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Preliminary Calibration System Report for High-Temperature Irradiation-Resistant Thermocouples

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SUMMARY

An established calibration procedure is presented for the high-temperature irradiation-resistant thermocouple (HTIR-TC). Standard calibration curves for both individual (i.e., case by case) and common calibrations are shown. The error limits within ±3 σ are ±1% for the individual calibrations and ±2% for the common calibrations.

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ACRONYMS

AGR	Advanced Gas Reactor
EMF	Electromotive Force
F&OR	Functional and Operational Requirements
GUI	Graphical User Interface
HTIR-TC	High-Temperature Irradiation-Resistant Thermocouple
TC	Thermocouple

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1. GENERAL

The high-temperature irradiation-resistant thermocouple (HTIR-TC) is a refractory metal thermocouple (TC) featuring a molybdenum (Mo) wire coupled with a niobium (Nb) wire. When in use, the HTIR-TC can measure up to 1600°C during highly radioactive experiments (e.g., in a thermal neutron flux field). This report documents the process and results of calibrating the HTIR-TC. The HTIR-TC is assumed to have been manufactured correctly and heat treated to the appropriate temperatures, for the appropriate length of time [1].

2. SETUP

After constructing the HTIR-TC within mineral-insulated, metal-sheathed cabling, the assembly is leak checked, the extension wiring (e.g., soft jacketed Mo/Nb wire duplex) welded in place, and the TC heat treated. Then the HTIR-TC can be calibrated in situ in the laboratory furnace, using a type B reference TC as the true furnace temperature measurement, as per the American Society of Testing and Materials E2846-20 specification [2]. The type B reference TC is a platinum/rhodium high-temperature design with a known published response and negligible error. The reference type B TC was calibrated and featured a National Institute of Standards and Technology traceable calibration number (i.e., 75036B).

The HTIR-TC is tightly wrapped in an Nb foil with a thickness of 0.001–0.002 in. (This provides a gettering process for any remaining oxygen in the system, thus preventing embrittlement of the TC.) The TC is next installed within an inert atmosphere capable of reaching 1450–1600°C. A type B reference TC is installed adjacent to the HTIR-TC tip, within ¼-in. spacing. The HTIR-TC is wired through an ice point cell held at 0°C, ensuring a constant reference temperature for the electromotive force (EMF) generated by the TC. Both the HTIR-TC and type B reference TC are hardwired into the computer data acquisition system, which is capable of measuring TC voltage and/or temperature.

3. FURNACE TEMPERATURE CYCLE

With the setup completed, the HTIR-TC can now be heated over a range of temperatures while recording both the HTIR-TC voltages and the NIST standard, type B TC known temperatures. Ensure the voltage and temperatures are being recorded at a sampling rate of approximately 1 Hz. Prior to a full heat-up cycle, the furnace must be brought to a constant temperature of 250°C for at least 2 hours to clean the environment of any organics. After, the temperature of the furnace can be brought up to 700°C at a rate of 5°C/min. Hold at this temperature for 1 hour or until the temperature has remained steady for 30 minutes. This is then repeated by increasing the furnace temperature in steps of 100°C until reaching a maximum temperature equal to or less than the sensor's associated heat treatment temperature. This maximum temperature is held for twice as long (e.g., two 1-hour segments) to begin the cycle in reverse. The temperature is then decreased in steps of 100°C, holding at the end of each step, until 700°C is reached. **Error! Reference source not found.** shows a typical HTIR-TC output voltage as the furnace undergoes this up and down cycle, with the left axis showing the voltage (V) from the HTIR-TC and the right axis showing the associated temperature as measured by the type B reference TC.

