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ABSTRACT

Over the preceding ten years, continual research and development has been performed on the high temperature irradiation resistant thermocouple (HTIR-TC) by the team at Idaho National Laboratories (INL) High Temperature Test Laboratory (HTTL). The HTIR-TC has the capability of achieving high temperatures up to 1600°C or more. Further, the HTIR-TC has gone through many longevity tests both in and out of pile to show the amount of drift is minimal compared standard thermocouples. Key considerations for properties and materials of the HTIR-TC have been final diameter, ductility after heat treatment, and minimizing transmutation of materials during operation. Further, heat treatment and calibration processes have been established in order to consistently produce repeatable and reliable temperature readings. The current work provides further background for the standardization process of the HTIR-TC.

Key Words: In-pile, instrumentation, thermocouple, HTIR-TC, High Temperature

1 INTRODUCTION

As the name implies, the High Temperature Irradiation Resistant Thermocouple (HTIR-TC), is the world's leading temperature probe for high temperature environments in radioactive environments, e.g. nuclear reactors. The current work performed at the Idaho National Laboratory (INL) High Temperature Test Laboratory (HTTL) in conjuction with Boise State University (BSU) and Idaho Laboratories Corp (ILC) has shown the HTIR-TC have lasting results in the reactor at 1450°C and the TC could eventually be pushed to the range of 1600°C or beyond. There is also reason to believe the HTIR-TC outlasts – in the max range – some of the lower temperature thermocouples (e.g. Type K and Type N) around 1000°C to 1200°C [1]. Past research studies have shown the HTIR-TC build/materials down selection, heat treatment process, and calibration technique utilized in building a single HTIR-TC [1, 2]. Further work has shown that HTIR-TC can withstand high temperatures, multiple thermal cycles, and in-pile irradiation for extended periods of time. These studies have shown specifically that the HTIR-TC can go without significant thermocouple aging (drift) and/or transmutation to the thermoelements and other components of the TC [3]. The main advantage over other commercially available thermocouples is the low calibration drifting at these heightened temperatures. The purpose of this current work is to segue into aiding the commercialization process of HTIR-TCs by providing Seebeck coefficients for the individual thermoelements similar to that in ASTM E207 – 08 [4], an averaged calibration curve to be used in conjunction with the ASTM E220 – 13 [5], and drift analysis for verification purposes in ASTM E2846 – 14 [6].

2 HTIR-TC THERMOCOUPLE

The HTIR-TC is comprised of a doped molybdenum and doped niobium wire that form a junction at one end. The HTIR-TC is pre-constructed prior to any heat treatments or individual calibrations taking place. This avoids thermoelements that are too brittle to work with and ultimately swaged to a final overall diameter. Although, future work may address the option of heat treating individual spools of wire in a vacuum or other specialty furnace prior to constructing the thermocouple. This will avoid the unnecessary amount of time taken to perform an individual heat treatment on each TC, and to avoid EMF accidentally being generated in the untreated sections of the wires [2].

2.1 Thermal Electromotive Force (EMF) of a HTIR-TC Individual Thermoelement

Similar to the procedure performed in ASTM E207-08 [4], the individual molybdenum and niobium thermoelements were placed into their own respective thermocouples with pure platinum as the reference leg. These secondary thermocouples were then placed at the centerline of a tube furnace and heated gradually to 1600°C at 5°C/min. Upon reaching this max temperature, the furnace was held there for 72+ hours, immediately following a cool off phase back to room temperature at 5°C/min until the natural cooling off the furnace took over. The voltage of each thermocouple was recorded throughout the entire process.

The EMF generated in each individual leg sums to a total EMF generated in the overall thermocouple by

$$E_T = E_{ref,Pt} + E_{xr} \tag{1}$$

where E_T is the total EMF generated in the individual thermocouples, $E_{ref,Pt}$ is the contribution to the EMF from the platinum leg, and E_{xr} is the contribution to the EMF from the thermoelement being considered that is cross referenced with the platinum leg.

The voltage of pure platinum can be represented by a 5th order polynomial

$$E_{ref,Pt}(T) = AT^5+BT^4+CT^3+DT^2+ET+F. \tag{2}$$

By doing a least squares fit of data from [7] the 5th order polynomial in Eq. 2 has units of µV, where T is the sensing temperature of the platinum (assuming a 0°C reference temperature), and the coefficients to the curve fit can be seen in Table I.

