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THERMAL CONDUCTIVITY OF ELECTROPLATED COPPER ONTO BULK NIOBIUM AT CRYOGENIC TEMPERATURES*

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Abstract

Superconducting radio-frequency (SRF) cavities made of high-purity bulk niobium are widely used in modern particle accelerators. The development of metallic outer coatings with high thermal conductivity would have a beneficial impact in terms of improved thermal stability, reduced material cost and for the development of conduction-cooled, cryogen-free SRF cavities. Several high-purity, fine-grain Nb samples have been coated with 2-4 mm thick copper by electroplating. Measurements of the thermal conductivity of the bimetallic Nb/Cu samples in the range 2 – 7 K showed values of the order of 1 kW/(m K) at 4.3 K. Very good adhesion between copper and niobium was achieved by depositing a thin Cu layer by cold spray on the niobium, prior to electroplating the bulk Cu layer.

INTRODUCTION

Modern particle accelerators rely more and more often to superconducting radio-frequency cavities (SRF) to accelerate the beam because of their higher efficiency, compared to normal-conducting ones. Research and development over the past 40 years has made bulk Nb the material of choice for this application and accelerating gradients close to the thermodynamic critical field of Nb have been achieved in state-of-the-art prototypes [1, 2]. One of the limitations towards reliably achieving accelerating gradients close to the theoretical limit is a thermal quench at lower field, initiated typically at defects introduced during the cavity manufacturing process [3].

The cost of high-purity Nb, having a residual resistivity ratio (RRR) greater than 300, is of the order of $1,000/kg, significantly higher than that of other metals used to produce accelerator cavities, and the requirement for the high RRR-value is dictated by the need for high thermal conductivity to push the quench field to higher values [3]. The availability of a method to provide a metallic coating with high thermal conductivity, κ, at liquid helium temperatures onto the outer surface of a Nb cavity would address both the thermal stability and the cost issues, as thinner Nb with lower RRR could be used to fabricate the cavities. The alternative approach of coating a high-κ substrate with a Nb thin film has yet to produce cavities which can achieve as high accelerating gradients with a high quality factor, as what can be routinely obtained with bulk Nb [4].

Alternative superconductors, other than bulk Nb, are being actively explored and significant progress has been made in recent years for Nb_3Sn films onto Nb substrate, with cavities achieving up to 17 MV/m with a quality factor of $\sim 1\times10^{10}$ at 4.3 K, much higher than what can be achieved in the same cavity with bulk Nb at the same temperature [5]. The capability of operating SRF cavities efficiently at 4.3 K, instead of 2 K, is a key to the development of SRF accelerators for industrial applications. In such applications, the cavities could be conductively cooled using cryocoolers if a high-κ coating is applied to the outer cavity surface [6-8].

In the past there were two most notable R&D efforts to produce bimetallic bulk Nb/Cu cavities: in one case, hydroforming was used to form the cavity from a Nb/Cu tube made by explosion bonding or hot-isostatic pressing [9-11], in the other plasma-spray techniques were used to add a copper coating on the outer surface of a standard bulk Nb cavity [12, 13]. It was shown that Nb/Cu cavities made by hydroforming achieve a performance similar to that of bulk Nb cavities [14], however this method is not applicable to the current method of Nb_3Sn thin film formation, which relies on thermal diffusion of tin into bulk Nb at temperatures above ~1000 °C. The plasma-spray methods investigated at IPN-Orsay were high-velocity oxy-fuel and inert plasma spraying, however the thermal conductivity at 4.3 K was not as high as that of high-RRR Nb [12, 13].

In this contribution we report on the deposition of 3 - 5 mm thick copper coatings by electroplating on high-purity bulk Nb samples and their thermal conductivity at cryogenic temperatures. Electroplating is a relatively inexpensive method which could be applied to fully finished cavities, whether bulk Nb ones or bulk Nb with a Nb_3Sn film on the inner surface.

SAMPLE PREPARATION

Samples Plated at AJ Tuck, Co.

