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Function and Operational Requirements for High-Temperature Irradiation-Resistant Thermocouples

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SUMMARY

This report sets forth the function and operation requirements for the high-temperature irradiation-resistant thermocouple (HTIR-TC). A mission and product are proposed. The function and operational requirements involve temperature range, accuracy, drift, life, mechanical ruggedness, and response time. Each has a criterion set for establishing thermocouples (TCs) that can withstand a typical 18-month nuclear power plant (NPP) refueling cycle.

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ACRONYMS

ATR	Advanced Test Reactor
BWR	boiling-water reactor
EFPD	effective full-power days
HTIR-TC	high temperature irradiation resistant thermocouple
LOCA	loss-of-coolant accident
NPP	nuclear power plant
PWR	pressurized-water reactor
TC	thermocouple

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1. GENERAL

A temperature sensor that offers continued reliability in high-temperature/high-radiation environments is paramount for understanding nuclear fuel behavior under accident conditions—conditions in which both the existing nuclear power plant (NPP) fleet and upcoming advanced reactor concepts can experience temperatures pushing 1600°C. These advanced reactor concepts require extensive modeling and validation that must be verified with experimental work—specifically, testing performed on advanced fuel materials and form factors in U.S. material and test reactors. Most needed is a method of measuring fuel centerline temperatures. Furthermore, since advanced reactor concepts are proposed to operate at higher temperatures, they require measurements in a range similar to that of higher fuel cladding melt temperatures.

A technology gap exists in regard to temperature sensors that measure reactor cores in the range of 1290°C–1600°C. Reactor core temperature sensors are exposed to extremely high temperatures coupled with fast or thermal neutron bombardment. The reactor temperatures can sometimes exceed the melting temperatures of most other thermocouple (TC) candidate materials, and the high neutron flux leads to transmutation, changing the TC temperature response. Signal drift due to transmutation can be the most problematic, as the effect is subtle yet significant [1]. This gap is a serious problem for many advanced reactor concepts being proposed by commercial nuclear vendors, and only the high-temperature irradiation-resistant thermocouple (HTIR-TC) technology discussed herein has been able to address it.

2. OVERALL MISSION

The HTIR-TC's mission is twofold: (1) measure advanced fuel temperatures during experimental testing, and (2) measure NPP, reactor, and over-temperature conditions as a standalone device, particularly during accident scenarios.

2.1 Advanced Fuel Temperature Measurement Experiments

For new reactors still in the design phase, the design and development of new fuel is an important consideration. Development of new fuel necessitates measuring the fuel centerline temperature when the fuel is placed in a reactor neutron flux environment prototypic of the intended application. This temperature measurement provides data to assist in evaluating the fuel design, and is usually performed using a specially designed experimental rig containing the test fuel and a TC capable of measuring—as closely to the fuel centerline as possible—the high temperatures generated in the fuel.

2.2 Standalone Temperature Measurements

All nuclear reactors require temperature measurements at various locations, including the measurement of core inlet/outlet temperatures under normal and accident operating conditions. For this application, it is a requirement that the thermocouple provide acceptably accurate measurements for one fuel cycle which is typically 18 months but may be increased to 24 months. In recent years, measuring the temperatures generated close to the fuel during loss-of-coolant accidents (LOCAs), in which the fuel is partially or fully uncovered (e.g., what happened at Fukushima), has become of particular interest to NPPs operating in the U.S., Japan, and other countries throughout the world. If such an event ever again occurred, an accurate, high-temperature-measuring TC could be very useful in alerting operators to the possibility of fuel uncovery and potential fuel failure.

When used as a standalone device in a new or operating reactor, a reliable TC that produces accurate temperature measurements is desirable for both normal operation and for providing a margin to protect against fuel damage in the event of a LOCA. There is a need for accurate, high-temperature, standalone TCs in non-nuclear industrial applications, as well. The requirements that HTIC-TCs must meet to successfully perform their two basic functions are given in Section 4.