Algebraically rearranging Eq. 1 to solve for E_{xr} provides a curve of EMF for each individual leg of lanthanated molybdenum (La-Mo) and phosphorous doped niobium (P-Nb), respectively. The resultant curves were then, in turn, algebraically combined to make an indirect (or virtual) thermocouple comprising of the two thermoelements utilized in an actual HTIR-TC. In short,

$$E_T = E_{xr,1} - E_{xr,2}\ , \tag{3}$$

where $E_{xr,1}$ is the contribution to the total EMF from the positive leg of the thermocouple, and $E_{xr,2}$ is the contribution from the negative leg of the thermocouple. In the HTIR-TC the La-Mo corresponds to the positive leg and P-Nb is the negative leg.

Table I. The coefficients of a least squares curve fit to Pure Platinum Seebeck Coefficients

Coefficient	Platinum
A	-8.79464×10^{-15}
B	5.33941×10^{-11}
C	-1.23534×10^{-7}
D	1.36635×10^{-4}
E	-8.70359×10^{-2}
F	11.6316

The EMF results of the indirect thermocouple junction can be seen in Figure 1, where the two solid lines coincide with the before and after heat treatment processes on either end of the 72+ hours at 1600°C mentioned above. They can be seen to the far left and far right of the figure, respectively. The dotted line is a trend line of the after heat treatment curve of the indirect thermocouple and coincides with a direct HTIR-TC (dashed) after the thermocouple has been heat treated.

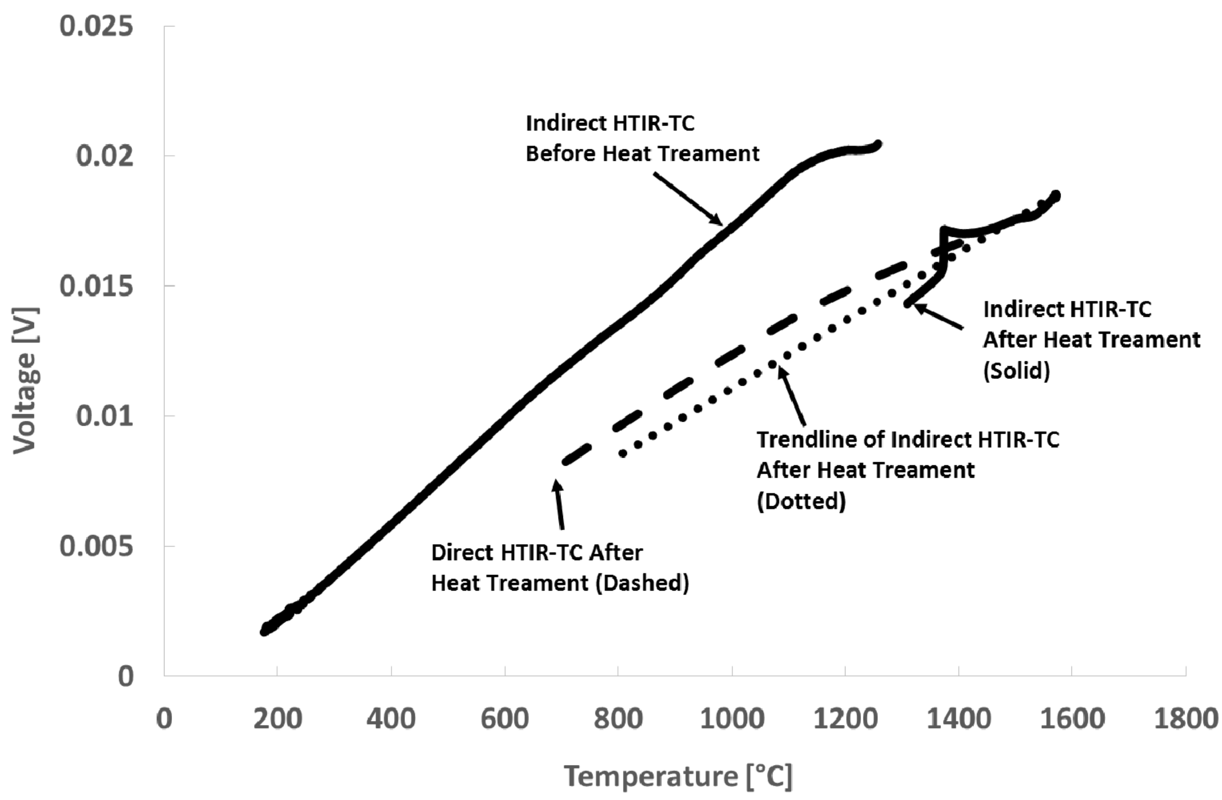

Figure 1. The indirect thermocouple EMF generated from removing the effect of pure platinum from a secondary thermocouple. The before and after heat treatment curves are so labeled and can be seen to the far left and far right of the figure, respectively. The trend of the after heat treated indirect thermocouple corresponds to a direct HTIR-TC after the respective heat treatment.

