# High Temperature Irradiation Resistant Thermocouples -- A Low Cost Sensor for In-Pile Testing at High Temperatures

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# High Temperature Irradiation Resistant Thermocouples – A Low Cost Sensor for In-Pile Testing at High Temperatures

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Abstract – Several options have been identified to further improve recently-developed Idaho National Laboratory (INL) High Temperature Irradiation Resistant Thermocouples (HTIR-TCs) for in-pile testing. These options have the potential to reduce fabrication costs and allow HTIR-TC use in higher temperature applications (up to at least 1800 °C). The INL and the University of Idaho (UI) investigated these options with the ultimate objective of providing recommendations for alternate thermocouple designs that are optimized for various applications. This paper summarizes results from these INL/UI investigations. Specifically, results are reported regarding options to enhance HTIR-TC performance (such as the use of specially formulated alloys, improved heat treatments, and the adoption of alternate geometries) and options to reduce HTIR-TC costs (such as automated fabrication techniques and the use of copper/nickel alloys as extension cable).

### I. INTRODUCTION

New fuel, cladding, and structural materials offer the potential for safer and more economic generation of energy from existing nuclear reactor and advanced reactor designs. However, insufficient data are available to characterize these materials in high temperature, radiation conditions. To evaluate candidate material performance, robust instrumentation is needed to measure temperatures under these conditions. Unfortunately, traditional thermocouples (TCs) either drift due to degradation at high temperatures (above 1100 °C) or drift due to transmutation of the TC components. Consequently, TCs are needed that can withstand both high temperature and high radiation environments.

To address this need, the Idaho National Laboratory (INL) developed and evaluated the performance of a High Temperature Irradiation-Resistant Thermocouple (HTIR-TC). Candidate TC components were first identified based on their ability to withstand high temperature and radiation. Components were then selected based on data obtained from materials interaction tests, ductility investigations, and temperature resolution evaluations relative to their electrical potential. The resulting INL HTIR-TC, which is designed to use commercially-available thermoelements primarily consisting

of molybdenum and niobium, has been shown to be stable and reliable based on minimal signal drift (typically less than 2%) throughout long duration (over 4000 hr) tests at high temperatures (up to 1400 °C) and thermal cycling tests. Furthermore, irradiation tests in INL's Advanced Test Reactor (ATR) are underway demonstrating the in-pile performance of these TCs.

Several options have been identified that could further improve the INL-developed HTIR-TCs for in-pile testing. These options have the potential to reduce fabrication costs and allow HTIR-TC use in higher temperature applications (up to at least 1800 °C). A joint INL and University of Idaho (UI) effort investigated these options with the ultimate objective of providing recommendations for an enhanced design. Following a brief description of the development and current status of the HTIR-TC, results from these INL/UI investigations will be summarized. Specifically, results are reported regarding options to enhance performance and options to reduce fabrication costs.

### II. BACKGROUND

The HTIR-TC was originally envisioned to be a swaged-type TC. Development of this TC began with identification of commercially available and potentially

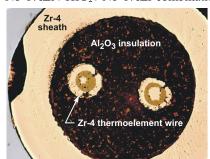
suitable materials. Relatively low cost materials with favorable high temperature and nuclear properties were deemed suitable. Materials initially considered are listed in Table I.

TABLE I Candidate TC Component Materials

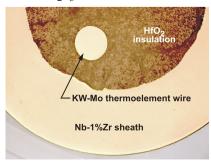
Component	Candidate Materials		
Thermoelements	Molybdenum,* Niobium-1%Zirconium,		
	Titanium-45%Niobium, Zircaloy-4		
Insulators	Aluminum Oxide, Hafnium Oxide,		
	Magnesium Oxide		
Sheaths**	Niobium-1%Zirconium, Titanium,		
	Zircaloy-4		

- \* Pure Mo, KW-Mo (doped with tungsten, potassium, and silicon), ODS-Mo (containing lanthanum oxide), and Mo-1.6%Nb were considered.
- \*\* Only materials amenable to swaging were initially considered.

Interaction tests were the first evaluations of the candidate materials. These tests were completed by heating representative samples in gettered argon at 1300 °C and 1600 °C for 30 minutes. As shown in Fig. 1(a), 1300 °C tests indicated significant materials interactions occurred with samples containing Zr-4 thermoelements, Al<sub>2</sub>O<sub>3</sub> insulators, and Zr-4 sheaths. However, 1600 °C results for Nb-1%Zr and KW-Mo thermoelements and Nb-1%Zr sheaths indicated that no discernible materials interactions occurred between these materials and HfO<sub>2</sub> insulators (see Fig. 1(b)). These results, and results from a series of similar interaction tests, provided one measure of suitability of the KW-Mo - Nb-1%Zr / HfO<sub>2</sub> / Nb-1%Zr combination.



