of SRF cavities consists mainly on four major components detailed in the following.

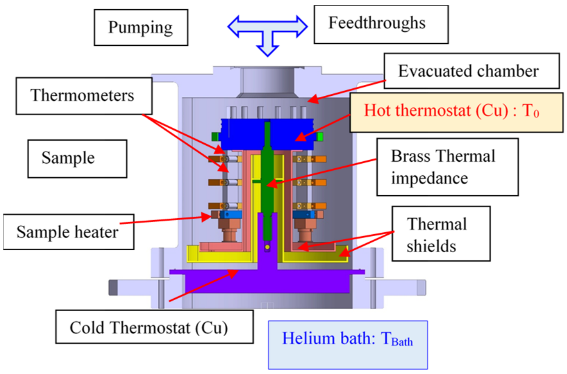

Figure 1: Thermal conductivity test-cell diagram.

A temperature controlled heat sink (T_o), or hot thermostatic O.F.H.C copper block, which is connected to a cold thermostat Cu block directly cooled by the surrounding liquid helium bath (LHe, temperature : T_{Bath}), via a carefully designed brass rod (diameter = 10 mm, length = 40 mm) acting as a thermal impedance between the heat sink and the cold source (Lhe). The heat sink temperature T_o is regulated by means of a resistive heater (manganin wire wrapped around the Cu block, R_{Heat}= 50 Ω) using a calibrated cernox resistor as a temperature sensor while the cold source (Lhe) is maintained at T_{bath}. Precise measurement of T_0 is performed by means of a reference cernox thermometer absolutely calibrated to within ± 10 mK in the temperature range 1.5 K – 20 K. Note that this thermometer is also used for referencing all the thermometers at the thermal equilibrium (q=0) and under isothermal conditions (see next section). Furthermore, T_{Bath} is also regulated via the control of the LHe bath vapour pressure (2 capacitive pressure sensors (0-1000 Torr and 0-100 Torr full scale) and automatic valve with a PID regulator) and monitored with another calibrated cernox thermometer. Four test-samples (length = 55 mm, width = 10 mm, thickness = 0.3 mm to 4 mm depending on the specimen), are measured simultaneously during each experimental run. (Fig. 2)

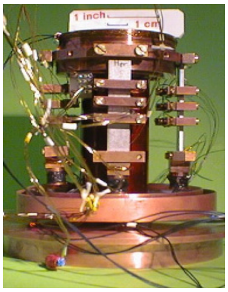

Figure 2: Close view to the samples mounted on the hit thermostatic Cu block.

Each of these test-samples is equipped with a removable heat source attached to its lower extremity and four calibrated (1.5 K–300 K) cernox resistors. Three of these thermometers are used to measure the temperature gradient along the test-sample. Note that as the experiment is performed at low heat flux (see next section), it is more reliable to use three temperature sensors instead of two (linear temperature profile along the sample). Moreover, a special attention was given to thermometers mounting and thermal coupling to the sample: knife edge–like O.F.H.C copper clamps are used and careful thermal anchoring of thermometer leads to the sample is insured using a copper filled grease as thermal bonding agent (CRYCON). It has to be stressed that the thermometers leads were carefully thermal anchored at different locations (thermometer location, heat sink, then LHe bath) so as to improve the accuracy of temperature measurement. The heat source which consists of a Joule heated O.F.H.C Cu block with manganin wire wrapped around is also mounted on the sample using CRYCON grease so as to improve thermal contact.

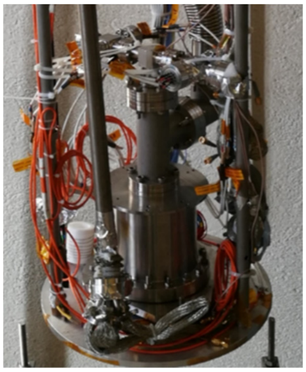

Figure 3: Test-cell on the cryogenic insert ready for testing.