2.2 Single Calibration Curve Standard for all HTIR-TCs

To date, the standard procedure for calibrating the HTIR-TC has been to individually calibrate each one as they are built. The natural evolution would be to have one standard calibration curve for all HTIR-TCs, given that their chemistry and manufacturing are near identical – similar to all commercially available thermocouples. The large database of HTIR thermocouples that have been built has produced a large array of individual calibration curves. A select few of the calibration curves can be seen in Figure 2 as solid colored lines. By averaging the coefficients to each least squares fitted curve (i.e. a 5th order polynomial), an average curve was generated as seen in Figure 2 as a black, dashed line. In short,

$$(\bar{A}, \bar{B}, \bar{C}, \bar{D}, \bar{E}, \bar{F}\ ) = \frac{1}{N}\sum_{i=1}^{N}(A_i, B_i, C_i, D_i, E_i, F_i) \quad (4)$$

where the overbar represents the average coefficient from the many HTIR-TCs that have been calibrated to date, the subscript i represents the per each HTIR-TC calibration index, and N is the total number of HTIR-TCs calibrated.

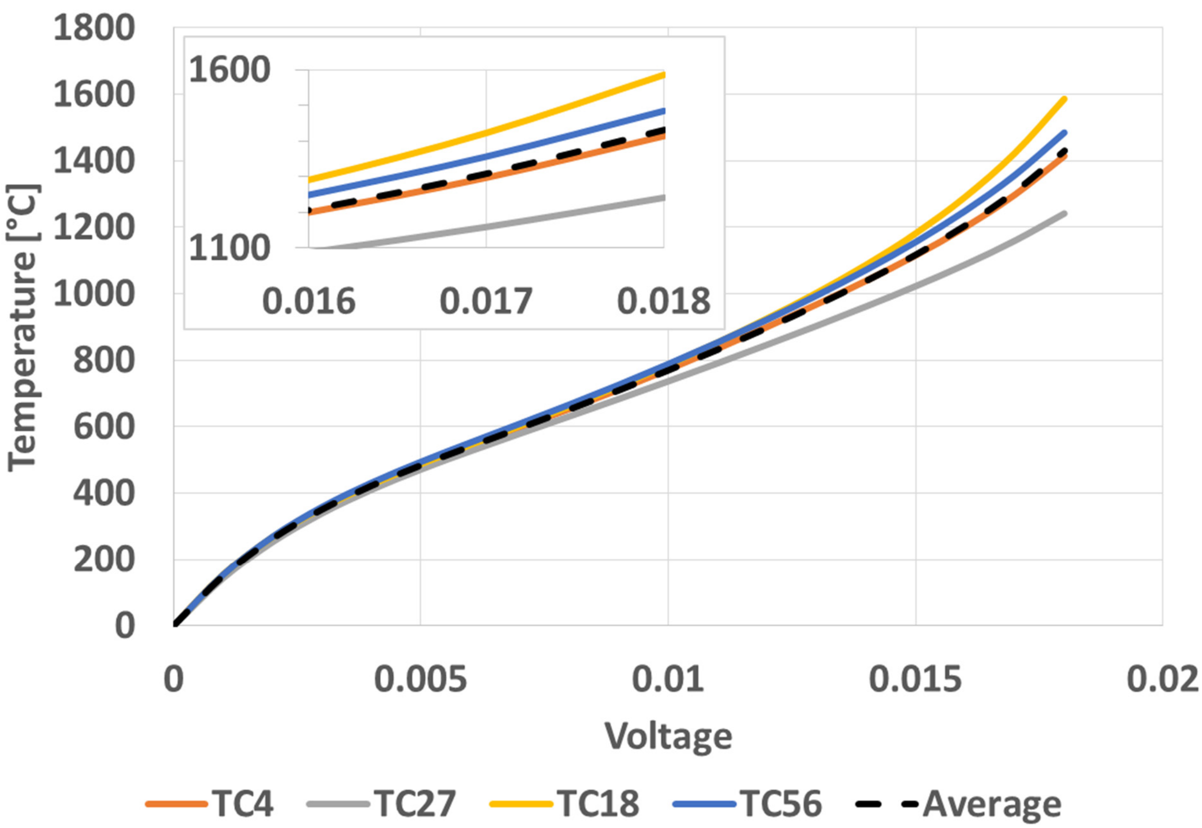

Figure 2. A set of calibration curves for the HTIR-TC from 0°C to ~1600°C. The black, dashed line is the average calibration for all HTIR-TCs to date. The insert is a zoomed view of the last ~500°C of the thermocouple range. The grey and orange curves are representative of the worst-case-scenario seen so far.