(a) Zr-4 - Al<sub>2</sub>O<sub>3</sub> - Zr-4 after heating at 1300 °C



(b) KW-Mo –  $HfO_2$  – Nb-1%Zr after heating at 1600 °C

Fig. 1. Materials (thermoelement/insulator/sheath) interaction test results.

Mandrel-wrap tests on wires exposed to temperatures up to 1600 °C for 30 minutes provided insights about thermoelement embrittlement. Wire samples from each of the thermoelements listed in Table I were heated and then wrapped on mandrels of two, five, ten, and twenty times the wire diameter. Those that wrapped without damage on a small-diameter mandrel after high-temperature exposure are better candidates from the standpoint of embrittlement. Most Table I thermoelements exhibited suitable ductility. The one exception, pure Mo wire, recrystallized at 1200 °C. As illustrated in Fig. 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 °C (see Fig. 2(b)).

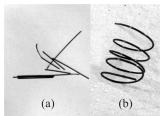


Fig. 2. Ductility test results [(a) Pure Mo wire after heating at 1300 °C, (b) KW-Mo wire after heating at 1600 °C].

Thermoelectric evaluations were also completed for candidate TC combinations at selected temperatures up to 1600 °C. The thermoelectric response, as shown in Fig. 3, was found to be single-valued and repeatable for all candidate combinations considered. In addition, results indicate that the high temperature resolution is acceptable for all combinations (although some of the candidates have limits associated with materials interactions at temperatures below 1600 °C).

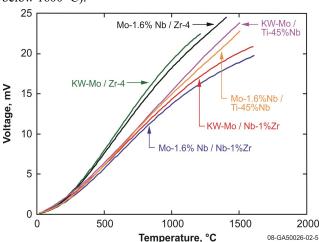


Fig. 3. Thermoelectric response for candidate TCs.

Evaluation of materials interaction, ductility, and thermoelectric test results indicate there may be several acceptable TC combinations among materials that were tested. However, for the high temperature in-pile applications envisioned for HTIR-TCs, a swaged design containing doped Mo / Nb-1%Zr thermoelements with

hafnia insulation and a Nb-1%Zr sheath was ultimately selected.

To demonstrate the long duration performance of HTIR-TCs, INL is conducting tests where the TCs are held at elevated temperatures (from 1200 to 1800 °C) for up to 6 months. The setup used for these tests is shown in Fig. 4. TCs are immersed in a continuous flow of gettered argon inside alumina tubes that are loaded in a tube furnace. A National Institute of Standards and Technology-traceable Type S TC is used for reference temperature measurements. Test data are automatically recorded at frequent intervals and stored electronically.

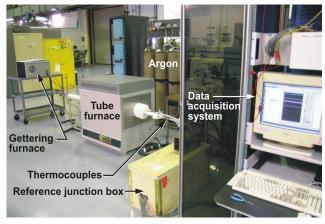


Fig. 4. Long duration TC test setup.

The 1200 °C test included nineteen commercially-available Type N TCs, three commercially-available Type K TCs, and nine INL HTIR-TCs. As indicated in Fig. 5, some of the Type K and N TCs drifted by over 100 °C or 8%. Much smaller drifts (typically less than 20 °C or 2%) were observed in the INL HTIR-TCs. Similar drift was observed in HTIR-TCs in a long duration (4000 hr) test completed at  $1400\ ^{\circ}\text{C}.^{1}$ 

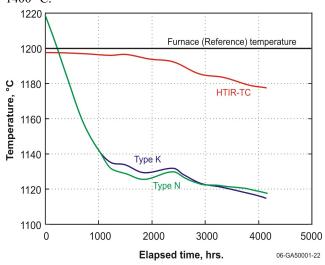


Fig. 5. Representative TC response in the 1200 °C test.

