Cryogenic thermometry: performance evaluation of hand-made prototypes for metal sheath SPRT calibration at the Triple Point of Argon

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Abstract: In the following paper a measurements data comparison between early vs latest version of the Triple Point for Argon (TP-Ar) system is presented. The updates made on the equipment improved the stability of the freezing-melting plateau, extending its duration above 240 minutes. The repeatability and Self-Heating (SH) parameters were maintained, having a difference in the order of 0.1 10^{-3} mΩ and 0.1 mK respectively. The slope parameter showed a difference of 1 mK between both versions and this improvement allow to generate more stables plateaus. Moreover, this article describes the performance evaluation of hand-made aluminum bushings in metal sheath SPRT's calibrations. In general, it has been possible to observe a decrease of a few mΩ in the SH effect when the bushings were used. The comparison between 25.5 Ω and 100 Ω SPRT showed that SH effect is ten times lower in the SPRT with nominal resistance of 25.5 Ω. Finally, heat flow measurements at different immersion depths are discussed.

1. Introduction

In recent years, the importance of having traceability to primary standards in the range of cryogenic temperatures has been demonstrated with the exponential expansion of the biopharmaceutical industry and the global impact caused by the pandemic that began in 2019. Consequently, the demand for low temperature calibration services by accredited laboratories and some industrial sectors is constantly increasing.

About 20 years ago, the thermodynamics department of the National Institute of Industrial Technology (INTI) anticipated the importance of offering traceability to the TP-Ar. The thermodynamics working group began to analyze the possibilities of extending their calibration capacity acquiring a commercial system for the realization of the TP-Ar. Several years later, the system K38 model 2006 version designed by Pond Engineering Laboratories Inc was chosen. Basically, this portable system has the advantage that a few liters of liquid nitrogen are enough to realize and maintain the TP-Ar, reaching the value of -189.34 °C [1]. In 2021, the K38 was restated to the laboratory after its respective repairs and upgrades. So, the first goal of this research work is carried out a measurement data comparison between the early vs latest version of K38 model: Version 2006 vs. Version 2021.

The second part of this research is focus on metal sheath SPRT's. It is known that calibration of this type of thermometers at cryogenic temperatures is remain a challenge because of the heat conduction effects generated by stainless stems. However, our focus is aimed at this kind of SPRT which are the most chosen by Argentine users in different industrial sectors due to their low cost, robustness, and easy handling [2]. The SH effect is one of the most important factors to consider when working at cryogenic thermometry. Papers have been reported indicating that the SH can be minimized when chemically inert close-fitting bushings are used and manufactured with materials of high thermal conductivity. In fact, the use of bushings is a recommendation that is documented in the ITS-90 Supplemental Information [3]. Basically, bushings consist of tubular structures made of a conductive material whose length and diameter depend on the size of the sensor and the diameter of the protective sheath of the SPRT. Bushings improve thermal contact between the cell and the SPRT, mainly when the diameter of the thermowell is larger than that of the SPRT [4]. Evidence of this were reported for the triple point of water [5-7] and Žužek et al. conducted tests at the fixed points from Zn to Hg [8]. At the time of writing this paper, only one work was found for the use of bushings at the TP-Ar [9]. So, another goal for this research work was to investigate the effect of bushings on the SH in metal sheath SPRT's at the TP-Ar.

2. Experimental procedure

2.1. Realization of triple point of argon

The Instituto Nacional de Tecnología Industrial (INTI) is the National Metrology Institute (NMI) of Argentina. For more than 40 years, it has thermometry laboratories where the ITS-90 is realized, maintained, and disseminated. For the realization of the TP-Ar, the K38 argon triple point cell and maintenance system from Pond Engineering Laboratories, Inc [1] was used. The procedure followed was the same as reported in [10].

2.2. Standard Platinum Resistance Thermometers (SPRT)

For this study, SPRTs were used mainly of metallic type and one of quartz sheath with different technical specifications (Table 1). The SPRT Fluke 5681, was designated as the monitoring SPRT at the TP-Ar and selected to verify that the system is in the freezing-melting plateau.