Finally, the test-samples are fixed and thermally clamped to the heat sink at their upper extremity. Two O.F.H.C copper radiation shields coated with one layer of superinsulation surround the heat sink as well as the test-samples. The innermost radiation shield, which is thermally clamped to the heat sink, is directly placed around the test-samples. The second radiation shield is thermally attached to the brass rod at an intermediate temperature between T_{bath} and T_{sink}. The whole assembly is placed inside a stainless-steel vacuum insulating jacket (Fig. 3). Note that as the major parts of this system are removable, a special care was given to insure proper clamping and good thermal contact: all copper pieces were first cleaned and deoxidized then coated CRYCON grease just before each mounting operation.

RESULTS AND DISCUSSION

Several samples of different materials (Nb, Ti,…), supplied by different companies were either tested as received or/and subjected to various Heat Treatment (H.T) prior to the experiment then tested. In particular, we studied the effect of both the material initial purity (i.e. RRR), which was also either measured and/or calculated from the material impurity content (O, N, H, Ta…) analysis, and the subsequent H.T on the sample thermal conductivity.

Heraeus Niobium

The data obtained with niobium sheets supplied by Hereaus are presented in Fig. 4. These data lead to the following observations: 1) Heat treatment with Ti gettering improves the thermal conductivity of Nb in whole temperature range 1.5 K-9 K, 2) This improvement is attributed to recrystallization of niobium, reduction of the density of dislocations in the material (annealing effect), purification (removal of interstitial impurities such as hydrogen, nitrogen and oxygen), 3) The measured initial (i.e as received sample) value of RRR is 137, 4) The experimental RRR value is close (to within 7%) to the calculated value according to Wiedemann-Franz law, namely RRR_{Th}=128, 6)At T=4.2K the thermal conductivities of as received and HT sample are respectively 40 W/m.K and 115 W/m.K, 5) HT with Ti gettering have a strong effect on the phonons peak at T=2K : for as received k_{2K}= 7 W/m.K, for HT k_{2K}= 30 W/m.K, 6) The enhancement of thermal conductivity is much stronger at T=2 K as compared to other temperatures.

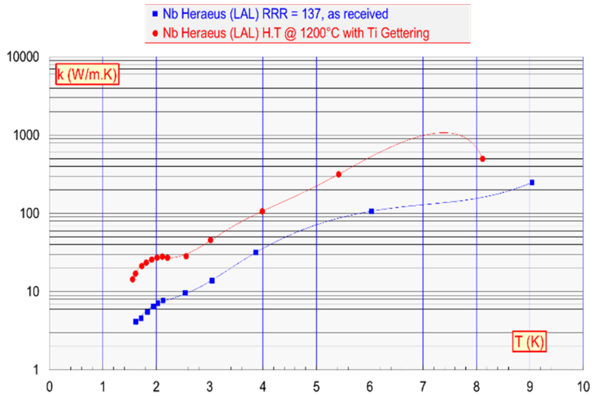

Figure 4: Effect of heat treatment at 1200 °C with Ti gettering on the thermal conductivity of Heraeus Nb.

Tokyo Denkai Niobium

The results obtained with niobium sheets supplied by Tokyo Denkai are presented in Fig. 5. These data show a different behaviour: 1) No phonon peak is observed for both as received and heat treated samples, 2) Heat treatment with Ti gettering improves the thermal conductivity of Nb above T=2.3 K. Below this temperature a reduction of k(T) is clearly observed. 3) The effect of HT seems dependent of Nb supplier and hence the production process, 4) according to the observed behaviour the crystal structure and defect density due to cold work and plastic deformation during production of Tokyo Denkai niobium seems to be of lower quality as compared to Heraeus Nb.

Wah Chang Niobium

Four samples of different initial RRR ranging from 46 (reactor grade Nb) to 200 was supplied by Wah Chang and tested. The data, presented in Fig. 6, show that Wah Chang Niobium has a behaviour similar to Heraeus samples.