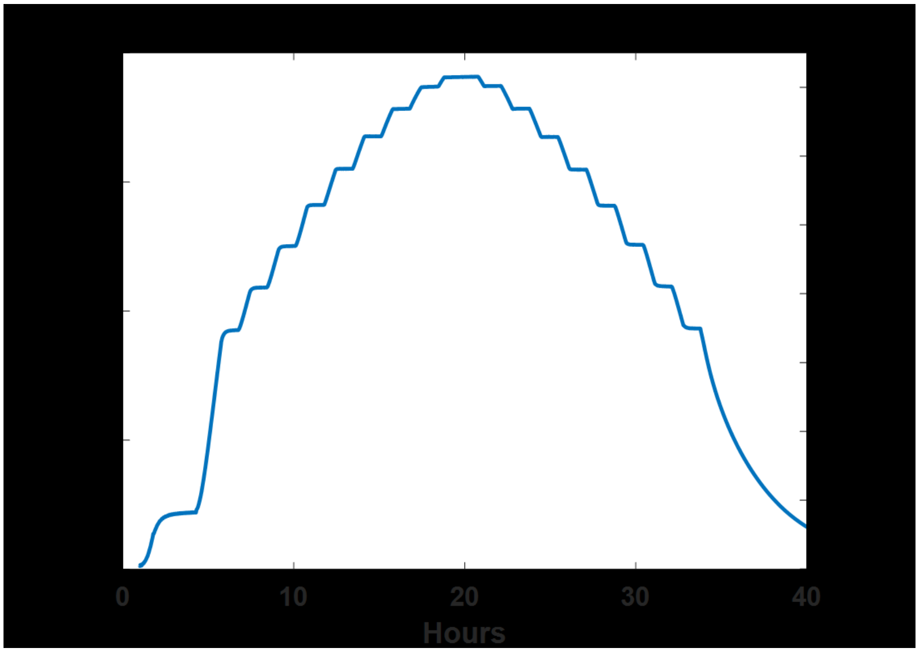

Figure 1. A typical calibration data sequence of the HTIR-TC in the furnace. The true furnace temperature was determined by the reference type B TC positioned less than a quarter inch from the tip of the HTIR-TC. The calibration data consist of the time-averaged values of the TC voltage output and the reference type B TC temperature reading at each temperature hold.

4. CALIBRATION SOFTWARE

Once the collecting of voltages at the various temperatures is complete, the voltage-vs.-temperature data are fit via a 5th order polynomial using the least-squares method. A graphical user interface (GUI) was created in the MATLAB environment to aide in the calibration process. This GUI can be seen in **Error! Reference source not found.**.

The following is a guide to using the GUI:

File Inputs:

1. The HTIR-TC voltage and type B temperature .txt files are loaded
2. The associated .txt column and number of calibration zones (i.e., 100°C increments) are input
3. This is followed by plotting the calibration curve and hand-picking the regions of interest
4. This is immediately followed by plotting the repeatability test and selecting the identical curve locations on the latter half of the calibration curve.

Calibration:

1. A drop-down menu allows users to select the polynomial degree (5th order was shown to work best)
2. Any extra data pairs of voltage and temperature can now be entered by hand, if needed
3. If desired, the origin of the plot—representing 0 V at 0°C—can be checked
4. Advanced: The Force extrema (min) at origin? checkbox can be checked, but this is not recommended.

