Low-temperature thermal conductivity of highly porous copper

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Abstract. The development and characterization of new materials is of extreme importance in the design of cryogenic apparatus. Recently Versarien® PLC developed a technique capable of producing copper foam with controlled porosity and pore size. Such porous materials could be interesting for cryogenic heat exchangers as well as of special interest in some devices used in microgravity environments where a cryogenic liquid is confined by capillarity.

In the present work, a system was developed to measure the thermal conductivity by the differential steady-state mode of four copper foam samples with porosity between 58% and 73%, within the temperatures range 20 - 260 K, using a 2 W @ 20 K cryocooler. Our measurements were validated using a copper control sample and by the estimation of the Lorenz number obtained from electrical resistivity measurements at room temperature. With these measurements, the Resistivity Residual Ratio and the tortuosity were obtained.

1. Introduction

In cryogenics, porous materials are of extreme importance, particularly those with high porosity and reduced pore size. Since this kind of materials have a high surface area and low mass, they can be used in many applications such as heat exchangers and for liquid retention by capillarity for devices used in microgravity like energy storage units [1-3].

A great variety of such materials are commercially available, each one with different characteristics that can be suitable for different applications. They can be poor conductors like ceramics or good conductors if made out of metals. Among these, highly porous copper (porosity in the range of 50 - 85 %) would be an especially interesting material since it can be easily soldered, as opposed to aluminum, and can have a high thermal conductivity. However, as far as we know, it is difficult to find highly porous copper with a pore size lower than 0.5 mm [4].

Recently, Versarien® PLC developed a new technique enabling the production of porous copper with pore size between 20 µm and 1 mm, keeping the porosity high (50 to 85%) [5]. In this paper, a system able to measure the thermal conductivity by the differential steady-state mode as a function of temperature in the range from 20 to 260 K of such a material is presented and the results are discussed against the bulk copper properties.
2. Sample preparation and measurement technique

Four samples with the same pore size (300 - 400 µm) and with different porosity (58, 66, 71 and 73%) were provided by Versarien for thermal conductivity measurements.

Using electrical discharge machining, bar shaped samples of 50 × 6 × 6 mm were prepared. This technique was used because it cuts the sample without mechanical contact, avoiding any property change due to more aggressive cutting techniques. The two extremities of the bar shaped copper foam were soft soldered along approximately 5 mm to two copper blocks (bases). Two thermometers (silicon diodes) are thermalized to the bases and, on the upper base, a heater is also connected in order to establish a heat flux. The lower base is attached to the cold finger of a 2 W @ 20 K Gifford-McMahon cryocooler that was used to cool down the sample and to control and sweep the temperature. This set makes up the measurement cell shown in Figure 1.

![Figure 1. Measurement cell mounted on the cryocooler’s cold finger. The distance between the platinum resistors is around 30 mm. See description in the text.](image)

Since porous copper has a low thermal resistance, the thermal contact resistance between the sample and the copper bases might not be negligible. So, in order to measure only the thermal resistance of the sample, two holes of 1.6 mm were drilled in the sample (distance between holes: 30 to 40 mm) to insert two 100 Ω platinum resistors (Figure 1), calibrated in our laboratory against diode sensors. A layer of high thermal conductivity grease was used to assure a good thermal contact between the platinum resistors and the samples. The grease should not migrate through the porous media since it has a high viscosity. Even if it does, its low thermal conductivity compared to that of copper one will not allow the copper to be short-circuited. Due to the poor sensitivity of the platinum sensors at low temperature, this experimental set up was limited to measurements down to 20 K.

To test the experimental measurement set-up, a control sample made out of bulk ETP copper was sized and machined to have approximately the same length and thermal conductance of the samples.

In order to minimize the uncertainty associated to the measurements, for each lower base’s temperature, five different powers \( \dot{Q} \) were applied in order to obtain a temperature difference between the bases of 2, 4, 6, 8 and 10%. Using these five points and the point obtained at null power, the slope \( d\dot{Q}/dT \), i.e. the thermal conductance between the platinum resistors was obtained. With this methodology (differential steady-state mode), the thermal conductance of the four samples and the control sample was measured (Figure 2).

In Figure 2, the global behavior of the thermal conductance of our porous copper samples and copper bulk control sample can be observed: a well defined maximum around 30 K and a thermal conductance almost constant at high temperature appears, a characteristic of a highly conductive metal. For the bulk control sample, in this figure, the peak appears at a lower temperature and is not clearly visible in our measurements.
The results of the control sample’s thermal conductivity were compared to those referenced for copper with a RRR 50 [6]: On Figure 3 our raw data are represented by closed symbols, whereas the open symbols display the same data with 8% correction. These corrections lead to a quite good matching on the whole temperature range between the nominal and the corrected curve. Three more runs were done as can be seen in the Figure 3 inset (the 8% correction was also applied on these results), showing the reproducibility of our measurements and allowing a more precise determination of the peak temperature. This last determination, despite not quite sensitive, is also consistent with a RRR 50 copper. This RRR 50 value is typical value for ETP copper used as provided without any further treatment. A constant 8% relative error is also quite compatible with uncertainties on geometric dimensions mainly due to the error on length resulting of the holes drilled and on section determination resulting from irregular thickness. The 8% correction factor, which is constant for all temperature range, gives an idea of the absolute error on our thermal conductivity measurements, and, since the data obtained for the control sample cover the same thermal conductance range than that of the copper foam samples (Figure 2), the same error is expected for the porous copper samples.