For the plating done at A. J. Tuck Co., Brookfield, CT, USA, seven Nb samples, 3.175 × 3 mm² cross-section, 75 mm long and three Nb samples, 25 mm wide, 1 mm thick, 140 mm long were cut by wire electro-discharge machining (EDM) from a high-purity, fine-grain (ASTM > 5) Nb sheet used for srf cavity fabrication. The samples were chemically etched to remove ~100 μm from the surface by buffered chemical polishing (BCP), annealed in a vacuum furnace at 600 °C/10 h, followed by BCP removing

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~25 µm at Jefferson Lab. Six of the 3 mm wide samples and two of the 25 mm wide samples were shipped to A. J. Tuck Co., where they were processed as follows:

- The side to be plated was sand-blasted with coarse grit on three of the samples (No. 1, 2 and 3)
- All samples were plated with ~5 µm thick Cu
- Mask 3 sides of the samples (the ones which were not grit-blasted in samples No. 1, 2, 3)
- Activate the substrate and plate up over 3 mm copper on the exposed side
- Machine the copper to 3 mm thickness on one side. Leave a very thin layer of copper on the other sides which is then removed by lap-sanding.

The average thickness of the Cu-plating was (3.0 ± 0.1) mm. It was noticed that even though molecular bonding of Cu onto Nb was not achieved, the adhesion was fairly good in the grit-blasted samples, whereas the others looked "flakey" under a microscope, sample No. 4 being the worst.

The thin, wide Nb strips were used to try developing a process with improved, reliable adhesion of the Cu on the Nb. The following methods were tried:

- Brush plating with a stainless steel wire brush, a ScotchBrite brush and a brush made with cotton balls fastened to a platinum plated titanium expanded mesh strip.
- Nickel-striking the Nb surface, following the method described in [15].
- The method of [16] which involves nickel striking, copper plating and vacuum annealing, done at 600 °C/3 h, final copper plating to full thickness
- BCP or electropolishing of the Nb test strip immediately prior to copper plating
- Depositing a "seed layer" of Cu, ~65 µm thick, by magnetron sputtering onto Nb, prior to copper plating.

Unfortunately, none of the above mentioned methods resulted in reliable, molecular bonding of Cu onto Nb. In most cases, the plated layer peeled-off when sanding the edges of the test strip, in a few cases a bend test was done according to ASTM B571-97 to judge the quality of the adhesion.

Finally, the cold-spray method [17] was used to produce a "seed" layer with good bonding to the Nb substrate prior to electroplating. The coating, ~76 µm thick, was applied to one of the 3 mm wide sample (CS 1) and one of the 25 mm wide sample at Concurrent Technologies, Corporation (CTC), Johnstown, PA, USA. Helium gas is heated to 300 °C and at a pressure of 2.76 MPa before injecting the Cu powder in the mixing chamber and spraying through a nozzle at ~1 cm distance from the samples. The thick Cu layer was then deposited by electroplating at A. J. Tuck Co. as described above. The bend test on the thin sample showed very good adhesion and a pull test done at CTC on a similarly coated cold-sprayed sample showed an adhesion of ~30 MPa [18].

Samples Plated at Nomura Plating

For the plating done at Nomura Plating, Osaka, Japan, a high-purity, fine-grain Nb sample was prepared at Jefferson Lab with the same procedure mentioned above and another Nb sample of the same quality was already on-hand at Nomura Plating. Prior to shipment, the Nb sample prepared at Jefferson Lab was annealed at 600 °C/3 h. About 3 mm thick Cu was plated on the "in house" sample at Nomura Plating, whereas ~300 µm Cu plating was done on the sample provided by Jefferson Lab. Nickel strike was used prior to Cu plating. Both samples were sent back to Jefferson Lab for additional vacuum annealing at 600 °C/3 h prior to additional plating at Nomura, up to ~4 mm total thickness for the "in-house" sample and up to ~6 mm for the JLab sample. After plating, the Cu-plated side "in-house" sample was machined to 6.2 mm overall thickness (3 mm thick Cu layer), then cut by wire-EDM into four 75 mm long strips, 2.5 mm wide, labeled N1-N4. The Cu-plated side of the JLab sample was machined to ~5.5 mm thickness (8.9 mm overall thickness) and cut by wire-EDM into four 75 mm long strips 2.5 mm wide, labeled NJ1-NJ4. The Cu separated from the Nb on sample NJ2 while engraving its label. A picture of samples N1-N4 is shown in Fig. 1. One of the samples was placed within 10 mm of an ion pump magnet (~120 µT). Remanent magnetization due to the nickel-strike layer was checked with a Hall probe in a shielding chamber and it was below the background level of ~2 µT.