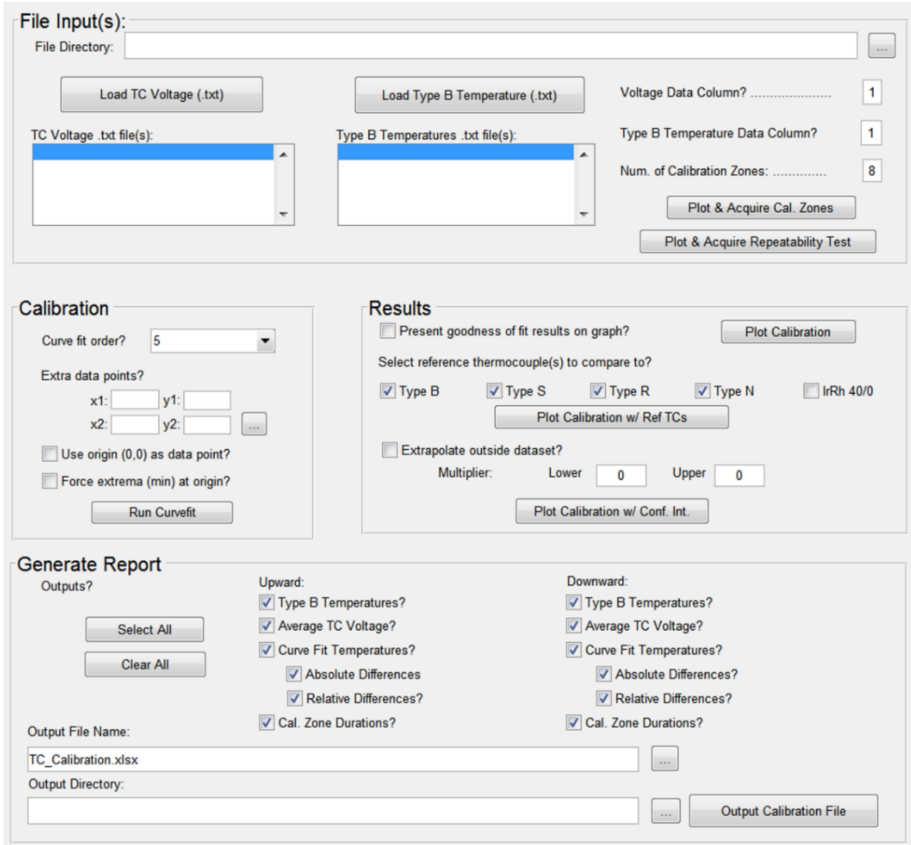

Figure 2. The GUI designed to aid in calibrating HTIR-TCs consistently.

Results:

1. In the Results section, the calibration curve can be presented by clicking Plot Calibration
2. The goodness of fit—represented as the sum of the squares of the error—can be presented on the plot by checking Present goodness of fit results on graph?
3. Comparative plots can be generated for comparing the current HTIR-TC calibration with other commercially available EMF curves from other TC types
4. Advanced: The calibration curve can be extrapolated outside the bounds of the standard plot by multiplying above (i.e., Upper) and below (i.e., Lower) the standard curve
5. The confidence interval curves at the 95% level can be plotted on the top and bottom of the calibration curve.

Generate Report:

1. A calibration report can be generated and output to an Excel spreadsheet
2. The individual checkboxes coincide with the columns of the spreadsheet report
    a. Type B Temperatures: The average type B TC measurement for the individual calibration zones
    b. Average TC Voltage: The average HTIR-TC voltage measurement for the individual calibration zones
    c. Curve Fit Temperatures: The associated temperature from the HTIR-TC voltage to the calibration. Must select one or both options: Absolute and/or Relative Difference.

   d. Cal. Zone Duration: The amount of time incurred for the time-averaged values above

3. An output file and directory can now be input by the user, and the file can be saved. The directory is, by default, where the voltage and temperature .txt files are located.

5. PRELIMINARY RESULTS

Utilizing the least squares method, HTIR-TCs have been successfully calibrated using a 5th order polynomial curve fit [3]. An evaluation of the 3rd, 4th, 5th, and 6th order polynomials was conducted to determine the best polynomial fit to the calibration data. The results showed that polynomial orders of less than five left too high a residual error for the least squares fit, while the improvement for polynomial orders higher than five was insignificant and also resulted in lower degrees of freedom that were deemed inappropriate for the number of data points to be fitted. Since the 5th order polynomial resulted in acceptably small residuals for all HTIR-TCs, it was used as the basis for correlating the HTIR-TC measured output voltage to the measured temperature.

The data from the furnace temperature cycle are time averaged at each temperature hold. The time-averaged voltage (mV) outputted by the HTIR-TC is paired to the time-averaged type B TC temperature (°C) measurements, and the measured temperature T(mV) is determined using the following general 5th order polynomial equation:

$$T(V) = a_0 + a_1V + a_2V^2 + a_3V^3 + a_4V^4 + a_5V^5 \tag{1}$$

The measurement error E(j) in temperature units (°C) at hold point j is the difference between the measured temperature $T(mV_j)$ at hold point j and the corresponding type B TC calibration source temperature $T(B_j)$ at hold point j.

$$E(j) = T(V_j) - T(B_j)\ [°C] \tag{2}$$

The measurement error in % is $100*E(j)/T(B_j)$ at the calibration temperature $T(B_j)$. Note that the error of the processing electronics is negligible and does not contribute to the measurement error—as seen in Section 5.3.

