INVESTIGATION OF THE NON-UNIQUENESS OF THE ITS-90 IN THE RANGE 660 °C TO 962 °C

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ABSTRACT

Preliminary results are reported on the non-uniqueness of the ITS-90 using high temperature SPRTs (HTSPRTs) in the range 660 °C to 962 °C. To achieve temperature stability and uniformity of the test HTSPRTs, the method employs the thermal isolation of the Inconel metal comparison block inside a sealed potassium heat pipe controlled to ±0.01 K. Eleven HTSPRTs of three designs from five manufacturers were tested. In addition to measurements near the calibration temperatures 663.3 °C and 966.6 °C to test the temperature uniformity of the comparison block, measurements were made at 703.3 °C, 724.4 °C, 753.2 °C, 801.7 °C, 847.5 °C, and 913.7 °C.

1. INTRODUCTION

This paper is a preliminary report on the investigation of the non-uniqueness, the irreproducibility of temperature values, of the International Temperature Scale of 1990 (ITS-90) between the defining freezing points of Al (660.323 °C) and Ag (961.78 °C) [1]. The high-temperature standard platinum resistance thermometers (HTSPRTs) that are recommended for this range have fused silica (SiO_2) glass supports for the sensor coils and leads, and fused silica glass for the protective sheaths. The electrical resistivity of fused silica glass decreases with increase in temperature. To reduce the electrical leakage, the temperature sensors of HTSPRTs have resistances at 0 °C of about 0.25 Ω to 5 Ω, instead of 25.5 Ω for those used at lower temperatures.

Except for the recent work of Marcarino, *et al*. [2], there is very little systematic information on the non-uniqueness of the ITS-90 using HTSPRTs in the range above 660 °C. The investigation of the non-uniqueness requires the resistance measurement of HTSPRTs to be made at the same temperature and the calibration of the HTSPRTs to remain stable. Because of the long exposures at high temperatures during the test measurements, the calibration of the HTSPRTs is susceptible to change. The calibration stability of the HTSPRT depends on the physical states of the Pt sensor wire, the coil supports and the length of time of exposure to high temperatures. Also, the subsequent re-calibration of the HTSPRTs may not be consistent with the earlier measurements made for non-uniqueness, because of physical changes that occur in the Pt sensor of the HTSPRTs at high temperatures. To obtain repeatable results, HTSPRTs require strict heat treatment as part of the measurement process. To minimize magnetic interferences in the measurements, our furnaces for both the calibration of the HTSPRTs and the non-uniqueness investigation are DC powered. The resistance measurements of the HTSPRTs were made using an AC bridge.

2. HTSPRT COMPARISON APPARATUS

The design of the HTSPRT comparison apparatus is based on the control of a sealed, closed-bottom potassium heat pipe (12.8 cm I.D. x 63.5 cm inside depth) to ± 0.01 K and on the thermal isolation of an Inconel metal comparison block inside the heat pipe to improve the temperature stability and uniformity by an order of magnitude or better. Seven thermometer wells (closed-bottom Inconel tubes, 14.29 mm O.D. x 13.27 mm I.D.) are inserted in holes, in close contact, to the bottom of the cylindrical block. The six wells are evenly spaced in a circle and the seventh well is in the center.

The block is surrounded by a cylindrical Inconel metal shield, with flat ends, and is suspended by the seven thermometer tubes inside the deep heat pipe. The space between the block and the shield and that between the shield and the heat pipe are tightly packed with thermal insulation made from aluminum-silicate fiber. The temperatures along the thermometer tubes are controlled to be at the temperature of the heat pipe to minimize the heat transfer along the tubes. To meet this condition, the seven tubes are in contact with holes in the flat top plate of the cylindrical shield. Above the shield, the tubes pass through nine six-mm thick disks spaced six mm apart to trap the radiation from the heat pipe to heat the tubes. At the mouth of the heat pipe, the tubes pass through holes of an 11 mm thick Inconel “cover” flange controlled with heaters and thermocouples (TCs) at the temperature of the heat pipe. At about the middle, between the upper opening of the thermometer tubes and the mouth of the heat pipe, the tubes pass through holes in a 13 mm thick Inconel “guard” flange controlled with heaters and TCs at the temperature of the heat pipe. The temperatures of the two flanges were controlled automatically or, when the required power was known, the power was set manually. Usually, less than 5 W were required in the cover flange. The space between the opening of the thermometer tubes and the cover flange was packed with thermal insulation.

