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J.L. Rempe
D.L. Knudson
K.G. Condie
S.C. Wilkins

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Evaluation of Specialized Thermocouples for High-Temperature In-Pile Testing

J. L. Rempe,* D. L. Knudson, and K. G. Condie
Idaho National Laboratory
P.O. Box 1625, MS 3840, Idaho Falls, ID 83415
Tel:208-526-2897 , Fax:208-526-2930 , Email:Joy.Rempe@inl.gov

S. Curtis Wilkins
Consultant
1258 Cameron Avenue, Idaho Falls, ID 83402
Tel: 208-523-9202, Email: curtwilkins@cableone.net

Abstract – *Many advanced nuclear reactor designs require new fuel, cladding, and structural materials. Data are needed to characterize the performance of these new materials in high temperature, oxidizing, and radiation conditions. To obtain this data, robust instrumentation is needed that can survive proposed test conditions. Standard thermocuoples for measuring temperature in-pile degrade at temperatures above 1100 ºC. Hence, INL initiated a project to develop specialized thermocouples for high temperature in-pile applications. Results from efforts to develop, fabricate, and evaluate specialized high-temperature thermocouples for in-pile applications suggest that several material combinations are viable. Tests show that several low neutron cross-section candidate materials are resistant to material interactions and remain ductile at high temperatures. In addition, results indicate that the thermoelectric response is single-valued and repeatable with acceptable resolution for the candidate thermoelements considered. The final selection of the thermocouple materials will depend on the desired peak temperature and accuracy requirements. If thermocouples are needed that measure temperatures at 1600 ºC or higher, the doped Mo / Nb-1%Zr and Mo-1.6% Nb / Nb-1%Zr thermoelement wire combinations are recommended with* HfO_2 *insulation, and a Nb-1%Zr sheath.*

Additional evaluations are underway to characterize the performance of this proposed thermocouple design. INL has worked to optimize this thermocouple's stability. With appropriate heat treatment and fabrication approaches, results indicate that the effects of thermal cycling on the calibration of the proposed thermocouple design can be minimized. INL has initiated a series of high temperature (from 1200 to 1800 ºC) long duration (up to six months) tests. Initial results indicate the INL-developed thermocouple's thermoelectric response is stable with less than 15 ºC drift observed in over 3500 hours of the planned 4000 hours of tests at 1200 ºC. In comparison, commercially-available Type K and N thermocouples included in these 1200 ºC tests have experienced drifts up to of over100 ºC.

I. INTRODUCTION

To resolve principal technical and scientific obstacles to the long-term future use of nuclear energy, new reactor designs must offer enhanced safety and overcome issues involving resistance to proliferation, economics, and nuclear waste disposition. To meet these goals, new materials are being considered for fuel, cladding, and structures in advanced and existing nuclear reactors. Data are needed to characterize the performance of these new materials in high temperature, oxidizing, and radiation conditions. To obtain this data, robust instrumentation is needed that can survive proposed test conditions. Base-metal thermocouples for measuring temperature in-pile degrade at temperatures above 1100 ºC. Hence, INL initiated a project to develop specialized thermocouples for high temperature in-pile applications. Initial efforts to select materials for these specialized thermocouples are reported in Rempe.[1] This paper reports results from additional evaluations of these specialized thermocouples.

II. BACKGROUND

Types of instrumentation that might be employed for in-pile, high temperature applications were first evaluated.[1] For temperatures above 1100 ºC, previously investigated in-pile instrumentation methods are limited to thermocouples (with thermoelements consisting of molybdenum, niobium, zirconium, or their alloys), Johnson Noise Power Thermometers (JNPT), and ultrasonic thermometers (UTs). UT and JNPT techniques weren't considered because of the cost and complexity associated with their probe and signal processing equipment. Optical pyrometer techniques were eliminated because viewing ports are not typically available. Likewise, considerably more development is needed before optical fiber methods overcome difficulties associated with signal degradation in radiation fields. Hence, specialized thermocouples were deemed to be the simplest and most economic approach for in-pile high temperature measurements. However, such thermocouples have not been used for over a decade,[2-6] and improved versions were needed.