The calibration data shows that the HTIR-TC response when the furnace temperature is rising is nearly the same as when it is falling, indicating that the HTIR-TC temperature measurement is reproducible, with no hysteresis errors. The repeatability result was calculated to be ~0.3% which is within the 1% accuracy requirement. Repeatability was determined during calibration by calculating the HTIR-TC measured temperature at each hold point in the calibration up and down cycle corresponding to the average temperature measured by the NIST standard at each hold point.

A numerical evaluation of the repeatability of the HTIR-TC is shown below in Tables 1 and 2 for two typical HTIR-TC thermocouples. The repeatability was calculated using the following procedure:

1) Determine the temperature measured by the NIST standard at the various hold points in both the up and down directions
2) Determine the voltage measured by HTIR-TC at the hold points in both the up and down directions and convert them to temperature using the 5th order polynomial
3) Calculate Average temperature measured by NIST standard at the various hold points
4) For each hold point, adjust the HTIR-TC measured temperature in the up direction (HTIR-TC (up, adjusted)) to correspond to the average NIST standard temperature at that hold point by multiplying the measured HTIR-TC up temperature by the ratio of the average NIST Standard average temperature to NIST standard up temperature, as follows:
   HTIR-TC (up, adjusted) = HTIR-TC (up) * NIST Standard (average)/NIST Standard (up)
5) For each hold point, adjust the HTIR-TC measured temperature in the down direction (HTIR-TC (down, adjusted)) to correspond to the average NIST standard temperature at that hold point by multiplying the measured HTIR-TC up temperature by the ratio of the average NIST Standard average temperature to NIST standard down temperature as follows:

HTIR-TC (down, adjusted) = HTIR-TC (down) * NIST Standard (average) / NIST (down)

6) Repeatability was determined by subtracting the adjusted HTIR-TC up and down temperatures and dividing by the NIST standard average temperature for each hold point as follows:

Repeatability (%) = 100* [HTIR-TC (up, adjusted) - HTIR-TC (down, adjusted)] / NIST (average)

The results of this calculation showed that the measured repeatability over the full measured temperature range was ~ ±0.3%, well below the requirement of ±0.5°C.

Table 1. The repeatability of HTIR-TC 1-14 during the calibration cycle in an out-of-pile furnace—prior to irradiation.

Hold Point	Temperature data of upward profile (°C)		Temperature data of downward profile (°C)		Average Temperature (°C)	Measured Average Temperature (°C)		Reproducibility between upward and downward temperature profiles	
	NIST Standard	HTIR-TC	NIST Standard	HTIR-TC	NIST Standard	HTIR-TC (upward)	HTIR-TC (downward)	°C	%
1	709.8	709.1	713.7	711.9	711.8	711.0	709.9	1.1	0.2
2	805.7	806.1	810.6	809.8	808.2	808.6	807.4	1.2	0.2
3	903.6	904.8	909.2	909.0	906.4	907.6	906.2	1.5	0.2
4	1005.2	1005.1	1006.2	1004.5	1005.7	1005.6	1004.1	1.5	0.1
5	1101.1	1099.2	1102.1	1098.7	1101.6	1099.8	1098.2	1.6	0.1
6	1196.8	1196.3	1198.0	1195.0	1197.4	1196.8	1194.5	2.3	0.2
7	1293.8	1297.0	1294.7	1294.2	1294.2	1297.4	1293.8	3.6	0.3
8	1391.4	1389.8	1391.7	1390.1	1391.5	1389.9	1390.0	-0.1	0.0

Table 2. The repeatability of HTIR-TC 3-5 during the calibration cycle in an out-of-pile furnace—prior to irradiation.