3. FUNCTIONAL DESCRIPTION OF THE PROPOSED PRODUCT

The HTIR-TC is comprised of molybdenum (Mo) and niobium (Nb). These refractory metals were specifically chosen due to their high melting temperatures and relatively low neutron absorption cross sections (see Table 1) [2], implying a relatively small number of transmutations during their residence inside the reactor. Such a TC would provide robust temperature measurements and an acceptably small drift due to transmutation and fast neutron damage during the TC's specified mission time inside the reactor. The melting temperatures of both elements exceed the HTIR-TC's specified maximum temperature measurement range.

As far as drift caused by transmutations is concerned, it should be noted that Mo is naturally endowed with approximately 6–7 stable isotopes, while Nb is essentially 100% ^{93}Nb. The neutron absorption cross sections and abundance of the Mo isotopes—along with the cross section of ^{93}Nb—are given in Table 1. The highest reported cross section (13.1 barns) lies with ^{95}Mo, which is naturally abundant at 15.92% of all the stable Mo isotopes. Mo's other stable isotopes have considerably lower cross sections. The cross section for ^{93}Nb is 1.15 barns [3]. The transmutations, primarily caused by thermal neutrons, would change the Seebeck coefficient (and, in turn, the sensitivity) and cause drift of the TC. For an HTIR-TC with two thermoelements, the absorption cross sections of both Mo and Nb would need to be considered in order to determine the transmutation rate applicable to the HTIR-TC.

The neutron absorption cross sections reported in Table 1 are 2200 m/s cross sections (based on a neutron velocity of 2200 m/s and a thermal neutron energy 0.0253 eV), and approximately correspond to those for thermal neutrons at room temperature (i.e., of 20°C). Note that, for thermal neutrons, the material-dependent cross sections decrease with increased thermal neutron temperature, T, at a rate of [4], making the cross section smaller for thermal neutrons at higher reactor temperatures. Note also that, as a general rule, the neutron absorption cross section is smaller for fast neutrons than for thermal neutrons, thus the neutron absorption cross sections reported in Table 1 are larger than they would be for a fast reactor. The absorption cross sections are most useful when discussing transmutations. The rate of transmutations is approximately proportional to the product of the absorption cross section and the thermal flux. For an HTIR-TC with two thermoelements, the combined absorption cross sections of Mo and Nb must be considered in order to determine the transmutation rate applicable to the HTIR-TC. The algebraically combined average absorption cross section is 3.63 barns. This is low enough to result in an acceptably low transmutation rate for most HTIR-TC applications, and matches the results obtained in previous experiments, as discussed in Section 4.3. Drift can also be caused by fast neutrons damaging the lattice structure and changing the TC's thermoelectric properties. It is not quantitatively known how this fast neutron bombardment contributes to changes in the thermoelectric behavior of HTIR-TCs, but the drift due to a certain fluence of fast neutrons is expected to be much less than that due to the same fluence of thermal neutrons.

In addition to the drift resulting from thermal and fast neutrons, drift at high temperatures can also be caused by prolonged residence time at high temperature. This drift is not dependent on neutron flux, but rather the difference between the temperature at which the TC was heat treated during manufacture and the temperature at which it is operating. Drift is expected—even when the operating temperature is less than the heat treatment temperature—but is predicted to decrease as the operating temperature decreases relative to the heat treatment temperature. This drift is due to potential metallurgical changes in the TC structure, and to the potential permeation of impurities into the TC as a result of prolonged lengths of time in operation at high temperature. Data from past experiments showing the magnitude of this high-

Table 1. Absorption cross sections for niobium and molybdenum. Table 1 also shows the natural abundance of Mo isotopes. The concertation of each Mo isotope is weighted in the average cross-section calculation.