III. APPROACH

Table I lists commercially-available materials initially considered for thermocouple components based on their high temperature thermal properties, nuclear properties, and cost.[1] Melting temperatures for these materials range from 1670 (titanium) to 2800 °C (hafnia). Thermal neutron absorption cross sections of the candidate thermoelement wire materials in Table I are below 10 barns. For comparison, the thermal neutron absorption cross sections of rhenium and rhodium are 86 and 150 barns, respectively. Hafnia has a thermal neutron absorption cross section of 100 barns. However, insulator choice doesn't affect in-pile thermocouple performance because the amount of insulation in a thermocouple is sufficiently small that it doesn't significantly disturb neutron flux.

TABLE I

Candidate Thermocouple Component Materials

Component	Candidate Materials
Thermoelements	Molybdenum,* Zircaloy-4, Titanium-45% Niobium, Niobium-1%Zirconium
Insulators	Aluminum Oxide, Hafnium Oxide, Magnesium Oxide
Sheaths	Titanium, Zircaloy-4, Niobium-1%Zirconium

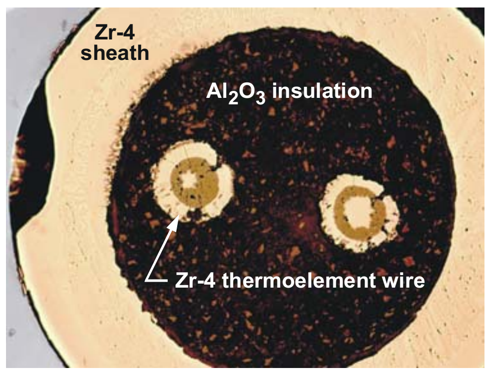
* Evaluations considered several types of Molybednum: undoped Mo, Mo-1.6% Nb, KW-Mo (doped with Tungsten, Silicon and Potassium), and ODS-Mo (containing Lanthanum Oxide).

Several types of evaluations were completed for candidate thermocouple materials at INL's High Temperature Test Laboratory (HTTL), which has key equipment and trained staff for such activities. Using representative samples, tests evaluated the potential for materials interactions between candidate insulation, sheath, and thermoelement materials. Then, candidate thermoelement wires were exposed to high temperatures to assess their ductility. Last, calibration tests were performed.

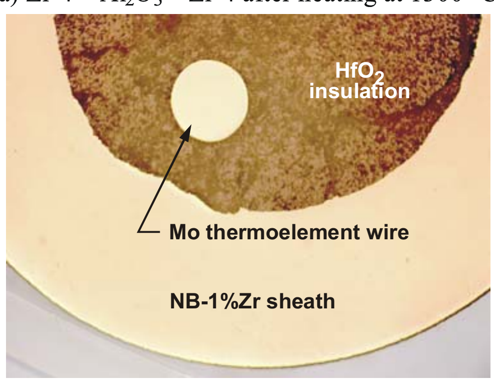
IV. THERMOCOUPLE COMPONENT SELECTION

IV.A. Materials Interaction Tests

Materials interaction tests were completed by heating representative thermocouple samples in gettered argon at 1300 and 1600 °C. As shown in Figure 1(a), 1300 °C tests indicated significant materials interactions occurred with samples containing Zr-4 thermoelements, Al_2O_3 insulators, and Zr-4 sheaths. However, 1600 °C results for Nb-1%Zr and Mo thermoelement wires and Nb-1%Zr sheaths indicate that no discernible materials interactions occurred between these materials and HfO_2 insulators (see Figure 1(b)). In summary, tests indicate that several thermoelement wire materials (Mo, Nb-1%Zr, and Ti-45% Nb) appear viable with HfO_2 insulation and Nb-1% Zr sheaths. Other sheath and insulator materials may also be viable if temperatures remain below 1300 °C.