Hold Point	Temperature data of upward profile (°C)		Temperature data of downward profile (°C)		Average Temperature (°C)	Measured Average Temperature (°C)		Reproducibility between upward and downward temperature profiles	
	NIST Standard	HTIR-TC	NIST Standard	HTIR-TC	NIST Standard	HTIR-TC (upward)	HTIR-TC (downward)	°C	%
1	709.1	709.1	714.0	711.7	711.5	711.5	709.3	2.2	0.3
2	805.6	805.4	810.9	808.3	808.3	808.1	805.7	2.4	0.3
3	903.5	904.0	909.3	907.3	906.4	906.9	904.4	2.4	0.3
4	1005.5	1005.5	1006.4	1004.1	1005.9	1006.0	1003.6	2.4	0.2
5	1101.4	1100.9	1102.6	1099.2	1102.0	1101.5	1098.7	2.8	0.3
6	1197.2	1197.0	1198.7	1195.5	1197.9	1197.7	1194.8	2.9	0.2
7	1294.1	1294.9	1295.3	1294.1	1294.7	1295.5	1293.5	2.0	0.2
8	1391.7	1391.4	1392.1	1391.7	1391.9	1391.6	1391.5	0.1	0.0

5.1 Individual Calibration

A list of the polynomial coefficients for the 10 HTIR-TC individual calibrations is shown in Table 3. (The sensor numbering is taken from the Advanced Gas Reactor (AGR) 5/6/7 test.)

Table 3. Calibration coefficients for the 10 HTIR-TCs from the AGR 5/6/7 test. Also listed is the observed measurement error of the calibrated HTIR-TC temperature compared to the type B TC reference measurement.

Sensor #	a_0	$a_1 \times 10^5$	$a_2 \times 10^7$	$a_3 \times 10^9$	$a_4 \times 10^{11}$	$a_5 \times 10^{12}$
TC-1-9	2.5586	1.5898	-2.4652	2.7917	-1.4937	3.0969
TC-1-10	1.4062	1.5997	-2.3369	2.4859	-1.2600	2.5074
TC-1-11	1.5198	1.5920	-2.4379	2.7115	-1.4229	2.8894
TC-1-12	2.8347	1.5670	-2.2598	2.4201	-1.2384	2.4891
TC-1-13	1.8931	1.5821	-2.3339	2.5420	-1.3111	2.6251

TC-1-14	2.3167	1.5844	-2.3149	2.4588	-1.2364	2.4226
TC-1-15	2.6841	1.5857	-2.1929	2.1709	-1.0184	1.8902
TC-1-16	1.4913	1.5871	-2.3790	2.6083	-1.3518	2.7185
TC-3-5	-0.3713	1.6518	-2.4254	2.5121	-1.2321	2.3740
TC-3-12	1.5757	1.6337	-2.3986	2.5145	-1.2553	2.4806

The calibration data and the coefficients of the 5th order polynomial slightly vary for the different HTIR-TCs, indicating the uniqueness of each HTIR-TC. For maximum accuracy, each must be calibrated separately before being used to measure temperature. However, the data show that the coefficients for all 10 TCs are approximately the same. This is as expected, since all the TCs were manufactured via the same process.

To determine whether the error for the HTIR-TC design falls within the requirements laid out in the functional and operational requirements (F&OR) report [5], the statistical error limit for the group of tested HTIR-TCs must be determined. To determine the statistical 3-σ limit of the measurement error, the evaluation was conducted in both a high and a low temperature range.

In the high temperature range (700–1400°C) the errors in °C increase as the temperature increases, thus the % error limit was determined by calculating the 3-σ limit of the relative errors of all the tested HTIR-TCs. For the group of data in this range, the 3-σ error limit was determined by adding triple the standard deviation to the absolute value of the average. Though the average is close to zero, it is conservatively accounted for by using its absolute value to give the same error limit in both the positive and negative directions.

In the low temperature range (room temperature to 100°C), the error in °C is small, and the error limit in °C was determined by calculating the 3-σ error limit of the absolute °C errors of all tested HTIR-TCs.

An error plot for the individual calibration case is shown in Figure 3. The data points, •, reflect the measured HTIR-TC temperature minus the reference type B TC temperature measured from 700 to 1400°C in 100°C increments for the high temperature range, and from room temperature to 100°C in the low temperature range. The black line above and below the data is the ±3 σ error limit (containing 99.7% of the data), which was determined to be ±0.4% in the high temperature range and ±0.86°C in the low temperature range. The red trace is the requirement of ±1% in the high temperature range and ±1°C in the low temperature range. The results show that, in the high temperature range, the measured error limit of ±0.4% is well below the ±1% requirement. For the low temperature range, the measured 3-σ error limit is ±0.86°C, which is below the requirement of ±1°C for this range. Note that no outliers were determined in this dataset of 100 samples.