Furthermore, as compared to previous studies [1-5], our experimental data on Nb supplied by Heraeus, Tokyo Denkia and Wah Chang show the same behaviour concerning: 1) thermal conductivity versus temperature (shape of the curve and phonon peak at T~2K), 2) the correlation between RRR and $k_{4.2}$ according to Wiedemann-Franz law (i.e. RRR=a. k4.2 (W/m.K) with a =4-5 m.K/W), 3) the heat treatment and impurities have a strong impact on the phonon peak

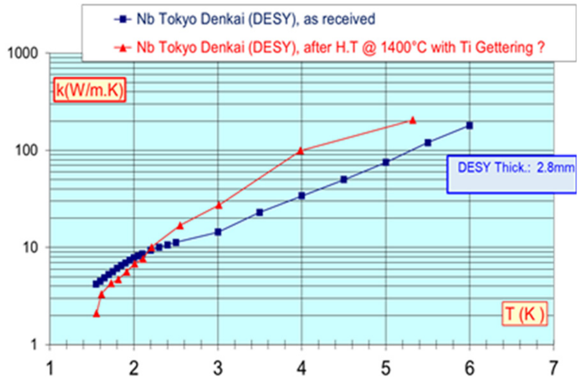

Figure 5: Effect of heat treatment at 1400 °C with Ti gettering on the thermal conductivity of Tokyo Denkai Nb.

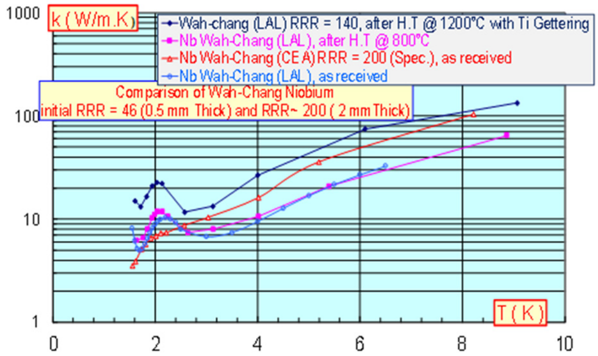

Figure 6: Effect of heat treatment at 1400 °C with Ti gettering on the thermal conductivity of Tokyo Denkai Nb.

CONCLUSION

A test facility dedicated to the measurement at low temperature (1.5 K- 60K) of the thermal conductivity of materials commonly used for the fabrication of SRF cavities. Several samples of niobium sheets supplied by different companies were either tested as received or/and subjected to various Heat Treatment (H.T) prior to the experiment then tested. The experimental data show that the phonons peak around T=2 K depends strongly on the following: 1) niobium sheets production process, 2) impurities, including hydrogen and heat treatment with and without titanium gettering. Heat treatment purification of Niobium with titanium gettering at temperature from 1200 °C to 1400 °C impacts strongly k(T). This effect is due to reduction of dislocations density in Niobium and purification (i.e reduction of oxygen content). Heat treatment at 800 °C (hydrogen outgassing) impacts also k(T). The reduction of quench field subsequent to baking and/or nitrogen doping could be attributed to reduction of k(T) the phonons peak. Finally, as expected according to the well-known Wiedmann-Franz law, the correlation (e.g $k_{4.2} \propto$ RRR) between RRR and $k_{4.2}$ is confirmed.

REFERENCES

[1] K. Mittag, "Kapitza conductance and thermal conductivity of copper, niobium , and aluminium in the range from 1.3 K to 2.1 K", *Cryogenics,* vol. 13, p. 94, 1973.

[2] P. H. Kes, J. G. A. Rolfes, and D. de Klerk, "Thermal Conductivity of Niobium in the Purely Superconducting and Normal States", *Journal of Low Temperature Physics*, vol. 17, nos. 3/4, pp. 341-364, 1974.

[3] M. Fouaidy *et al.*, "Kapitza conductance and thermal conductivity of materials used for SRF cavities fabrication", in *Proc. of the 9th workshop on RF superconductivity*, Santa Fe, Nov. 1999, pp. 263-266.

[4] F. Koechlin and B Bonin, "Parametrization of the niobium thermal conductivity in the superconducting state", *Supercond. Sci. Technol.*, vol. 9, pp. 453-460, 1996.

[5] S. K. Chandrasekaran *et al.*, "Hydrogen saturation and the thermal conductivity of superconducting niobium", in *Proc. SRF'13*, Paris, France, Sep. 2013, paper TUP067, pp. 589-592.