(a) Zr-4 – Al_2O_3 – Zr-4 after heating at 1300 ºC



(b) Mo – HfO_2 – Nb-1%Zr after heating at 1600 ºC

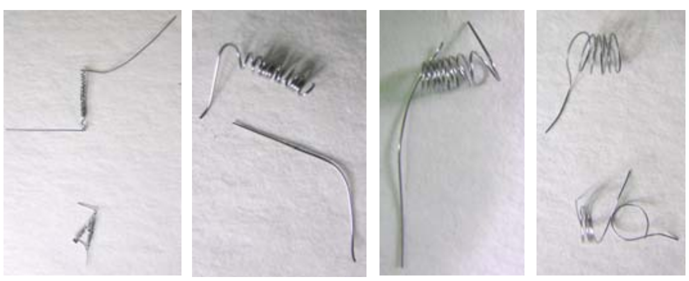
Fig. 1. Materials interaction test results (wire-insulator-sheath) using representative thermocouple samples.

IV.B. Ductility

Mandrel-wrap tests on wires exposed to temperatures up to 1600°C provided insights about thermoelement embrittlement. Wire samples from each of the thermoelement materials listed in Table 1 were wrapped on mandrels of two, five, ten, and twenty times the wire diameter. Those metals that wrap without damage on a small-diameter mandrel after high-temperature exposure are better candidates from the standpoint of embrittlement. Most Table 1 thermocouple wire materials exhibited suitable ductility. The one exception, undoped Mo wire, recrystallizes at 1200 °C. As illustrated in Figure 2(a), this wire was brittle after heating at 1300 °C. However, other tested Mo wires (e.g., KW-Mo, ODS-Mo, and Mo-1.6%Nb) remained ductile even after heating at 1600 ºC (see Figure 2(b)).

(a) 0.51 mm (b) 1.27 mm (c) 2.54 mm (d) 5.08 mm

(a) Undoped Mo wire after heating at 1300 °C.



(a) 0.51 mm (top unheated) (b) 1.27 mm (c) 2.54 mm (d) 5.08 mm (top unheated)

(b) Doped Mo-KW wire after heating at 1600 °C

Fig. 2. Ductility test results (0. 254 mm diameter wire).

IV.C. Thermoelectric Calibration

Calibration tests were completed for candidate thermocouple combinations. Tests were performed using "bare wire" thermoelement wire combinations that were inserted into hard-fired insulator material. Results (see Figure 3) indicate that the thermoelectric response is single-valued and repeatable for the candidate thermoelements considered. In addition, results indicate that the high temperature resolution is acceptable for all thermocouple element combinations considered (although some combinations are limited due to materials interactions at temperatures below 1600 ºC).

IV.D. Summary

Results indicate several candidate low neutron cross-section thermocouple component materials experience minimal interactions and remain ductile at high temperatures. Tests also indicate that the thermoelectric response for candidate thermoelement combinations is single-valued and repeatable with acceptable resolution. The selection of thermocouple materials will depend on the desired peak temperature and accuracy requirements. If thermocouples are needed that measure temperatures at 1600 ºC or higher, the doped Mo / Nb-1%Zr and Mo-1.6% Nb / Nb-1%Zr combinations are recommended. However, additional evaluations are needed to characterize the performance of this proposed thermocouple design.