HTIR-TCs have also been installed in a multi-capsule experiment that is currently being irradiated in INL's ATR at temperatures up to 1200 °C. The irradiation started in February 2007, and it is planned to continue for over two years. Signals from two INL HTIR-TCs and a Type N TC (which is located in a cooler region within one of the capsules) are illustrated in Fig. 6. As shown, HTIR-TC signals are higher but otherwise consistent with the signal from the Type N TC. (Data fluctuations are due to ATR power fluctuations.) These results provide an important indication of the irradiation resistance of HTIR-TCs.

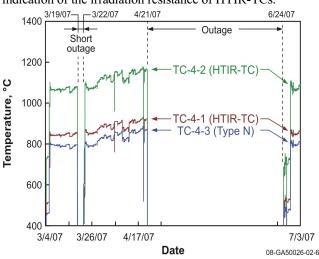


Fig. 6. Representative HTIR-TC and Type N data during ATR irradiation.

As discussed, recent INL efforts led to development of the HTIR-TC. Testing to date has indicated the stability and reliability of this instrument. However, INL/UI are also investigating options to further enhance HTIR-TC performance while reducing costs as outlined below.

### III. PERFORMANCE ENHANCEMENT OPTIONS

Efforts to enhance HTIR-TC performance have focused on the use of thermoelements made from specially formulated alloys, investigations to improve and optimize heat treatments, and the evaluation of alternate TC geometries.

### III.A. Specially Formulated Alloys

Historically, industry has relied on alloys (e.g., W/Re alloys and Pt/Rh alloys), rather than pure metals, to improve TC high temperature performance with respect to ductility, stability, and reliability.<sup>2</sup> Prior experience with Mo/Nb TCs suggests that similar efforts are warranted.<sup>3-5</sup>

Prokoshkin and Vasil 'eva indicate that addition of small amounts (less than 1%) of zirconium to niobium raises its recrystallization temperature by 25 °C.<sup>6</sup> The addition of molybdenum (up to 4%) may delay recrystallization by 75 °C for temperatures up to 1200 °C. Schley and Metauer

have shown that addition of small amounts of molybdenum (less than 10%) to niobium will improve its resolution.<sup>7</sup>

Efforts have also been made to improve the ductility and resolution of molybdenum. Prokoshkin and Vasil 'eva indicate that the recrystallization temperature of molybdenum is increased if it is alloyed with small amounts of niobium.<sup>6</sup> To control molybdenum crystal structure during recrystallization, suitable "dopants" can be added to molybdenum. In the case of molybdenum, the dopant is typically tungsten and potassium silicate. In more recent years, lanthanum oxide has been used as a dopant for molybdenum. Furthermore, investigations by Schley and Metauer suggest that the addition of small amounts of niobium (less than 5% to molybdenum) will improve its thermoelectric properties.<sup>7</sup>

Results from the earlier efforts prompted further evaluation into the possible benefits of using alternate alloys. Specific alloys of molybdenum and niobium evaluated here are listed in Table II. Two types of doped molybdenum, two alloys of molybdenum with small amounts of niobium, three alloys of niobium with small amounts of molybdenum, and one alloy with a small amount of zirconium were investigated. Note that none of the developmental alloys contained doped molybdenum, which may improve their performance.

TABLE II Molybdenum and Niobium Alloys Evaluated

Designator		Description	
	KW-Mo	Molybdenum doped with W, K, and Si	
+	ODS-Mo	Molybdenum doped with LaO	
wire	Mo-1.6% Nb	Molybdenum-1.6% Niobium alloy	
	Mo-3% Nb	Molybdenum-3% Niobium alloy	
	Nb-1%Zr	Niobium-1% Zirconium alloy	
-	Nb-4%Mo	Niobium-4% Molybdenum alloy	
wire	Nb-6%Mo	Niobium-6% Molybdenum alloy	
	Nb-8%Mo	Niobium-8% Molybdenum alloy	

Ductility testing was completed after wire samples were heated to 1400 °C, 1600 °C, and 1800 °C. Results indicate that ODS-Mo and KW-Mo samples retain suitable ductility (e.g., the wire samples survive mandrel wrapping) for all tested temperatures and heating durations. As shown in Fig. 7, the Mo-1.6%Nb and the Mo-3%Nb samples became brittle after 12 hr at 1800 °C, but the doped molybdenum samples remained ductile. However, test results indicate that niobium wires were generally less ductile than the doped molybdenum samples tested. Only Nb-1%Zr wires were ductile after heating at 1600 °C for 2 hr (see Fig. 8).

Thermoelectric comparisons were made using TCs where KW-Mo was paired with every Nb alloy and TCs where Nb-1%Zr was paired with every Mo-based material. Test results, along with ductility results, indicated that Nb-1%Zr was superior to every Nb alloy tested.