It can be concluded that, for individual calibrations—with each HTIR-TC being calibrated separately and relying on its own set of calibration coefficients—the measured error meets the F&OR report's requirement for both the high and low temperature ranges. For TCs, a commonly accepted method of stating the requirement over the full temperature measurement range is that, for individual calibrations, the error is 1% of point or 1°C, whichever comes first. Note that, although the HTIR-TC response was determined for calibration data collected at up to 1400°C—and good results at up to 1500°C were obtained in the AGR 5/6/7 test [4]—the response can be extrapolated to 1600°C. This is a minor extrapolation that, based on engineering judgement, is justified by the qualification procedures [6].

5.2 Common Calibration

The calibration data were also analyzed to determine the error if a common set of coefficients was used for all the HTIR-TC samples. Using a common set of coefficients for all such HTIR-TCs would increase the measurement error, due to the inherent variability in the samples. However, as stated in the F&OR report, common coefficients may be justified in terms of convenience for commercial use, assuming the common calibration error requirements meet the application needs. Using a common set of predetermined coefficients for all HTIR-TCs whenever a new batch is manufactured for customer delivery allows for the calibration of only a fraction (~10%) of random samples in the batch—as opposed to calibrating every manufactured HTIR-TC—to ensure compliance with the common calibration specifications.

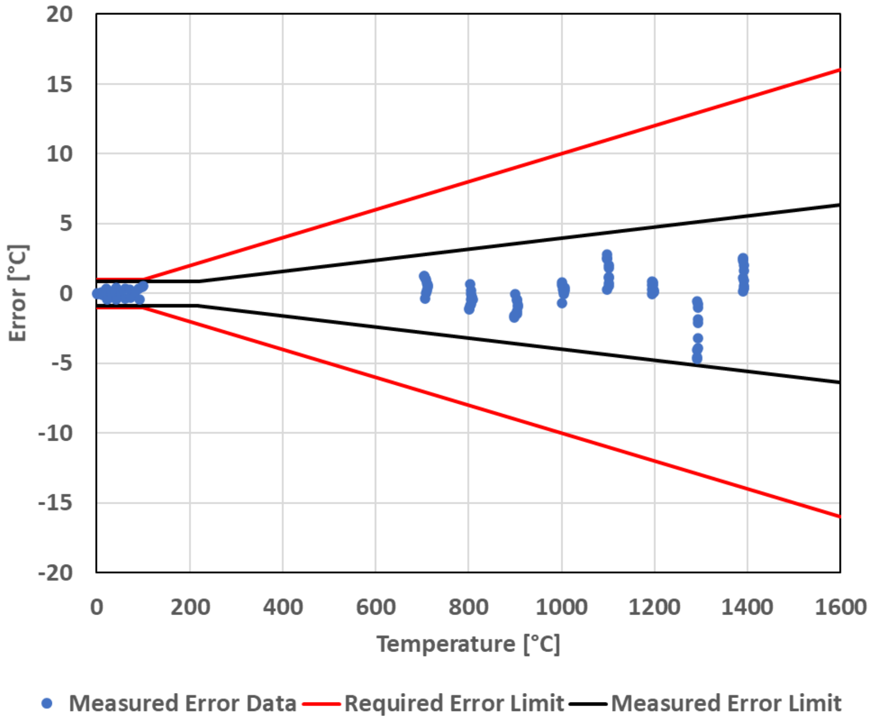

Figure 3. Measured error of individual calibrations vs. the requirements. The black trace represents the 3-σ error limit measured by the dataset at ~0.40% in the high temperature region and ~0.86°C in the low temperature region. The red trace represents the requirement of ±1% in the high temperature region and ±1°C in the low temperature region.