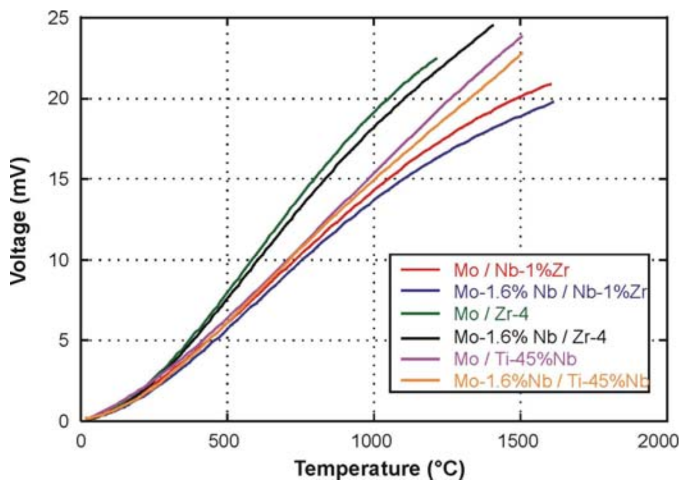


Fig. 3. Calibration curves for candidate thermocouples.

V. THERMOCOUPLE EVALUATIONS

Additional evaluations are underway to characterize the performance of the proposed thermocouple design. Tests were completed to assess the effect of thermal cycling, heat treatment, and selected fabrication processes. In addition, a series of high temperature tests were initiated to evaluate the long-term performance of these thermocouples. As indicated below, preliminary results suggest that the proposed design performs better than commercially available thermocouples.

V.A. Thermal Cycling Tests

Figure 4 compares data from "As-Received" bare wire (ARBW) and swaged (ARS) KW-Mo and Nb1%Zr thermocouples. For each thermocouple, data were obtained from several thermal cycles. Data are single-valued and consistent except for tests conducted with swaged thermocouples above 1400 °C. Above this temperature, more variation in the emf occurs. It is believed that this may be due to annealing of the cold-work introduced into the thermocouples by swaging and the onset of grain growth. Note that all thermocouples were fabricated with wire from the same lot.

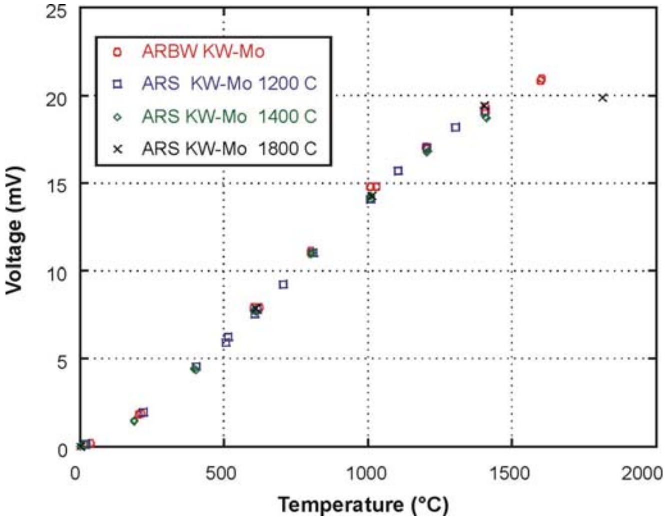


Fig. 4. Combined calibration data from tested ARBW and ARS KW-Mo/Nb1%Zr thermocouples.

V.B. Heat Treatment Temperature

Figure 5 compares the drift measured in as-received swaged KW-Mo/Nb1%Zr thermocouples heated at 1200 and 1400 °C. Note that none of these thermocouples were heat treated prior to testing. The percentage drift for the "ARS KW-Mo 1200" thermocouple was nearly an order of magnitude higher than that measured for the "ARS KW-Mo 1400" thermocouple. Although data were only obtained for approximately 60 hours, the output of the ARS KW-Mo 1400" thermocouple appears to have stabilized after approximately 20 hours after heating at 1400 °C. In comparison, the output of "ARS KW-Mo 1200" indicates that this thermocouple has not stabilized after even 140 hours of testing at 1200 °C. Hence, data suggests that at least 20 hours of heat treatment at 1400 °C is needed for these thermocouples (shorter time periods may be used if heat treatments are performed at higher temperatures).