Isotope	Concentration [%]	Absorption Cross Section [barns], σ_A
^{93}Nb	100	1.15
Mo (Weighted Average)	100	2.48
^{92}Mo	14.84	0.019
^{94}Mo	9.25	0.015
^{95}Mo	15.92	13.1
^{96}Mo	16.68	0.5
^{97}Mo	9.55	2.5
^{98}Mo	24.13	0.127
^{100}Mo	9.63	0.4

temperature drift will be used in the HTIR-TC Drift Model to determine the amount of drift during the qualification test.

Table 2 shows the high-temperature, irradiation-resistant capabilities of Mo and Nb thermoelements as well as other commercially available thermoelements. The material selection involves a balance between high temperatures and low neutron absorption cross sections, allowing for high-temperature measurements while simultaneously resisting the transmutation process that all materials undergo within a neutron flux field. The melting temperatures of Mo and Nb are higher than that of other commercially available base metal TCs, save tungsten and rhenium. However, when neutron absorption cross sections are also taken into consideration and compared, Mo and Nb are, understandably, the right selections.

Existing TC designs have either too low a maximum temperature range or, if maximum temperature is not an issue, the design quickly de-calibrates (e.g., drifts) under the reactor's neutron radiation effects.

Currently, type K and N TCs are utilized most often to measure temperatures in reactor cores but have an upper maximum temperature of 1290°C—several hundred °C less than the desired maximum temperature. Moreover, these TCs can only withstand this temperature for a short period of time. Further, they are used sparsely, and for fuel centerline measurements, they are placed farther from the fuel bundles

Table 2. Properties of base materials used in HTIR-TCs (Mo vs. Nb and others).

	Element	Atomic No.	Atomic Wt.	Density [kg/m3]x 10^{-3}	Melting Temperature [°C]	Absorption Cross Section [barns], σ_A[1]
HTIR	Niobium	41	92.906	8.57	2477	1.15
	Molybdenum	42	95.94	10.2	2623	*2.48
OTHERS	Nickel	28	58.71	8.90	1455	*4.49
	Tungsten	74	183.84	19.3	3422	*18.43
	Rhenium	75	186.207	21.02	3186	*89.7
	Platinum	78	195.09	21.45	1768	*10.3
	Rhodium	45	102.905	12.41	1964	144.8

[1]2200 m/sec cross sections (based on a neutron velocity of 2200 m/sec and a thermal neutron energy of 0.0253 eV)

*Average values of individual isotope cross sections

than desired. This means fuel temperatures must be approximated via interpolation between two or more low-temperature TCs [5].

4. PERFORMANCE REQUIREMENTS

This section covers the HTIC-TC performance requirements, along with the bases thereof. A summary of these requirements is given in Table 3.

4.1 Temperature Range

For application in advanced fuel temperature measurement experiments, the requirement is to measure temperature (ranging from room temperature to ~1600°C) in order to the evaluate the behavior of advanced fuels in a test reactor. The basis of the room temperature requirement is to ensure accurate temperature measurements, even when the reactor is shut down. The basis for the high temperature requirement is to evaluate several experiments designed to test advanced fuels in test reactors. For these experiments, the desirable maximum temperature was >1500°C, based on the results of theoretical models for calculating fuel temperature. Moreover, preliminary evaluations show that this temperature range can be measured by HTIR-TCs.

For standalone temperature measurements at various locations in operating nuclear power reactors (i.e., boiling-water reactors [BWRs] and pressurized-water reactors [PWRs]), the temperature range requirements are room temperature to <400°C for long-term operation under normal operating conditions, and high-temperature operation (~1000°C) for the purpose of measuring temperature during accidents (e.g, LOCAs) in which the core is partially or fully uncovered. Note that, since this standalone temperature range requirement is contained within the HTIR-TC fuel temperature measurement range, if the HTIR-TC meets the temperature range requirement for advanced fuel temperature measurement experiments, it will certainly meet the temperature range requirements for the standalone application.