There are two side heaters and one bottom heater for the heat pipe. One of the side heaters extends over the full length and the other covers the bottom half of the heat pipe. For the bottom, the heater was mounted inside an Inconel metal platform fitted to the curved bottom of the heat pipe. All heaters were non-inductively wound. The heat pipe and the heater system were mounted inside a thermally insulated system.

The temperature of the heat pipe was controlled by means of a Au/Pt TC attached to the inner surface of the heat pipe. The thermocouple was referenced to a Ga melting-point cell. The relative voltage at the desired operating temperature was balanced by a “reference voltage” that was generated using a constant current source and a series of thermostated tapped resistors. Any voltage deviation of the Au/Pt TC from the reference voltage was amplified by means of a microvolt amplifier and directed to a PID controller which adjusted the precision DC power supply to the heater wound over the full length of the heat pipe. The power in the bottom heater and in the lower-half side heater was usually manually set. Differential TCs were used with similar electronic equipment to control the temperature of the two flanges (cover and top guard) attached to the thermometer tubes. Otherwise, the heat pipe was set to control automatically at a given temperature and the comparison block was allowed to come to thermal equilibrium. The heaters are powered by DC power supplies, which in turn are powered by uninterruptible power supplies for a uniform mains voltage.

To minimize the contamination of the HTSPRTs, two nesting closed-end fused silica glass tubes, with a closed end Pt tube in between, extend to the bottom of each of the Inconel thermometer tubes. All three protection tubes fitted closely. Sections of Inconel sleeves were used to fit the outer fused silica glass tubes closely inside the Inconel thermometer tubes. The I.D. of the inner fused silica glass tubes was selected to be (7.80 ± 0.05) mm in order to give a close fit with the HTSPRTs. To reduce the vertical heat conduction of the thermometer protection assembly, the Pt tubes were cut and separated by 6 mm near the top of the block. The use of “precision bore” fused silica glass tubes was avoided because they are usually manufactured using metal inserts that may leave behind metal impurities. Before the three protection tubes were assembled, they were soaked in nitric acid solution (8 mol/L) for several hours, thoroughly rinsed repeatedly with distilled water, and air dried.

3. HTSPRTs AND RESISTANCE MEASUREMENT INSTRUMENTATION

A selection of eleven HTSPRTs from five manufacturing sources and three designs were investigated: Rosemount* birdcage (0.25 Ω), Rosemount notched silica blade coil (2.5 Ω), Hart

Scientific notched silica blade coil (0.25 Ω and 2.5 Ω), Isotech notched silica blade coil (2 x 0.25 Ω), Chino notched silica cross coil (2 x 0.25 Ω and 2.5 Ω), and Yunnan notched silica blade coil (0.25 Ω and 2.5 Ω). These HTSPRTs were calibrated twice at the ITS-90 fixed points from 0 °C to 962 °C. Details of the calibration process and the calibration uncertainties are described in references [3,4].