The resultant values from Eq. 4 for the overall HTIR-TC calibration coefficients can be seen in Table II, where the constant term was forced to be 0 under the assumption that the 0°C reference temperature is in fact 0 Volts. These coefficients will prove to be useful in the near future when the HTIR-TC is standardized in either ASTM E230/E230M-17 as a lettered thermocouple or ASTM E608/E608M-13 as a base metal thermocouple.

Table II. The averaged coefficients of all least squares curve fits for HTIR-TCs that have been calibrated to date.

Average Coefficient	
$\bar{A}$	2.5367×10^{12}
$\bar{B}$	-1.27917×10^{11}
$\bar{C}$	2599767053
$\bar{D}$	-25341242.23
$\bar{E}$	173060.805
$\bar{F}$	0

2.3 Longevity Test Using Different Insulation Packing

To show the effect of various thermocouple builds on the actual thermoelements themselves, three different insulators were used in a longevity test at elevated temperatures. The first thermocouple was constructed of HTIR-TC thermoelements and alumina (Al_2O_3) crushable insulators, the second was with magnesia (MgO), and the third with hafnia (HfO_2). Each were held in a well-controlled furnace at an elevated temperature of 1247°C +/- 5°C. Over 1,000 hours of data was recorded, and the overall trend in the drift can be seen in Figure 3 – where no more than a 0.6% °C temperature offset can be seen. The temperature of the furnace was spot checked weekly by a Type B thermocouple for a few hours at a time.

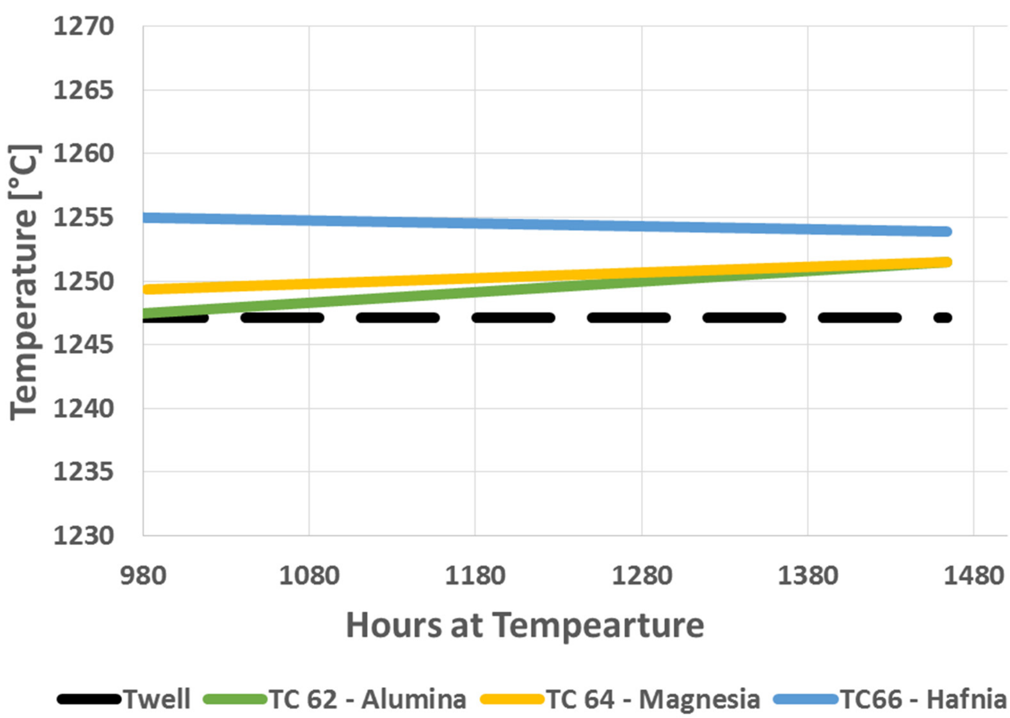

Figure 3. The trending results of a drift test using three different mineral insulations in a HTIR-TC. The T_{well} temperature control was spot checked weekly by a Type B thermocouple for a few hours at a time. The furnace itself never drifted more than +/- 5°C.

3 CONCLUSIONS

A series of tests have been set up to facility the standardization of the HTIR-TC. Individual EMF curves for each HTIR-TC thermoelement were calculated and algebraically combined to form an indirect (or virtual) thermocouple. This provides the EMF curve for each thermoelement of La-Mo and P-Nb. An averaged curve of past HTIR-TC calibration curves was presented to give an appropriate range of voltages vs. temperatures for the standardization of the HTIR-TC. Finally, a longevity test has been performed in a well controlled furnace at the elevated temperature of 1247°C. This latest test showed that the HTIR-TC never drifted more than 0.6% °C after 1,000+ hours of operation.

4 REFERENCES

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