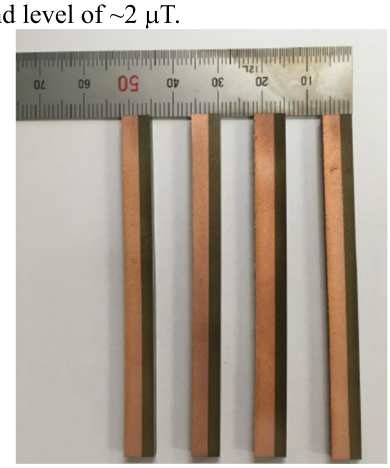

Figure 1: Nb/Cu samples N1-N4 produced at Nomura Plating.

Additional details of the plating process are proprietary to AJ Tuck Co. and Nomura Plating, however, a copper sulfate bath and a DC power supply were used for the plating, with no use of brighteners or additives.

TEST RESULTS

Each sample is mounted on a setup which has been used to measure the thermal conductivity below 10 K at JLab for many years [19]. One end of the sample is clamped to a copper "cold finger" on a 152 mm diameter Conflat flange, which is in contact with the He bath. A constantan wire, 0.08 mm in diameter and to be used as a heater, is wrapped around the opposite end of the sample and glued

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to it using Stycast 2850FT epoxy. Two calibrated Cernox resistance-temperature devices (RTDs) are attached to the Nb side of the sample with GE Varnish, at a center-to-center distance, *d*, from each other. A picture of a sample mounted on the setup is shown in Fig. 2. The Conflat flange with the sample is sealed to a cylindrical can which is mounted on a vertical test stand and evacuated to a pressure lower than 10^{-5} mbar. The test stand is inserted in a vertical cryostat which is filled with liquid He at 4.3 K and then pumped to 1.5 - 2.0 K, depending on the available pumping capacity.

The thermal conductivity, κ, as a function of temperature is calculated using one-dimensional Fourier's law:

$$\kappa(T) = \frac{P\,d}{S\,(T_1 - T_2)}, \tag{1}$$

where P is the heater power, calculated as the product of the voltage across the heater and the current through the heater, S is the cross-sectional area of the sample, T_1 and T_2 are the steady-state temperatures of the RTD closer to the heater and to the cold finger, respectively, and T is the average of T_1 and T_2.

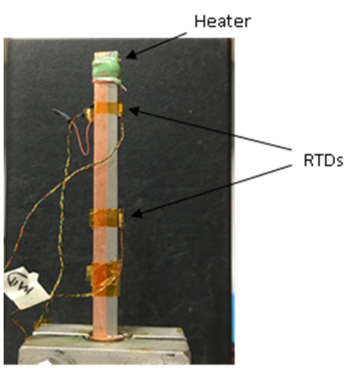

Figure 2: Sample clamped to the cold-finger as part of the thermal conductivity test setup.

A plot of κ(T) measured on the bare Nb sample and on Nb/Cu samples plated at AJ Tuck Co. is shown in Fig. 3. After testing sample CS 1, the Cu bar was cut from the Nb bar by wire-EDM and the thermal conductivity of the electroplated Cu bar only was measured and it is also shown in Fig. 3. A plot of κ(T) measured on the Nb/Cu samples plated at Nomura Plating is shown in Fig. 4. The thermal conductivity at 4.3 K ranged between 600 – 1400 W/(m K) which is about one order of magnitude higher than that of high-purity Nb. A summary of the samples preparation and thermal conductivity at 4.3 K is given in Table 1.

DISCUSSION

The factor of ~2 variation in thermal conductivity data of samples plated at AJ Tuck Co. seems to be correlated to the adhesion at the Nb/Cu interface: for example, the thermal conductivity of grit-blasted samples No. 1 and 2 is higher than that of samples No. 4 and 5, on which the Cu had poorer adhesion to the Nb. The average thermal conductivity of the samples plated at Nomura Plating is similar to that of samples plated at AJ Tuck Co, although with a larger spread, possibly indicating larger process variability. The thermal conductivity of the electroplated copper at 4.3 K, κ_{Cu}, can be obtained from the value measured on the Nb/Cu sample as:

$$\kappa_{Cu} = \frac{\kappa\,S - \kappa_{Nb} S_{Nb}}{S_{Cu}}, \tag{2}$$

where S_{Cu} and S_{Nb} are the cross-sectional areas of the Cu and Nb bars, respectively, and κ_{Nb} is the thermal conductivity of the Nb bar at 4.3 K. The data shown in Figs. 3 and 4 indicate that κ_{Cu} was as high as ~2 kW/(m K), consistent with the value measured on the Cu bar, which corresponds to a RRR of the order of 300 [20], similar to that of oxygen-free high conductivity copper.