4.2 Accuracy

The accuracy of fuel temperature measurements depends on the design of the experimental test fixture (e.g., the steepness of the temperature profile and the TC positioning error in the fixture) and the effect of immersion depth. Consequently, the accuracy of this fuel temperature measurement application depends on the design of the experimental fuel test fixture configuration and how well the temperature is controlled; thus, it cannot be well specified. However, the standalone accuracy requirement can be used as the basis for calculating the HTIR-TC accuracy in fuel temperature measurement experiments.

When each HTIR-TC is calibrated individually, the HTIR-TC accuracy requirement for standalone device measurements is ±1% of the instrument measurement error [6]. (This also includes the specified repeatability error.) The ±1% accuracy is a percent of point measurement, meaning that if the TC is reading 1000°C, the accuracy requirement is ±10°C absolute. The basis of this requirement is that it approximately matches the accuracy specifications of other commercially available TCs that cover a much lower temperature range. Moreover, in most NPP applications, the ±1% accuracy is generally sufficient for standalone TC measurements and is valid in the high-temperature region above 100°C, where the error would be greater than ±1°C absolute. That is considered reasonable, since HTIR-TCs are primarily intended for high-temperature measurements. For lower temperatures, the requirement is kept at a constant ±1°C, due to other factors such as the constancy of the cold reference junction temperature affecting the accuracy of low-temperature measurements.

If a common (or collective) calibration is used for all the HTIR-TCs, the accuracy requirement is doubled to ±2%, since the accuracy is expected to be lower than when each HTIR-TC is calibrated individually. This difference in the accuracy requirement is considered reasonable based on the potential differences in the samples and their polynomial fits. For lower temperatures, the requirement is kept at a constant ±2.5°C absolute. This is reasonable when using a common calibration, due to the expected variability in the low-temperature range. As for using individual calibrations, the ±2% accuracy is valid in the high-temperature region above 125°C, where the error would be greater than ±2.5°C absolute.

Although the overall accuracy requirement is less for common calibration than for individual calibration, use of the common calibration can be considered in order to facilitate manufacturing and delivery of the HTIR-TC as a marketed or commercial product (i.e. if the accuracy requirements are appropriate). For most applications, especially those in which new fuel designs are being tested and data for the evaluation are of critical importance, each HTIR-TC will likely be individually calibrated to obtain the most accurate data possible.

4.3 Drift

For HTIR-TCs, the drift requirement for commercial nuclear power reactors using a conservatively high neutron absorption coefficient which includes the cross sections for both Mo and Nb is less than -1% sensor drift over a typical 18-month refueling cycle, and -1.2% for a 24 month refueling cycle. If the cross section for Nb is not included in the overall HTIR-TC cross section, since Nb contributes insignificantly to the HTIR-TC EMF, the more realistic drift requirement for 24 months would also decrease to < 1.0%. This is considered low enough for most predictable HTIR-TC standalone applications in nuclear power reactors. The drift in a well-thermalized, water-cooled reactor is mostly due to transmutations caused by thermal neutron reactions with the thermoelements in TC cable regions where the temperature is changing. Drift caused by fast neutron damage to the thermoelements is small in these reactors, with such damage including atom dislocation in the thermoelement lattices as a result of neutron bombardment, resulting in increased brittleness and hardening and potentially causing changes in the TC response. However, this effect—which, due to annealing, is weaker at high temperatures than at low ones—is generally only significant in fast reactors, which cause the neutron energies of the fast neutron flux spectrum to peak at around 1 MeV. Also, since HTIR-TCs are heat treated to a temperature (i.e. 1450°C) far exceeding the typical operating power reactor (e.g. BWR/PWR) temperature of typically ~350°C, any drift due to prolonged residence at normal operating reactor temperature or accident temperatures of < 1000°C is negligible. As mentioned above, these relatively low temperatures are more than 450°C below the heat treatment temperature which has a depreciating effect on the drift.