The calibration data for all 10 HTIR-TCs using a common set of polynomial coefficients were determined by first analyzing the set of 10 HTIR-TCs for differences in mean error. One of the 10 HTIR-TCs was determined to be statistically different enough to justify removing it from the dataset. Upon doing so, a single least squares curve fit was applied to the remaining dataset as a whole. The coefficients for the 5th order polynomial are as follows:

$$a_5 = 2.131 \text{ x } 10^{12}$$

$$a_4 = -1.095 \text{ x } 10^{11}$$

$$a_3 = 2.233 \text{ x } 10^{9}$$

$$a_2 = -2.182 \text{ x } 10^{7}$$

$$a_1 = 1.567 \text{ x } 10^{5}$$

$$a_0 = 2.453$$

A plot of the errors for the common calibration case is shown in Figure 4. As for the case of individual coefficients in Figure 3, the data points are the measured HTIR-TC temperature minus the reference type B TC temperature measured from 700 to 1400°C in 100°C increments for the high temperature range, and room temperature to 100°C for the low temperature range. The black line above and below the data is the ±3 σ limit (containing 99.7% of the data), which was determined to be ±1.8% in the high temperature range and 2.3°C in the low temperature range. The red trace is the requirement of ±2% in the high temperature range and ±2.5°C in the low temperature range. The results show that, in the

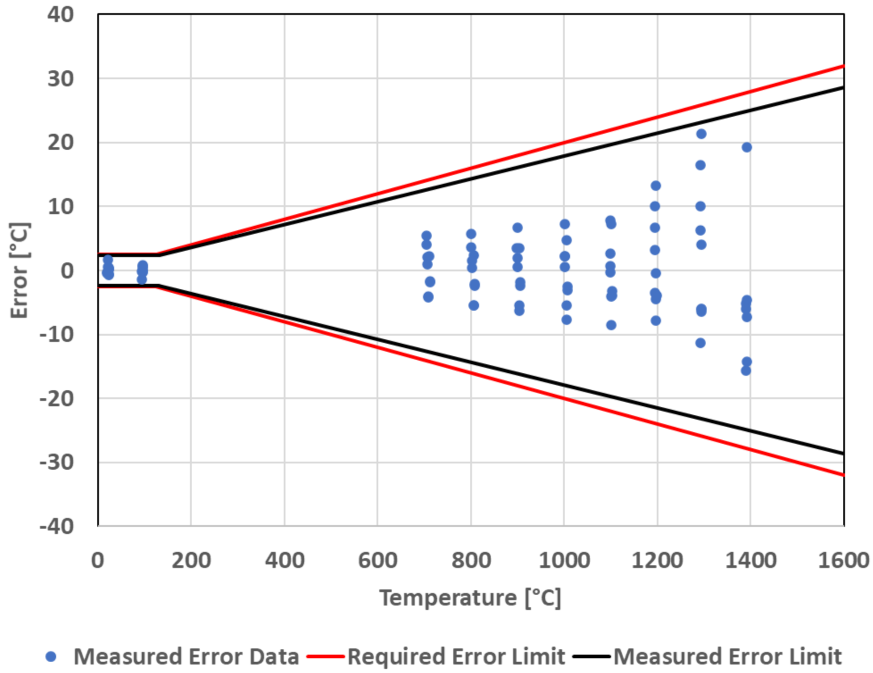

Figure 4. Measured error of common calibrations vs. the requirements. The black trace represents the 3-σ error limit measured by the dataset at ~1.8% in the high temperature region and ~2.3°C in the low temperature region. The red trace represents the requirement of ±2% in the high temperature region and ±2.5°C in the low temperature region.

high temperature range, the measured error limit of ±1.8% is below the ±2% requirement. For the low temperature range, the measured 3-σ error limit is ±2.3°C, which is below the requirement of ±2.5°C.

It can be concluded that, for common calibration—in which a common set of coefficients is used for a full set of HTIR-TCs—the measured error meets the F&OR report's requirement for both the high and low temperature ranges. An acceptable method of stating the requirement over the full temperature measurement range for common calibration is that the error is 2% of point or 2.5°C, whichever comes first.

Note that the error for common calibration is approximately double that of individual calibrations, making it likely that, where high accuracy is required (e.g., in-pile, high-temperature measurements), the HTIR-TCs will be individually calibrated. But for other commercial uses (less accurate lower temperature measurements, perhaps), the common calibration technique can help facilitate the commercial delivery process.