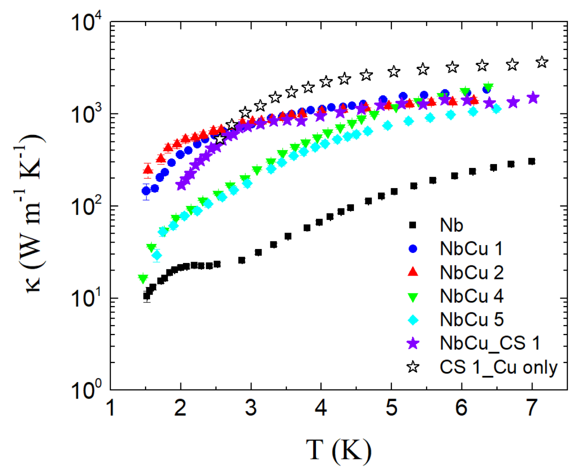

Figure 3: Thermal conductivity as a function of temperature measured on a high-purity Nb sample used for sample No. 1 and on electroplated Cu on Nb samples prepared at AJ Tuck Co. Also shown are the data measured on the electroplated Cu bar after separation from the Nb bar in sample CS 1.

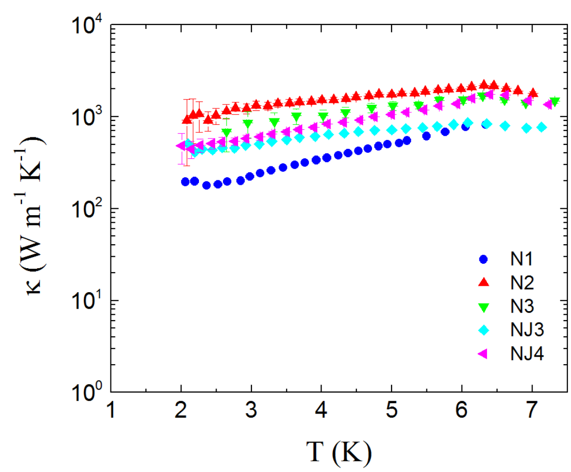

Figure 4: Thermal conductivity as a function of temperature on electroplated Cu on Nb samples prepared at Nomura Plating.

Recent developments at CERN resulted in electroplated Cu samples, prepared with a pulsed-plating technique, with exceptionally high κ(6 K), of the order of 6 kW/(m K) [21].

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Table 1: Summary of Processing Prior to Cu Plating and Values of the Thermal Conductivity At 4.3 K. The Average Value of κ is Given for Samples Processed in the Same Way. Samples N1, N2, N3, NJ3 and NJ4 were Plated at Nomura Plating, All the Others were Plated At AJ Tuck Co.

Sample Label	Processing Steps	κ(4.3 K) [W/(m K)]
Nb	~100 µm BCP, 600 °C/10 h, ~25 µm BCP	86 ± 4
NbCu 1, 2	Same as "Nb", sand-blasting, 3 mm Cu plating	1159 ± 30
NbCu 4, 5	Same as "Nb", 3 mm Cu plating	640 ± 72
NbCu CS 1	Same as "Nb", ~76 µm Cu cold-spray, 3 mm Cu plating	1040 ± 50
Cu CS 1	Cut from "NbCu CS 1"	2400 ± 100
N1, N2, N3	Same as "Nb", 600 °C/3 h, nickel strike, 3 mm Cu plating, 600 °C/3 h, 1 mm Cu plating	1024 ± 590
NJ3, NJ4	Same as "Nb", 600 °C/3 h, nickel strike, ~300 µm Cu plating, 600 °C/3 h, 6 mm Cu plating	765 ± 106

CONCLUSION

The availability of a technique to coat the exterior surface of Nb SRF cavities with a high-thermal conductivity metal would be very beneficial to improve the cavity thermal stability and to expand the application of the SRF cavity technology by using cryocoolers for industrial accelerators.

Different methods of electroplating a few millimeters-thick Cu layer onto 3 mm thick high-purity Nb samples were explored. The thermal conductivity of the coated samples at 4.3 K was ~1 kW/(m K), about a factor of ten higher than that of high-purity Nb at the same temperature. Good adhesion between the Cu and the Nb was achieved by depositing a thin interface Cu layer by cold-spray.

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