Though the transmutation-caused change in the TC's elemental makeup can be correlated to the expected drift, such correlation depends on the properties of the thermoelements. To determine the correlation for Mo/Nb TCs, the drift data in the Mo/Nb TC literature [1, 5] was reviewed, revealing that, for Mo/Nb TCs—in regions where spatial temperature gradients are present—after a thermal neutron fluence of 1.6×10^{21} nvt and a fast neutron fluence of 2.7×10^{21} nvt, the drift was approximately 5°C at 1080°C, corresponding to a drift of 5/1080 * 100 = 0.46%. The theoretical basis for this drift measurement is unknown, and it is hoped that the HTIR-TC would have a similarly small drift. However, the HTIR-TC drift specification would need to be based on actual HTIR-TC performance over the full 125 effective full-power days (EFPDs) of exposure in the Advanced Test Reactor (ATR) qualification test, after adjusting for the differences between operation in an ATR qualification test and operation in a BWR/PWR.

To specify HTIR-TC drift in an operating, well-thermalized, light-water thermal power reactor (BWR or PWR), a conservative approach is recommended. Thus, it is appropriate to specify a drift that is conservatively high yet still acceptable for a single 18-month refueling cycle, and that can be linked to the drift caused by thermal neutrons in the qualification test. Based on preliminary evaluations of performance in the qualification test, a thermal-neutron-caused drift of less than 1% is desired for 18 months of operation in a commercial, well-thermalized, light-water reactor, for which the typical average thermal flux is 8×10^{13} nv, corresponding to a thermal neutron fluence of 3.8×10^{21} nvt. Thus, the requirement of -1% drift for HTIR-TCs in standalone commercial thermal neutron power plant applications is both acceptable and conservatively reasonable.

It is recommended that, over 125 EFPDs, the HTIR-TC drift specification in the ATR test fixture be approximately -3.5%. It is recognized that, for this application, the drift is due to thermal and fast neutrons, as well as to the length of time in the test fixture at high temperatures.

Note that this drift is a function of the materials used in the HTIR-TC design, the flux and temperature variations over the TC cable length, and the temperature at which the TC was heat treated. The measured HTIR-TC drift in the ATR test fixture is expected to be much higher than for a commercial

BWR/PWR, since the measurement temperature is much lower in a BWR/PWR. Note that drift is not the only factor determining the effective life of the TC. Unexpected excessive mechanical failures due to factors such as excessive thermal shocks—especially for fuel temperature measurement experiments—must be considered in determining a TC's effective life, and these are discussed in the mechanical ruggedness requirements outlined in Section 4.5.

4.4 Life

The required life of the TC depends on its application. For the standalone application of an HTIR-TC in an NPP, a life equal to or greater than the refueling interval is appropriate, since the TC can be replaced during the refueling interval. Therefore, an end of life, that extends to drift reaching -1% (and without opportunity to recalibrate), is also an appropriate specification for the neutronic life of HTIR-TCs in NPPs. The specification for end of life due to drift in the ATR test fixture depends on the temperature being measured. The HTIR-TC is heat treated to 1450°C, so for temperatures of ~1200°C a drift of -3.5% after 125 EFPDs is appropriate. For temperatures higher than 1200°C, the drift could exceed this specification, due primarily to prolonged residence time at high temperature.

The neutronic life requirement when the drift reaches -3.5% is also appropriate for HTIR-TC application in an ATR fuel test fixture, where the drift is due to thermal/fast neutrons and the length of residence time at high temperature. However, in fuel test applications within a test reactor, an excessive number of thermal shocks can also limit the HTIR-TC life. As described in Section 4.5, a thermal shock requirement of five rapid startups and five rapid shutdowns—each covering a temperature range of room temperature to ~1500°C—was established as an HTIR-TC life requirement. The HTIR-TC life in a fuel test experiment extends up until either the aforementioned 10 thermal shocks are reached or a drift of -3.5% occurs, whichever comes first.