To record the resistance values detected by the SPRTs, a precision thermometry bridge MicroK 70 Isotech and a 25 Ω or 100 Ω standard resistor (immersed in silicone oil thermostatized bath at 23 °C) were utilized. The environmental conditions of the laboratory were maintained at 23 °C and relative humidity<80%.

Table 1

SPRT's specifications

Sensor	Nominal Resistance (Ω)	Sheath material	Sheath diameter (mm)	Sheath length (mm)	Temperature range (°C)
Fluke 5699	25.5	Inconel	5.56	482	-200 to 661
Fluke 5626	100	Inconel	6.35	508	-200 to 661
Fluke 5681	25.5	Quarz	7.00	520	-200 to 661

2.3. Aluminum bushings

The bushings were manufacture in a hand-made way. For this work, bushings made of aluminum (thermal conductivity k≈ 230 W m^{-1} K^{-1} [6]) were chosen. Their sizes were approximately 60.0 mm of length and outer diameter between 8.00 and 8.20 mm.

2.4. Equations and data analysis

The calculations to estimate the SH was made with the two-current method as reported [8]. Resistance values were measured in two current values: R_1 with 1mA and R_2 with $\sqrt{2}$ mA. Then the SH value was calculated as:

$$SH = (R_1 - R_2)\frac{\partial T}{\partial R} \quad (1)$$

where $R_0 = 2(R_1) - R_2$ is the resistance value obtained at 0 mA current.

The measurements data were collected through software that has been designed several years ago for exclusive use in the laboratory, using the Basic programming language. Data analysis and graphs were performed using other software wrote in Python language. The main libraries that have been used are OS, Pandas and Matplotlib. Spyder was the open-source cross-platform integrated development environment employed.

3. Results and discussion

3.1. Data analysis in Python

From files with the .ASC extension, obtained from laboratory measurements, a Python program was designed that allows processing the data more efficiently and quickly. Briefly, the program consists of a main module (Figure 1) that displays a menu with the different active options for data processing. When an option is selected, the program calls the module and executes it. Upon completion, an image and/or data file (as appropriate) is obtained (Figure 2). Currently, the program is still in development.

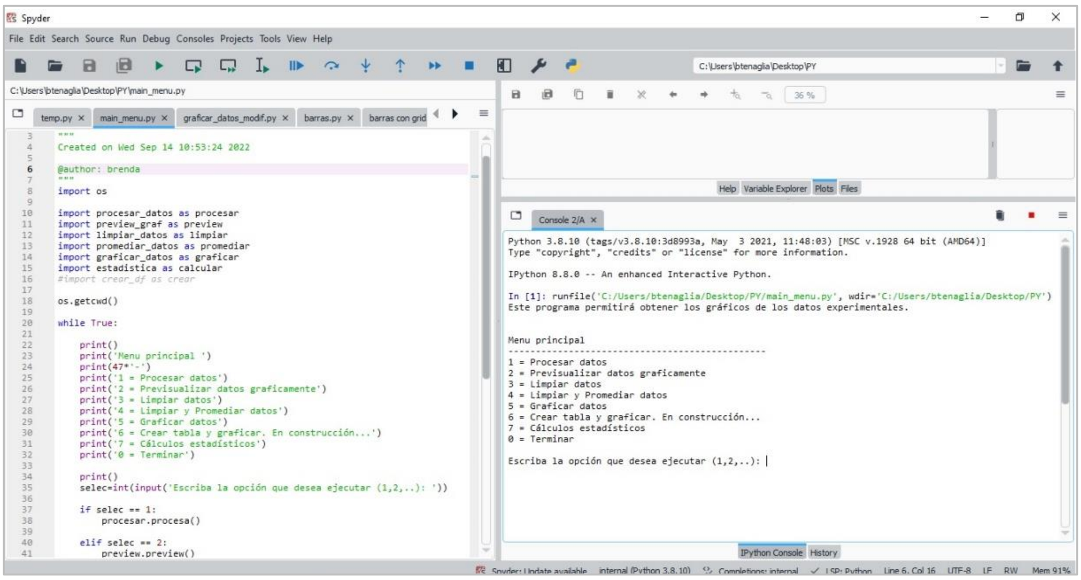

Figure 1 Main menu of the program developed in Python.