The resistance measurements were conducted automatically under programmed computer control. An Automatic Systems Laboratories, Inc. (ASL) Model F18 AC bridge, which is capable of 9.5 digit resistance-ratio measurement resolution, was used with thermostated ac/dc Tinsley reference resistors of 10 Ω for 2.5 Ω HTSPRTs and 1 Ω for 0.25 Ω HTSPRTs, giving effectively 0.05 μΩ and 0.005 μΩ resolution, respectively. The bridge was operated at 30 Hz (with detector gain at 5.90×10^4, bandwidth of 0.1 Hz and quadrature detector gain at x10). For the comparison work, a single excitation current of 5 mA for the 2.5 Ω and 14.14 mA for the 0.25 Ω HTSPRTs were used. Sixteen resistance-ratio readings were recorded for the measurement of a HTSPRT at a given current. The bridge was first zero balanced; then, after discarding the first three readings, the next four ratio measurements were recorded. The process was repeated three more times and the sixteen readings were averaged. The standard deviations of such sixteen readings ranged from 4 μK to 11 μK for the 2.5 Ω HTSPRTs and 10 μK to 40 μK for the 0.25 Ω HTSPRTs.

4. METHOD, EXPERIMENTAL PROCEDURE AND ANALYSIS OF MEASUREMENTS

The metal block method requires the interchange of the positions of HTSPRTs to check their temperature uniformity. The interchanging of the positions of HTSPRTs at 660 °C to 962 °C is a very hazardous operation, fraught with many chances of changing the calibration of the HTSPRTs. Because of the hazards, the HTSPRT positions were not regularly interchanged in this preliminary work to check the temperature uniformity of the comparison block. Also, if the block temperatures were uniform at two slightly different temperatures, e.g., 0.02 K to 0.1 K apart, the readings on the HTSPRTs should be different by the same amount. This method was mostly used to test the temperature uniformity of the block.

Another check on the temperature uniformity of the comparison block is to control the block temperature near the freezing temperatures of Al and Ag where the HTSPRTs were calibrated. The readings on the HTSPRTs should reflect the calibration, as long as the temperature non-uniformity of the block and the change in calibration of the HTSPRTs are not balancing out. It is unlikely, however, that the calibrations of all HTSPRTs being tested would change to balance the temperature gradients in the comparison block.

After the comparison block temperature had stabilized at the selected temperature, the HTSPRT resistances were continuously measured sequentially and converted to temperatures. About 35 min were required to make a cycle of readings of seven HTSPRTs. When the temperature was unstable or the range of temperatures of the HTSPRTs was considered too broad, the power in the heaters was adjusted until the temperature variations and the spread in the temperature readings were minimized. The vertical temperature profile of the block was tested by changing the "immersion" of one or two HTSPRTs in the range –10 cm to +4 cm from the normal position and the readings compared. The heater power in the two flanges and in the bottom was adjusted until the temperature variations did not exceed 0.2 mK. Of the many readings, those over a period of three to seven hours or longer during which the readings were nearly stable and their range was relatively narrow, were selected for detailed analysis. On occasions, a new H_2O TP calibration was performed on a HTSPRT if it indicated a large deviation from other HTSPRTs.

Temperature readings of the HTSPRTs at a common time were calculated from the selected data. First, the deviation of the seven HTSPRTs from their average was calculated for every 35-minute cycle of readings over the selected measurement period. The deviations of each thermometer and the average temperatures were fitted to linear equations in time, from which the deviation of each HTSPRT was calculated at a common time and selected average temperature. The range of the experimental deviations represents the combined effect of the non-uniqueness of the ITS-90, calibration errors, changes in calibration, non-uniformity of temperature of the comparison block, stem conduction, self-heating differences, and measurement errors (see Section 5). If the HTSPRTs were at the same temperature, the range of the deviations of HTSPRT readings should indicate the combination of non-uniqueness and HTSPRT calibration uncertainty and the range should be a minimum. On the other hand, in an experimental situation, the range can be smaller or larger, depending upon the manner in which the temperature and calibration of the HTSPRTs are affected. Before the measurements were taken, the furnace temperature was allowed to equilibrate for many days to several weeks. The ranges of plotted data were compared. Of the data sets, represented in each row of Table 1, the narrowest range of observations at a given temperature was taken to be the non-uniqueness, unadjusted for the many sources of uncertainty. Those ranges that were broader were taken to have larger temperature non-uniformity components.