Note that technical specifications for commercial power reactors state that the heat-up and cooldown rates are typically quite mild (~100°F/hr), so severe thermal shocks due to rapid startup/shutdown are not a significant concern. A requirement of <100°C/hour was specified for thermal shocks related to the standalone application of HTIR-TCs in NPPs, so TC life in an NPP is not expected to be limited by thermal shocks.

4.5 Mechanical Ruggedness

For the HTIR-TC to survive the expected duration in either a fuel temperature measurement configuration or standalone configuration, it must be mechanically rugged and able to absorb the thermal shocks expected for that particular application.

First, the TC must be ruggedly designed so that the TC junction is protected and does not incur mechanical damage from handling and installation. The requirement in this regard is that the junction be secure and well protected from external forces. Also, apart from the junction, the entire length of the TC should be protected from potential damage by ensuring that the bend radius of the entire HTIR-TC cable assembly exceeds 20 in.

Excessive thermal shock can also disable the HTIR-TC by causing changes in the thermoelement lattice structure or potentially causing mechanical failure of the electrical connections within the TC. The HTIR-TC thermal shock requirement for the fuel temperature test application is five rapid shutdowns and five rapid restarts of the reactor during the course of the experiment, with each shutdown or restart resulting in a thermal shock of room temperature to ~1500°C. The basis for this requirement is that such shutdowns/restarts could reflect the nature of the fuel test experiment, meaning that the HTIR-TCs must be designed to survive this set of thermal shocks. For standalone use in NPPs, the thermal shock requirement is 100°C per hour. The basis for this requirement is that, in NPPs, the thermal shock is never as great as in test reactors, since the normal NPP procedures found in the technical specifications require the temperature changes to be gradual (typically <100°F [~38°C] per hour) to protect the reactor structures and equipment.

Excessive drift can also be a life-limiting parameter and has been observed in other types of TCs. It was attributed to improper mechanical construction of the TC, an issue that can lead to surface changes occurring due to chemical processes (e.g., oxidation or solid-state diffusion) and permeating the surface layer down to the TC junction. The requirement for preventing such excessive drift is to rigidly control the HTIR-TC design and manufacturing process [7].

4.6 Response Time

The required HTIR-TC response time is <0.5 seconds since this is sufficiently fast for HTIR-TC applications. Moreover, a study of the literature shows that the response time for bare-wire Mo/Nb TCs is <0.2 seconds [8, 9], so increasing the time constant to 0.5 seconds to account for the effect of encasing the Mo/Nb junction within a thin protective casing is technically reasonable. However, since the response time is so heavily dependent on the thickness of the cover being used in the HTIR-TC application, and since the measurement of time constant is not included in the HTIR-TC qualification test the time constant requirement is not included in the overall HTIR performance requirements.

4.7 Summary of Performance Requirements

Table 3 summarizes the HTIR-TC requirements for both a fuel performance test application and as a standalone application for use in BWRs/PWRs and beyond (i.e. non-nuclear).

Table 3: Summary of performance requirements.

Performance Parameter	Performance Requirement Fuel Test Application	Performance Requirement Standalone Application in BWRs/PWRs
Temperature Range	Room Temp. to 1500°C	Room Temp. to 1500°C
Accuracy	Not Specified	±1%
Drift	3.5% for 125 EFPDs	<1% for 18 months, corresponding to a typical NPP refueling cycle using conservatively high neutron absorption cross section; and <1% for 24 months using a lower and more realistic neutron absorption cross section.
Life	-3.5% drift, or 10 thermal shocks (room temp. to 1500°C)	18 or 24 months, corresponding to an applicable NPP refueling cycle
Mechanical Ruggedness:		
Rugged Junction	Rugged mechanical junction design	Rugged mechanical junction design
Bend Radius	Minimum of 2 ft	Minimum of 2 ft
Thermal Shock	5 sudden startups and 5 sudden shutdowns, each causing a thermal shock of room temp. to 1500°C	<100°C/hr

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