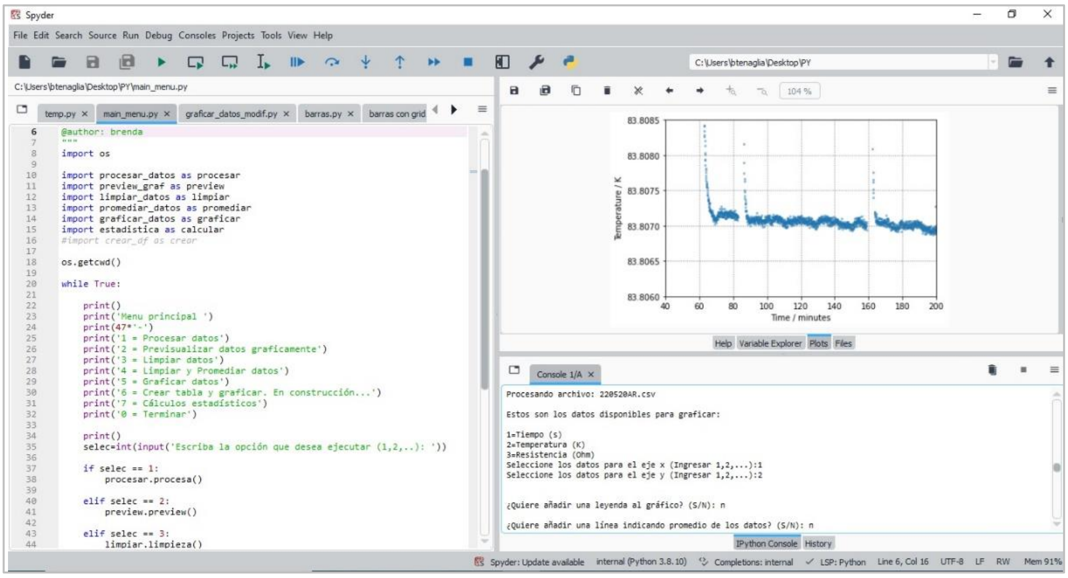

Figure 2 An example of data processing. The output of this module (an image file) is shown.

3.2. Evaluation of TP-Ar system before and after reparation-updated

3.2.1. Historical background

In previous work, B. Tenaglia Giunta et al. reported that INTI has traceability to TP-Ar since 2006 [10]. From its arrival at the laboratory until its irreversible damage (during 2017), there were difficulties for the realization of the TP-Ar. This version had elementary electronics and, consequently, the success of achieving the TP depended entirely on the skill and expertise of the metrologist. Initially, the laboratory did not have a procedure, rather it could only follow some general lines that were in the user manual.

The failure of the freeze-melt experiments of TP-Ar (Figure 3) allowed begin to understand the operation of the K38 model and perform the first draft of the procedure. After several tests, a plateau of approximately 20 minutes was achieved, but not stable enough to be able to calibrate metal sheath SPRT at different immersion depths. When trying to calibrate the latter, the equilibrium of the phases was destabilized and, therefore, the end of the TP-Ar was observed (Figure 4).

During the years 2007-2008 the experiments continued, perfecting the procedure for the realization of this fixed point. However, it was not yet possible to calibrate metal sheath SPRT. The effects produced by the heat fluxes generated due to the high thermal conductivity of Inconel sheath are enough to generate the destabilization of the system. Especially this one that operate at cryogenic temperatures as it had reported in [10].