Table 1 is a consolidation of selected data sets that were analyzed in the manner described above. The data are listed in the chronological order of those that were taken during August 1994 to September 2000. During that period, the heaters on the apparatus burned out twice, which required complete re-assembly of the apparatus. Although seven HTSPRTs were always compared, they were not always the same ones because when a HTSPRT began to show large deviation from the others it was replaced by another (see the end and beginning of data on HTSPRTs). The second column lists the average temperature. Columns A to K give the deviation from that average for each of the HTSPRT. The last column lists the range of the deviations of the HTSPRTs that were compared together. Those values of range that are considered to represent the non-uniqueness of the HTSPRTs at the temperature are given in bold type and left justified. Those values of range that are given in italics are considered to have large changes in thermometer calibration.

5. DISCUSSION OF THE RESULTS

Marcarino, et al. [2] investigated the non-uniqueness of fourteen HTSPRTs between the freezing points of Al and Ag employing a pressure controlled sodium heat pipe which directly incorporated six thermometer wells. Tests showed the stability and uniformity of the heat pipe to be ± 0.3 mK to ± 0.05 mK. The authors observed changes in the HTSPRT calibrations during two sets of non-uniqueness measurements. After adjusting for calibration changes in the HTSPRTs, the non-uniqueness in the first set was found to be within ± 27 mK and in the second set to be within ± 35 mK. The authors reported a range of uncertainties (1σ) with the peak at 835 °C of 8.5 mK.

The measurements at NIST, made on commercially available HTSPRTs, show non-uniqueness values (bold figures in the range column of Table 1) considerably smaller than those previously reported. The broader range of temperatures that were observed after the measurements at 966 °C is taken to indicate change in HTSPRT calibration.

Measurements were made at 663.2 °C and 966.6 °C, near the HTSPRT calibration temperatures of 660.323 °C and 961.78 °C, respectively, to check the temperature uniformity of the comparison block and the stability of the calibration. As shown in Table 2, the bold values of **0.3 mK** and **3.3 mK** that were selected at these respective temperatures are within the uncertainty of the measurements. Judging from the data, the calibration of the HTSPRTs was fairly stable until the HTSPRTs were compared at 966 °C. Afterwards we were not able to reduce the range of the readings at lower temperatures to the values that were obtained before the comparison at 966 °C. Experiments were continued and the HTSPRTs were allowed to remain undisturbed until we were certain that their calibrations had changed (see runs 34 on). Near the end of the experiment (runs

Table 1. Summary of HTSPRT non-uniqueness comparison measurements (660 °C to 962 °C). ΔT is the deviation (mK) of the linear fit of the temperature data of the HTSPRT at the average temperature. Italicized data indicates large changes in the HTSPRT calibration. Bolded and left justified numbers in range column are the best estimate of the non-uniqueness. All temperatures were calculated to 0.01 mK; the average temperature is rounded to 0.1 K and the deviations to 0.1 mK. HTSPRT Key for five manufactures and three sensor designs: A: 0.25 Ω; B: 0.25 Ω; C: 0.25 Ω; D: 0.25 Ω; E: 2.5 Ω; F: 2.5 Ω; G: 0.25 Ω; H: 2.5 Ω; I: 2.5 Ω; J: 0.25 Ω; K: 2.5 Ω.