As can be seen in the Figure 2, there is a slight thermal conductivity increase at high temperatures, so experiments regarding the radiation losses were made. In a new run a shield of aluminum foil thermalized to the cold finger was used to cover the measurement cell, this run did not present significant discrepancies (less than 1%), meaning that the radiation losses are negligible.
3. Analysis

Figure 4 shows the thermal conductivity of the different different samples, $K_{\text{Foam}}$, as a function of temperature.

As can be seen in Figure 4, the samples with higher porosity present lower thermal conductivity. This result is expected since the porosity reduces the area through which the heat flux is transported.
Porosity alone is not enough to describe the difference between the thermal conductivity of a porous material and a bulk one, \( K_{\text{Bulk}} \). The modification of the thermal path due to the porous microstructure also needs to be taken into account. Hence, a topology factor referred frequently as the tortuosity \( t \) has to be used and can be defined as:

\[
K_{\text{Bulk}} = K_{\text{Foam}} (1 - P) t
\]  

Like porosity, this tortuosity factor is purely geometric so it does not depend on the temperature and its value ranges from 0 to 1, where 1 is obtained, for instance, for bulk materials. By normalizing each curve of Figure 4 to the control sample thermal conductivity between 200 K and 260 K where the thermal conductivity is constant and does not depend on the RRR, the \((1-P)t\) and \(t\) factors can be obtained (last columns of Table 1).

Applying this normalization factor for the whole temperature range (the factor is temperature independent), the bulk thermal conductivity of the copper within the sample is calculated (Figure 5).

As can be seen in Figure 5, the peak of thermal conductivity of the samples reaches a lower value and is situated at a slightly higher temperature when compared to the control sample.

This indicates that the copper within the samples has a RRR between RRR 10 and 20, smaller than the usual ETP copper. These results also seem to indicate that this RRR slightly decreases with porosity: this result could be explained by an increase of impurities or crystalline defects during the fabrication. Further analyses are under progress.
By measuring the electrical resistivity of the samples at 300 K, and assuming that the thermal conductivity is constant between 200 K and 300 K, the estimation of the Lorenz number for each sample at 300 K was calculated and compared to the reference value for copper at room temperature \( L_0 = 2.23 \times 10^{-8} \text{W.}\Omega.\text{K}^{-2} \) [8]. These values are presented in Table 1.

| Porosity | k (200 K - 260 K) W.m\(^{-1}.\text{K}^{-1}\) | \( \rho (300 K) \times 10^{-7} \Omega.\text{m} \) | \( L_0 \times 10^{-8} \text{W.}\Omega.\text{K}^{-2} \) | \((1 - P)t\) t | t % |
|----------|---------------------------------|-----------------|-----------------|---------------|------|
| 73       | 34.0                            | 1.97            | 2.23            | 0.0897        | 33   |
| 71       | 45.0                            | 1.61            | 2.41            | 0.1189        | 41   |
| 66       | 66.0                            | 1.11            | 2.45            | 0.1802        | 53   |
| 58       | 83.5                            | 0.80            | 2.22            | 0.2310        | 55   |

As can be seen in Table 1, the different Lorenz numbers obtained for each sample are very similar to each other and to the reference value for copper at room temperature (less than 10 %). Then, since the Lorenz number does not depend on the RRR, tortuosity or porosity, in this temperature range, this agreement between the Lorenz number is a good indicator of the quality of the thermal conductivity measurements at least in the 200-260 K range and confirm that the absolute errors are probably due to geometrical errors.

It can also be concluded that the thermal conductivity of porous copper not only depends on the porosity but also on the tortuosity, that decreases with the porosity.

4. Conclusions
A thermal conductivity measurement system working in the 20-260 K temperature range was built and tested by measuring an ETP copper control sample. This validation is also corroborated by obtaining a Lorenz number (at room temperature) very close to that of copper.

This system allowed to measure the thermal conductivity of four copper samples of relatively high porosity and with a pore size around 300-400 \( \mu \text{m} \). The results show a large peak around 30 K and an almost constant thermal conductivity at higher temperatures: despite the fabrication process used to obtain the porous structures, the samples maintained features that are typical of metals with rather high conductivity.

The normalization of the absolute values of the copper foam’s thermal conductivity to the bulk copper can be obtained using a tortuosity between 30-60%, that decreases with the porosity, and a RRR of the copper within the samples between 10 and 20, allowing a complete description of the results.

This system will be upgraded allowing more precise measurements at low temperatures (down to 13 K) and the results will be published later.

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