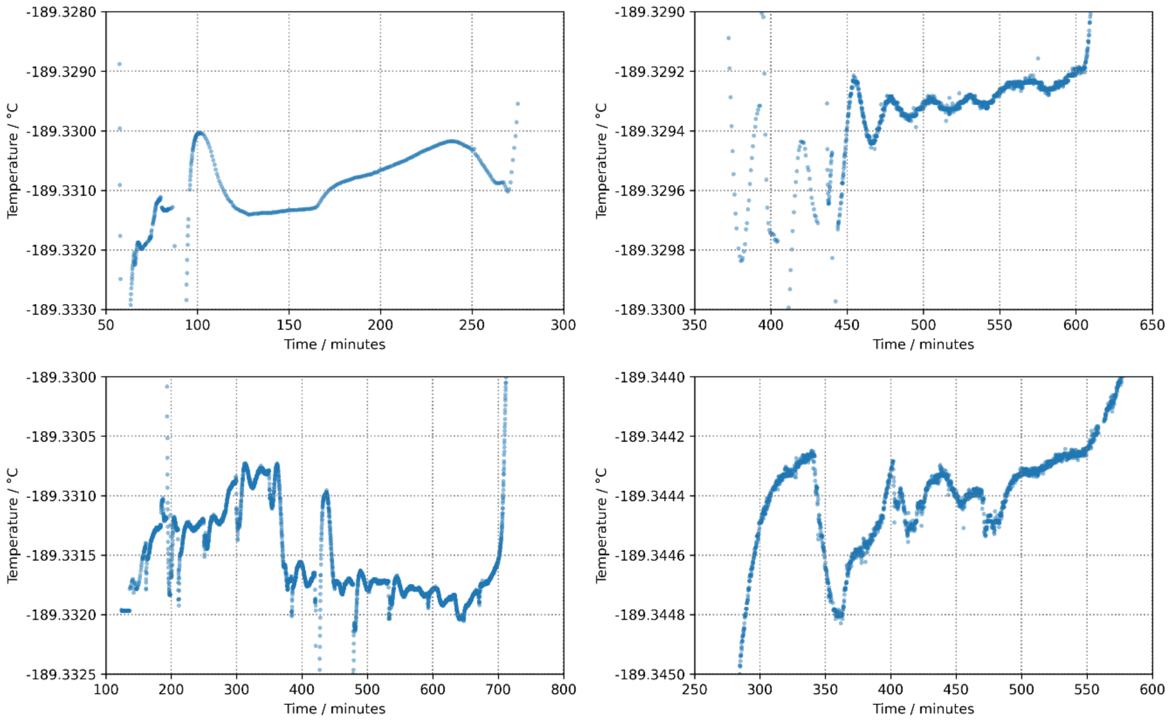

Figure 3 Failed plateaus of the performance of TP-Ar in its beginnings.

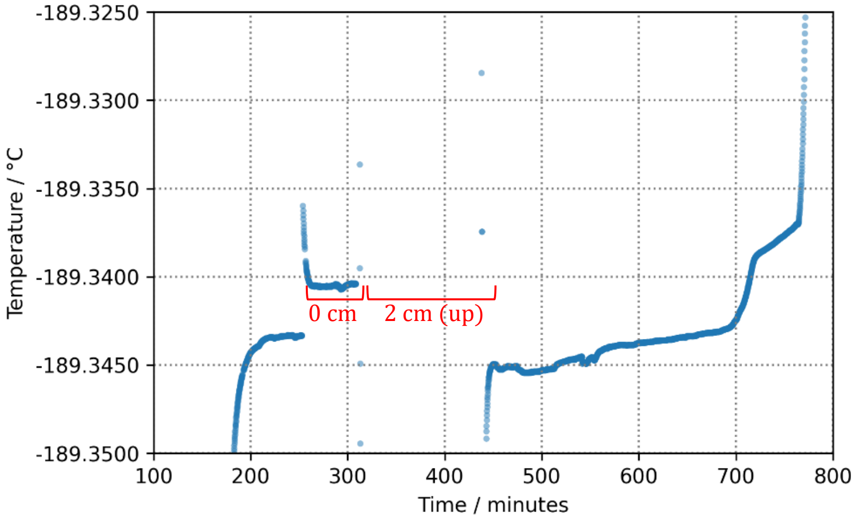

Figure 4 Plateau obtained in 250 to 270 minutes. Attempting to calibrate at immersion depths destabilized the system.

Finally, in 2021 the repaired and updated argon triple point system returned to INTI. A series of experiments were performed to define a new procedure which allowed the realization of stable and reproducible plateaus [10]. It was possible to obtain a freezing-melting plateau with a duration greater than 240 minutes (Figure 5b). The latter is achieved by adding liquid nitrogen to the Dewar when its level drops 75% of the capacity of the container.