5.3 Measurement and Test Equipment (M&TE) Errors

The M&TE devices in the HTIR-TC temperature measurement loop included the Voltage measurement device and the zero-temperature device. The overall error of the HTIR-TC temperature measurement is the square root of the sum of the squares (SRSS) of the errors of the HTIR-TC and the errors of the M&TE devices. The HTIR-TC measurement errors reported in Section 5.1 and 5.2 include the errors from all the M&TE devices. However, since the M&TE errors are small, the error is conservatively reported as the HTIR-TC thermocouple error. Following is a calculation of the M&TE device errors:

a) The data acquisition system (DAS) is model NI-1102B, and the specified accuracy of this device is 0.015% the reading.

b) The temperature reference device is a TRCIII ice-point reference cell from Omega with a specified accuracy of 0.1°C.

c) The total M&TE error, E_T, is calculated by combining the error sources—with like units—by utilizing the following

, (4)

where x_i is the error from an individual source. In this case there are two error sources, the DAS and the ice-point reference cell. The error domain is further discretized into temperature ranges as seen below.

M&TE = Statistical combination of error of ([A]0.1°C and [B]0.015%)

$$\text{M\&TE}(0°C < T \leq 100°C) = 0.10°C = \sqrt{}(0.1^2 + (0.015 \times 100/100)^2)\ °C$$

$$\text{M\&TE}(100°C < T \leq 500°C) < 0.10\% = \sqrt{}\ ((0.1*100/100)^2 + 0.015^2)\ \%$$

$$\text{M\&TE}(500°C < T \leq 1000°C) < 0.03\% = \sqrt{}\ ((0.1*100/500)^2 + 0.015^2)\ \%$$

$$\text{M\&TE}(1000°C < T) < 0.02\% = \sqrt{}\ ((0.1*100/1000)^2 + 0.015^2)\ \%$$

A, reported error in Omega Ice-point Cell [7]

B, reported error in NI DAS SCXI 1303 32-channel temperature block [8]

This translates to a depreciating error at higher temperatures starting at 0.10°C for temperature measurements below 100°C, less than 0.1% of the temperature measurements between 100°C – 500°C, less than 0.03% for measurements between 500°C – 1000°C, and lastly less than 0.02% for temperature measurements above 1000°C.

d) The calculation shows the M&TE errors are much less than the HTIR-TC measurement errors so it can be conservatively assumed that the overall measurement errors reported in Section 5.1 and 5.2, which include the M&TE errors, are the HTIR-TC errors.

6. REFERENCES

1. Rempe, J.L., Knudson, D.L., Condie, K.G., Wilkins, S.C., "Thermocouples for High Temperature In-Pile Testing," *Nuclear Technology* 156, no. 3, (2006): 320-331, https://doi.org/10.13182/NT06-A3794.
2. ASTM Standard E2846-20 , "Standard Guide for Thermocouple Verification," *ASTM International*, (2020), www.astm.org
3. Björck, Å., "Numerical Methods for Least Squares Problems," SIAM, ISBN978-0-89871-360-2, (1996).
4. Skifton, R., "Function & Operational Requirements for High Temperature Irradiation Resistant Thermocouples developed by the In-Pile Instrumentation Program," INL/EXT-21-63173, 2021.
5. Palmer, A.J., Skifton, R.S., Scervini, M., Haggard, D.C., Swank, W.D., "Performance of Custom-Made Very High Temperature Thermocouples in the Advanced Gas Reactor Experiment AGR-5/6/7 during Irradiation in the Advanced Test Reactor," *EPJ Web Conf.* 225 no. 04010, (2020), ANIMMA 2019, https://doi.org/10.1051/epjconf/202022504010.
6. Dayal, Y., Jensen, C., "Guidelines for Developing and Qualifying Instrumentation Systems at Idaho National Laboratory", Idaho National Laboratory, Document ID: GDE-947 Rev 0 (2019).
7. Omega, Ice-point Cell, Datasheet, https://assets.omega.com/spec/TRCIII.pdf

8. National Instruments, SCXI 1102 32-channel Temperature block, Datasheet, https://www.ni.com/datasheet/pdf/en/ds-206, 2014