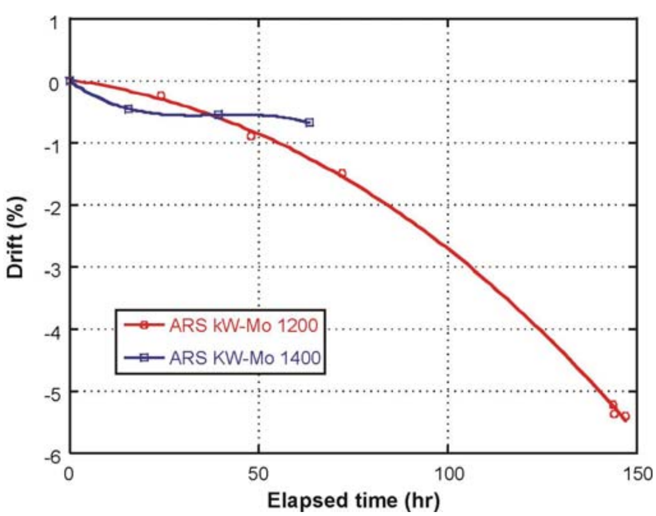
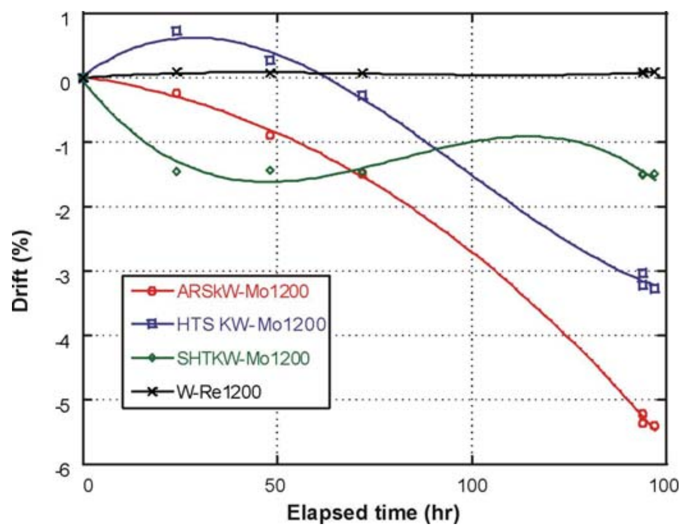


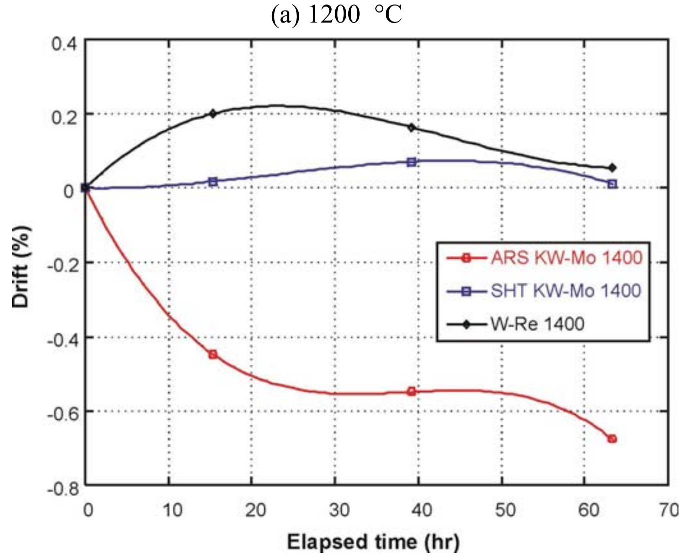
Fig. 5. Drift (in percent) observed for ARS thermocouples heated at 1200 and 1400 °C.