It is important to clarify that the resistance measurements from 2021 were made with the MicroK 70 Isotech thermometry bridge while for the previous ones the MI model 6010 automatic resistance bridge was used.

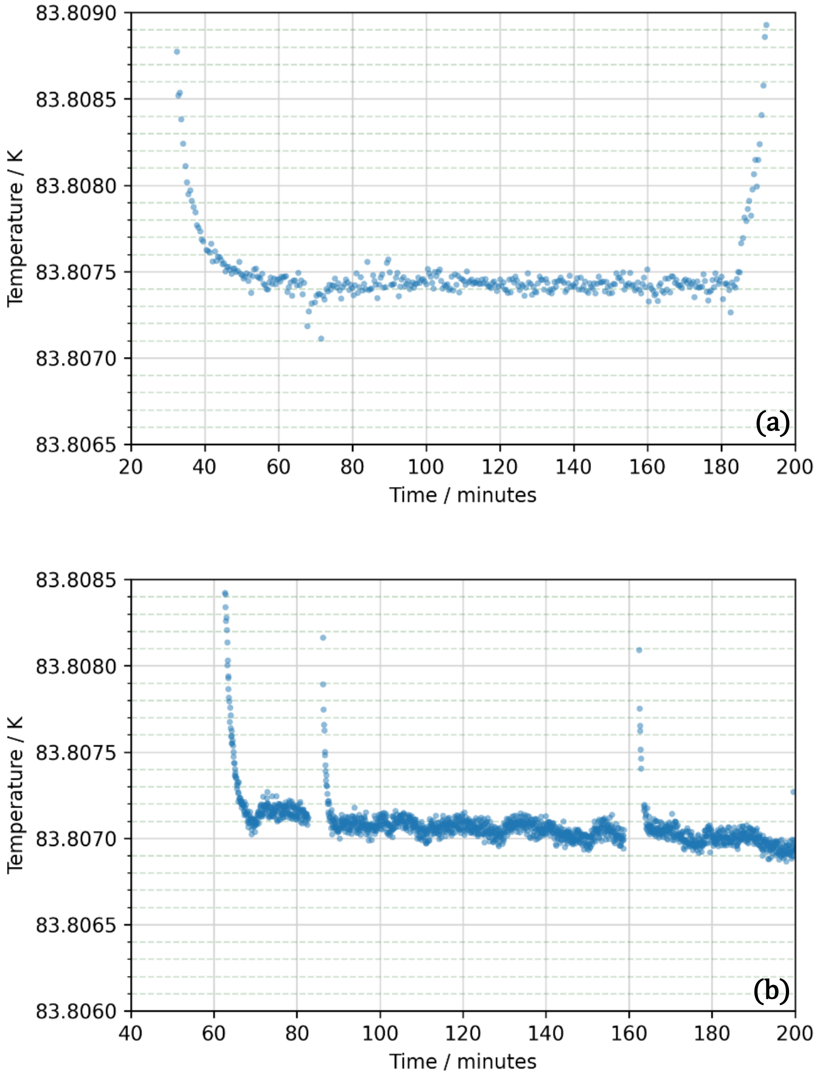

Figure 5 Plateaus realized in : (a) 2017 and, (b) 2022.

3.1.2. Estimated measurement parameter between both versions

A series of parameters were determined to evaluate the performance of early vs. latest versions of the K38 model. Table 2 shows that repeatability and SH did not vary significantly. There is a difference of the order of 0.1 10^{-3} mΩ and 0.1 mK respectively. However, a difference of 1 mK can be seen in the slope parameter, in favor of the 2021 version. This means that the improvements in the equipment and procedure have significantly benefited the stability of the plateau.

A more stable freezing-melting plateau allowed to study metal sheath SPRT's and perform the immersion profiles at two heights. The latter had not been achieved with the old version (Figure 4), meaning a breakthrough in calibration capability in range 4 of the ITS-90 [11] of our laboratory.