Run	*t* avg.	HTSPRT deviations (ΔT) from the average temperature / mK											range
ID	°C	A	B	C	D	E	F	G	H	I	J	K	mK
1	663.2	-0.1	0.2	-0.4	-0.4			0.2	0.4	0.2			0.8
2	663.2	-0.1	0.4	-0.3	-0.3			0.2	0.1	0.1			0.7
3	663.3	-0.1	0.1	-0.1	0.1			0.0	0.1	-0.1			0.2
4	663.3	-0.1	0.1	-0.1	0.0			0.0	0.1	-0.1			**0.3**
5	663.2	-0.3	0.3	-0.3	0.2			0.0	0.1	0.0			0.6
6	703.4	-0.1	0.4			-0.8	0.1	-0.5	0.3	0.6			**1.4**
7	703.3	0.1	-0.1			-0.8	-0.3	-0.9	0.9	1.0			1.9
8	753.2	-0.7	1.0			0.6	-1.1	0.1	-1.1	1.3			**2.4**
9	753.2	0.0	1.3			-0.4	-1.3	-0.8	-0.4	1.6			2.9
10	724.4	0.3	-0.6			-0.1	0.3	-0.4	0.5	-0.1			1.1
11	724.4	0.4	-0.5			-0.1	0.2	-0.4	0.5	0.0			**1.0**
12	801.8	-0.2				-0.3	-0.5	0.1		-0.1	0.7	0.3	**1.2**
13	801.7	-0.3				-0.3	-0.7	-0.2		0.2	1.0	0.3	1.7
14	801.7	-0.4				-0.8	-1.3	-0.6		-0.1	2.2	1.1	3.4
15	801.7	-0.3					-1.0	-0.5		0.4	1.0	0.5	2.0
16	801.7	-0.3				0.0	-1.0	-0.5		0.5	0.4	0.9	1.9
17	847.5	2.5				-2.2	-3.0	-3.6		0.4	3.5	2.4	7.1
18	847.5	0.4				0.3	-1.3	-0.8		1.0	-0.2	0.6	**2.4**
19	847.5	0.4				0.2	-1.5	-1.0		1.0	0.0	1.0	2.5
20	847.5	-0.2				-1.7	-1.1	-0.1		1.6	0.9	0.6	3.2
21	913.8	1.5				-1.9	-1.4	-0.4		0.8	-0.6	2.1	4.0
22	913.9	0.8				-1.5	-1.5	-0.8		1.3	-0.4	2.1	3.6
23	913.8	-0.5				-0.2	-0.8	0.4		0.3	0.5	0.3	**1.3**
24	913.8	-0.8				0.7	-1.1	0.2		1.0	-0.8	1.2	2.3
25	913.6	-0.8				-0.3	0.1	0.4		0.7	0.2	0.1	1.5
26	966.8	2.5				-2.3	0.4	-1.2		-1.2	1.5	0.4	4.8
27	966.7	3.1				-3.5	-0.1	-0.4		-1.5	5.3	-1.2	8.8
28	966.7	2.4				-0.9	0.4	-0.9		-0.6	1.8	-0.1	**3.3**
29	966.8	3.6				-2.9	-0.9	0.0		-1.4	2.0	0.2	6.5
30	966.8	2.5				-1.6	0.7	-0.7		-0.7	0.3	0.3	4.0
31	966.8	5.5				-0.4	-2.1	-0.2		-1.7	0.7	-1.5	7.6
32	966.8	5.3				-0.9	-1.7	-0.5		-1.2	-0.3	-0.1	7.0
33	966.4	3.6				1.6	-1.1	-1.7		-1.2	-1.0	-0.3	5.3
34	*503.0*	*16.8*				*-9.0*	*-7.0*	*24.7*		*-6.5*	*-10.3*	*-8.8*	*35.0*
35	*502.8*	*7.3*				*-1.2*	*-1.3*	*-1.1*		*-0.1*	*-2.4*	*-1.2*	*9.7*
36	*502.8*	*8.3*				*-1.7*	*-1.8*	*-0.4*		*-0.6*	*-2.2*	*-1.5*	*10.5*
37	*502.8*	*13.0*				*-3.4*	*-3.4*	*3.5*		*-2.4*	*-4.1*	*-3.1*	*17.1*
38	*705.9*	*-73.2*				*12.2*	*14.9*	*6.2*		*11.8*	*15.3*	*12.8*	*88.5*
39	*705.8*	*-73.3*				*12.3*	*14.3*	*5.7*		*12.3*	*15.0*	*13.6*	*88.3*
40	*754.2*	*-80.4*				*13.6*	*12.2*	*6.9*		*17.1*	*13.5*	*17.1*	*97.5*
41	*754.2*	*-80.5*				*12.9*	*12.0*	*7.0*		*16.6*	*13.9*	*18.1*	*98.6*
42	*801.3*	*-84.8*				*15.5*	*11.3*	*7.9*		*18.2*	*12.3*	*19.5*	*104.3*
43	*801.3*	*-83.5*				*16.4*	*8.9*	*7.9*		*20.4*	*11.7*	*18.3*	*103.9*
44	*801.2*	*-83.7*				*14.9*	*10.7*	*11.6*		*16.4*	*14.2*	*16.4*	*100.1*
45	*847.0*	*-71.7*				*-2.6*	*-46.0*	*-12.8*		*59.4*	*0.9*	*72.8*	*144.5*
46	*846.9*	*-72.2*				*-2.9*	*-48.2*	*-13.9*		*59.8*	*0.7*	*76.6*	*148.8*
47	*847.0*	*-71.5*				*-3.0*	*-48.8*	*-13.7*		*59.9*	*0.7*	*76.5*	*148.0*
48	*847.0*	*-71.3*				*-3.1*	*-49.3*	*-14.0*		*60.3*	*0.6*	*76.8*	*148.1*