V.C. Fabrication Effects

It is possible to heat treat the wires prior to swaging and after swaging. To gain insights about the effects of various fabrication options, calibration and drift tests were completed at high temperatures for thermocouples fabricated from as-received bare wires (ARBW), from as-received wire that was then swaged (ARS), for heat treated wires that were subsequently swaged (HTS), and for as-received bare wires that were swaged and then heat treated (SHT). Figures 6(a) and (b) compare the drift (in percent) measured for the 1200 and 1400 °C tests. Note that heat treatments for thermocouples tested at 1200 °C were performed at 1500 °C for 30 minutes and those for 1400 °C tests completed at 1600 °C for 4 hours. A reference Type C thermocouple (W-Re 1200 or W-Re 1400) was included in each test.



(a) 1200 °C



(b) 1400 °C

Fig. 6. Drift (in percent) observed for KW-Mo/Nb1Zr thermocouples.

Data from the test conducted at 1200 °C indicate that the ARS and HTS thermocouples had not stabilized during the period that output was monitored. In contrast, the SHT thermocouple appears to have stabilized within 50 hours. Data from the test conducted at 1400 °C indicate that the ARS thermocouple stabilized within 24 hours. However, the SHT KW-Mo 1400 thermocouple output appears to be stable throughout the test. The measured drift remained below 0.1%. which is less than that measured for the Type C W-Re 1400 reference thermocouple.

V.D. Long Duration Testing

Longer-duration evaluations were initiated in which proposed INL doped Mo/Nb1%Zr thermocouples and commercially available thermocouples are held at elevated temperatures (ranging from 1200 to 1800 °C) for up to 6 months. The 1200°C tests use the setup shown in Figure 7. Thermocouples are inserted into a tube furnace containing gettered argon. In these tests, data are continuously recorded and stored on a computer.

Fig. 7. Long duration 1200 ºC thermocouple test setup.

The measured emf from representative thermocouples in the 1200°C test is compared in Figure 8. This 1200 °C test includes nineteen commercially-available Type N thermocouples, three commercially-available Type K thermocouples, and nine of the INL-developed doped Mo/Nb1% Zr thermocouples. Several transients are included in this test (to assess the effect that transients have on the thermocouples' response). As indicated in Figure 8, the measured emf for some thermocouples (e.g., see the curve labeled "P2K1-Type K", which corresponds to one of the Type K thermocouples included in this test) is starting to drift downward. It's interesting to note that this downward drift isn't influenced by the transients.

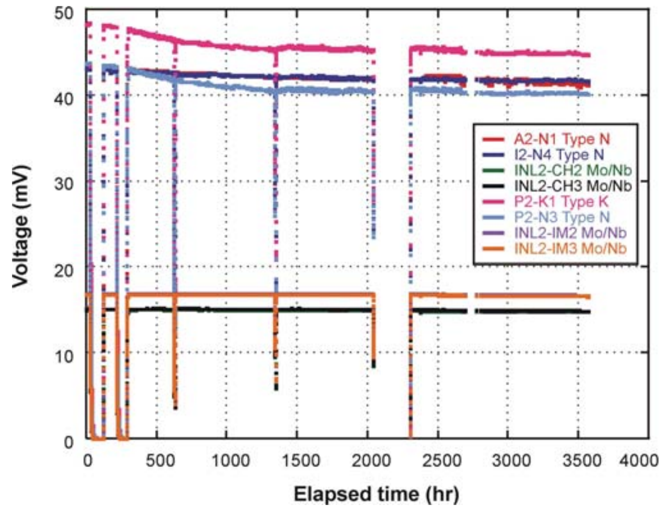


Fig. 8. Measured emf voltage for representative thermocouples in 1200 °C tests.