Table 2
Estimated measurement parameters

	Version 2006	Version 2021
Repeatability	4.6 10^{-3} mΩ	4.7 10^{-3} mΩ
Self-heating	0.3 mK	0.2 mK
Slope	1.5 mK	0.5 mK

Figure 6 shows the measurement of the Fluke 5699 and Fluke 5626 SPRT's at two immersion depths (0 cm and 2 cm). In both cases it was possible to perform the measurement with 0 cm immersion (green) and 2 cm immersion, that is, 2 cm above the bottom of the thermometric well (red).

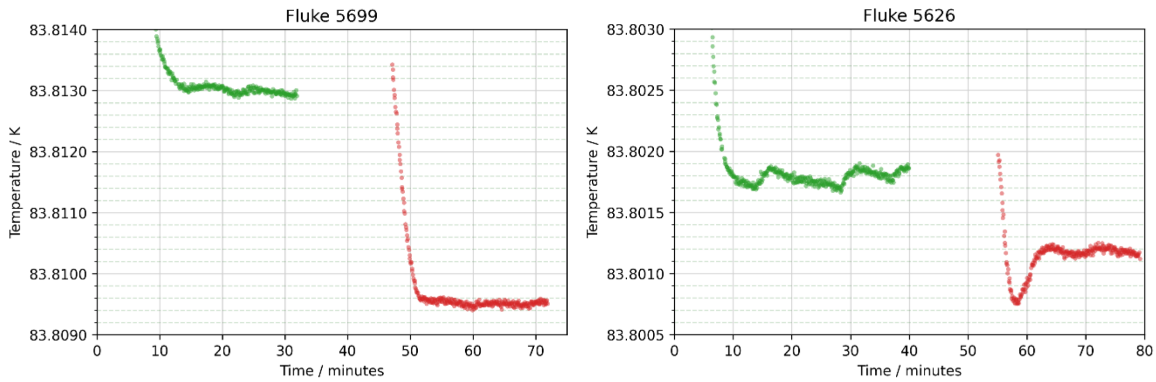

Figure 6 Immersion depth profiles of Fluke 5699 and Fluke 5626.

The difference in mK observed between the two depths can be attributed to poor thermal contact between the cell and the SPRT. It has been observed that by placing the SPRT in another position, the gap decreases. In turn, one would expect that when lifting the SPRT 2 cm, the temperature value increases a few orders of magnitude. However, in this test was observed that the opposite happens. This behavior has not yet investigated, but it can infer that it would be directly related to the design of the K38 system.

3.2. Metal sheath SPRT's: Experimental investigation of thermal contact with aluminum bushings

The placement of SPRTs in the K38 generates air currents and therefore heat flows. Because the diameter of the thermowell (8.2 mm) is greater than the average value of the sheaths (6 -7 mm) [10]. It is known that the SH correction can be minimized filling the air interface with a higher heat transfer capacity [12]. In general, distilled water, alcohol and silicone oil are the most chosen to low SH but sometimes the use of liquid mediums is not a possibility in the range of cryogenic temperature. A possible solution is the placement of heat-conducting bushing. Until now, most of the research work with bushings has been carried out up to the triple point of Hg [4-9]. In this work, the use of bushings in the TP-Ar was proposed.

After analyzing the results presented by Žužek et al. [8] it was decided to begin experiments on the TP-Ar using aluminum bushings. The choice to use this metal was mainly due to its intermediate thermal conductivity between the three metals proposed by them. The main objective is to allow thermal contact between the SPRT and the thermometric well, dissipating the heat produced by the currents, but without these heat flows alter the melting-freezing plateau.

In Figure 7, it was plotted the SH data obtained for the SPRT Fluke 5699 and Fluke 5626. In both instruments it is observed that the aluminum bushing decreases the effects of SH generated by the heat fluxes in the TP-Ar. For Fluke 5699 the difference is 0.019 mΩ and for Fluke 5626 is 0.040 m Ω. In addition, it was possible to verify that the SPRT with nominal resistance of 100 Ω generates an SH ten times greater than that of 25.5 Ω. This is expected due to the high sensitivity of the former [8].