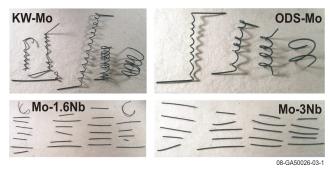


Fig. 7. Doped molybdenum and molybdenum alloy samples heated for 12 hr at 1800 °C.

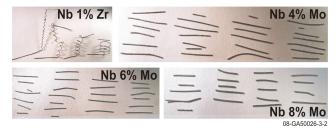


Fig. 8. Niobium alloy samples heated for 2 hr at 1600  $^{\circ}$ C.

A comparison of the thermoelectric response of Nb-1%Zr paired with all molybdenum-based wires is provided in Fig. 9. As indicated, all combinations exhibit suitable resolution, although the combination with ODS-Mo was slightly superior.

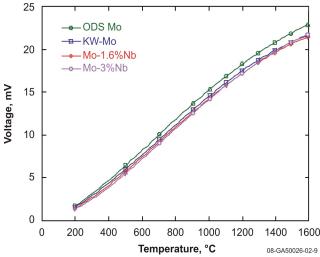


Fig. 9. Thermoelectric response of TCs containing various molybdenum-based materials paired with Nb-1%Zr.

These evaluations indicate that doped molybdenum (either ODS-Mo or KW-Mo) and Nb-1%Zr retain ductility better than the molybdenum/low niobium alloys and the niobium/low molybdenum alloys that were evaluated. Although thermoelectric resolution of the ODS-Mo / Nb-1%Zr combination is slightly superior, the lower cost of commercially available KW-Mo makes the KW-Mo / Nb-1%Zr the best combination at this time.

### III.B. Improved Heat Treatments

Grain growth in thermoelements can result in signal degradation and drift. For that reason, all HTIR-TCs are heat treated in an attempt to stabilize grain growth. However, data are limited relative to selecting appropriate temperatures and durations for this heat treatment. For that reason, heat treatment temperatures and durations listed in Table III were investigated for TCs to be operated at two different temperatures. Evaluations completed relative to those heat treatment variations are also listed.

TABLE III Heat Treatments Investigated

Op Temp (°C)	Heat Treat Temp (°C)	Heat Treat Duration (hr)	Designator	Evaluations Completed
		0	12-1300-0	Dagmanga ta
		5	12-1300-5	Response to 1200 °C, 100 hr
	1300	10	12-1300-10	@ 1200 °C
		20	12-1300-20	(ii) 1200 C
1200	1400	20	12-1400-20	Response to 1200 °C
	1500	20	12-1500-20	Response to 1200 °C
		0	15-1600-0	Dagmanga ta
		4	15-1600-4	Response to 1500 °C, 100 hr
	1600	8	15-1600-8	@ 1500 °C
1500		16	15-1600-16	W 1300 C
1300	1700	4	15-1700-4	Response to 1500 °C

Results from these evaluations indicate there is some decrease in thermoelectric response as heat treatment temperature increases. However, results for TCs heat treated at  $1300\,^{\circ}\text{C}$  suggest that the response is not significantly impacted by differences in heat treatment duration as shown in Fig. 10.

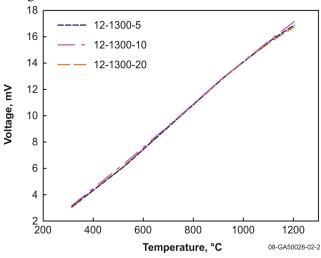


Fig. 10. Thermoelectric response of TCs heat treated at 1300 °C for 5, 10, and 20 hr (labeled 12-1300-5, 12-1300-10, and 12-1300-20, respectively).

Signal drift in TCs heat treated at 1300 °C for various durations is compared in Fig. 11. These results indicate that the highest drift occurs without heat treatment (see the curve labeled 12-1300-0). However, the observed drift was minimal, and essentially comparable, in all of the heat treated TCs. Hence, investigations suggest that heat treatment temperature may be more important than heat treatment duration in minimizing signal drift.