Figure 9 plots temperatures for each of the TCs shown in Figure 8 (with the transients omitted to better reflect the observed trends). In over 3500 hours of this planned 4000 hour test, some of the Type K and N thermocouples (e.g., see the curves labeled, "P2K1 Type K" and "P2N2 Type N") indicate that these thermocouples have drifted over 100 °C. Much smaller drifts (up to 15 °C) are observed in INL-developed thermocouples with hafnia and magnesia insulation (see the curves labeled "INL CH2 Mo/Nb" and "INL CH3 Mo/Nb," which correspond to two doped Mo-Nb1%Zr thermocouples with Nb-1%Zr sheaths and HO_2 insulation and the curves labeled "INL2-IM2 Mo/Nb" and "INL2-IM3 Mo/Nb," which correspond to two Mo-Nb1%Zr thermocouples with Inconel 600 sheaths and MgO insulation. These results led several customers to request doped Mo-Nb1%Zr thermocouples in proposed irradiation tests at INL's Advanced Test Reactor.

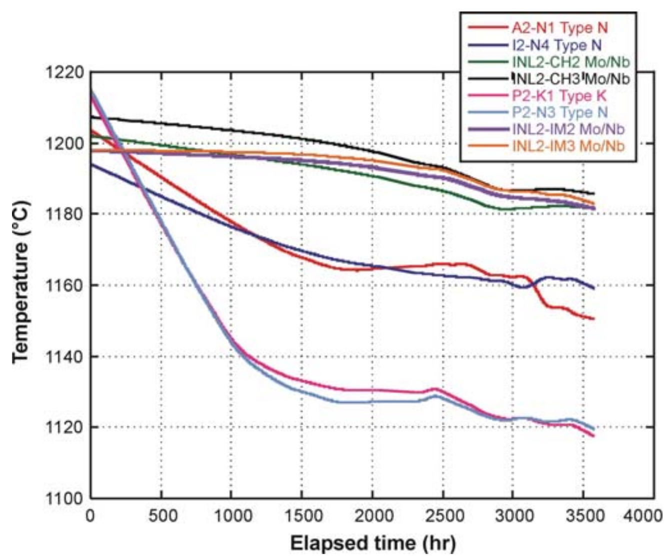


Fig. 9. Temperatures for representative thermocouples in 1200 °C tests.

VI. CONCLUSIONS

Results from efforts to develop, fabricate, and evaluate specialized high-temperature thermocouples for in-pile applications indicate that several material combinations are viable. Tests show that several candidate low neutron cross-section thermocouple component materials experience minimal interactions and remain ductile at high temperatures. In addition, results indicate that the thermoelectric response for the candidate thermoelements considered is single-valued and repeatable with acceptable resolution. The selection of the thermocouple materials will depend on the desired peak temperature and accuracy requirements. If thermocouples are needed that measure temperatures at 1600 ºC or higher, the doped Mo / Nb-1%Zr and Mo-1.6% Nb / Nb-1%Zr combinations are recommended with HfO_2 insulation and a Nb-1%Zr sheath.

Additional evaluations are underway to characterize the performance of this proposed thermocouple design. INL has worked to optimize this thermocouple's stability. With appropriate heat treatment and fabrication approaches, results indicate that the effects of thermal cycling on the calibration of the proposed thermocouple design can be minimized. INL initiated a series of high temperature (from 1200 to 1800 ºC) long duration (up to six months) tests. Initial results indicate that the INL-developed thermocouple's thermoelectric response is stable, with less than 15 ºC drift observed during the first 3500 hours of the planned 4000 hours of tests at 1200 ºC. In comparison, commercially-available Type K and N thermocouples included in these 1200 ºC tests have experienced drifts of over 100 ºC.

These preliminary results have motivated several customers to request that doped Mo-Nb1%Zr thermocouples be included in upcoming irradiation tests at INL's Advanced Test Reactor. Activities are underway to fabricate specialized thermocouples for these irradiation tests. In addition, INL thermocouple development activities will continue to explore additional options that could further enhance the performance of these thermocouples for higher temperature applications. In particular, INL will be exploring the effects of additional fabrication options, geometries, and non-commercial component materials on thermocouple performance.

ACKNOWLEDGEMENTS

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