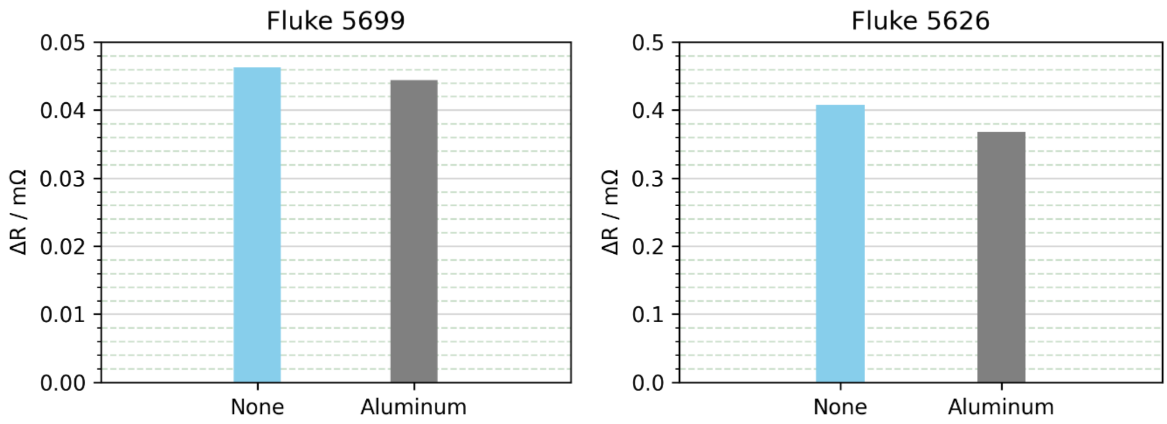

Figure 7 Self-heating in mΩ for SPRT Fluke 5699 and 5626 with aluminum bushing and without bushing.

At the end of the calibration procedure a verification using the SPRT Fluke 5681 is carried out to check if the TP-Ar plateau remains constant. The difference between the initial state and the final state of the plateau, allows us to evaluate if the plateau suffered a variation in resistance after calibrating the metal sheath SPRT's. The graph in Figure 8 compares two situations: the first (light blue) corresponds to SPRT calibrations without bushings and the second (gray) uses aluminum bushings. From the results, the use of bushings protects the duration of the plateau, altering its slope in the order of 0.3 mK. In addition, it is shown that the use of aluminum bushings provides more reproducible plateaus.

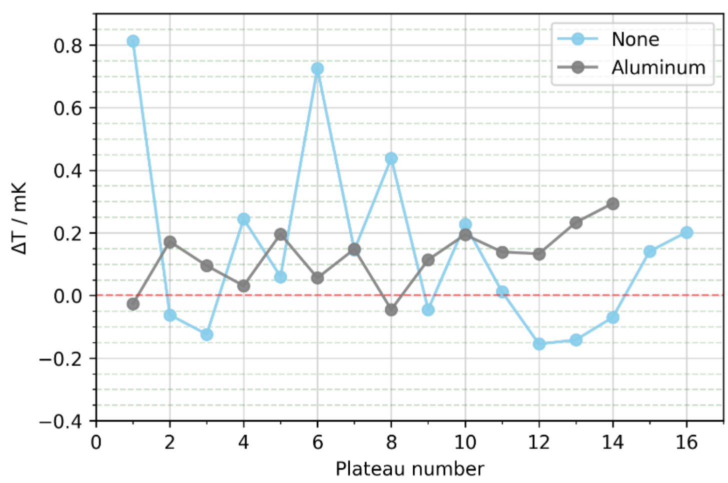

Figure 8 Temperature difference (ΔT) of freezing-melting argon plateau with aluminum bushing and without bushing.

Currently, measurements using different metal bushing are still in progress. Our next goal is test with bushings made of copper and brass (Figure 9) and it expected to find out if a remarkable decrease of SH is possible.

Figure 9 Bushings of copper, aluminum, and brass respectively.

5. Summary

A comparison was made between the early vs. the latest versions of the K38 model in order to evaluate whether the updates improved the realization of the TP-Ar. Notable improvements were found in the stability and duration of the plateau allowing SPRT calibration of metal sheaths at different immersion depth. In addition, tests were carried out using aluminum bushings with the aim of reducing the self-heating effects generated by the Inconel stems. Finally, these preliminary results encourage us to continue measurement testing with other types of metal bushings, focus on improve measurements in TP-Ar.

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