45 and 46 at 847.0 °C), two HTSPRTs that were located on the opposite sides of the circle were interchanged. The two sets of analysis of the readings, before and after the interchange, showed that the HTSPRTs were at the same temperature within a k=2 uncertainty of 1.4 mK. On the other hand, the readings of runs 47 and 48, in which the HTSPRTs were not interchanged, indicate that the HTSPRTs were at the same temperature within a k=2 uncertainty of 0.24 mK. The results of runs 45 and 46 suggest that the calibration of the two interchanged HTSPRTs had changed.

Table 2 gives the principal uncertainty components of the bold values at the indicated temperatures. As shown in Table 2, the uncertainties of the non-uniqueness measurements are about the same as the non-uniqueness values, which were found to be not larger than 2 mK or 3 mK. In our analysis, we found the instability of the HTSPRT is a large part of the uncertainty, which may be improved by more regular calibration. It is our plan to continue the measurements with new calibrations of the HTSPRTs as well as improvements of the furnace control conditions.

Table 2: Uncertainty estimates of the non-uniqueness measurements. The values of the range and inner-quartile (Q3-Q1) range are derived from Table 1.

t / °C	range mK	Q3-Q1 mK	u (Cal) mK	u (EP) mK	u (SH) mK	u (B) mK	u (SC) mK	U, $k=2$ mK
663.3	0.3	0.2	0.2	0.2	0.3	0.04	0.2	0.9
703.4	1.4	0.6	0.5	0.3	0.1	0.4	0.2	1.5
753.2	2.4	1.7	0.4	0.2	0.3	0.4	0.2	1.4
724.4	1.0	0.6	0.5	0.3	0.4	0.4	0.2	1.7
801.8	1.2	0.4	0.4	0.4	0.9	0.4	0.2	2.3
847.5	2.4	1.1	0.4	0.3	0.7	0.4	0.2	1.9
913.9	1.3	0.7	0.5	0.3	0.9	0.4	0.2	2.3
966.7	3.3	1.9	1.4	0.4	0.4	0.5	0.2	3.2

Uncertainty components are defined as follows: u (cal) = HTSPRT calibration, u (EP) = error propagation, u (SH) = self heating, u (B) = block uniformity, and u (SC) = stem conduction

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* Commercial equipment or materials are identified in this paper to adequately specify the experimental procedure. Such identification does not imply recommendation or endorsement by NIST, nor does it imply that the materials or equipment are necessarily the best available for the purpose.

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