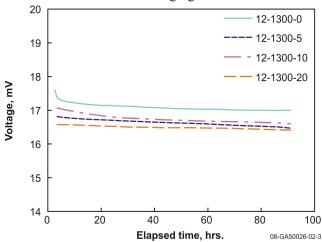


Fig. 11. Drift during a  $1200\,^{\circ}\mathrm{C}$  constant temperature test of TCs heat treated at  $1300\,^{\circ}\mathrm{C}$  for 0, 5, 10, and 20 hr (labeled 12-1300-0, 12-1300-5, 12-1300-10, and 12-1300-20, respectively).

The thermoelectric response of TCs relative to heat treatments evaluated for operation at 1500 °C is shown in Fig. 12. Similar to Fig. 10, these results also indicate that thermoelectric response is not significantly affected by heat treatment duration. Furthermore, heating at 1700 °C has no apparent benefit relative to heat treating at 1600 °C. Subsequent evaluations also indicate temperature is more effective than duration when heat treating to reduce drift.

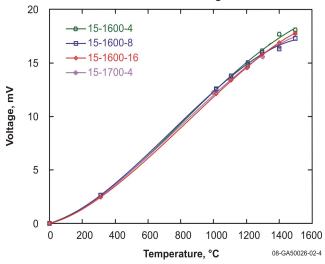


Fig. 12. Thermoelectric response of TCs heat treated for 4 hr at 1600 °C (labeled 15-1600-4, 15-1600-8, and 15-1600-16, respectively) and for 4 hr at 1700 °C (labeled 15-1700-4).

### III.C. Alternate Geometries

Alternate geometries that have been considered include evaluations of various thermoelement (wire) diameters and comparisons of swaged versus loose assembly TC configurations. As discussed below, both alternates may offer benefits relative to HTIR-TC stability and reliability.

Initial INL efforts focused on the development of swaged TCs using 0.254 mm diameter thermoelement wires. However, evaluations by Ludtka, et al., indicate the reliability of Type K and Type N TCs increases with wire diameter, especially at higher temperatures. Based on these investigations, TCs were fabricated with thermoelements of three different diameters: 0.127 mm, 0.254 mm, and 0.508 mm.

Thermoelectric results during a long duration (1000 hr) test at 1500 °C are shown in Fig. 13. As indicated, all three previously-specified thermoelement (wire) diameters were included in this test. The results illustrate there may be some stability advantage to using larger diameter thermoelement wire.

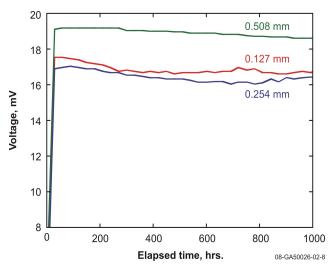
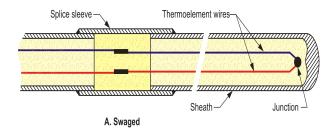


Fig. 13. Impact of thermoelement diameter on HTIR-TC stability during a long duration 1500 °C test.

As previously discussed, initial HTIR-TC development and evaluation efforts focused on a swaged design. A swaged TC is fabricated by loading pre-formed, crushable insulator beads onto thermoelement wires and placing the insulated thermoelements in a sheath (tube) that is then swaged (compacted) to form a single, cohesive component (see Figure 14a). However, a loose assembly configuration was also considered because it may have the potential for inpile measurements at temperatures above those possible with a swaged design. In a loose assembly, pre-formed, hard-fired insulator beads are used; and there is no swaging. Instead, a loose fit is left between thermoelements, insulators, and the sheath (see Figure 14b).



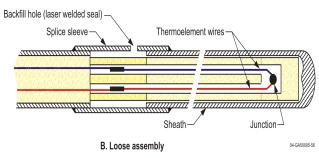


Fig. 14. Swaged and loose assembly TC configurations.

Higher temperatures may be possible using a loose assembly because thermoelement thinning and irregular deformation associated with swaging are avoided. Furthermore, the loose fit can accommodate some differential thermal expansion of TC components without inducing thermoelement stress. However, some specialized components and fabrication techniques required development, including the splice between loose assembly components and extension cable as shown in Fig. 15.

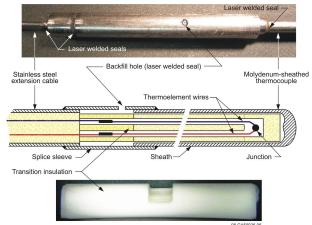


Fig. 15. Loose assembly splice sleeve.

The splice between thermoelements and wires in the extension cable is actually achieved inside a transition piece of insulation. A window is machined into the insulation as shown in Fig. 15 so that wires can be joined using a laser welder. Both halves of the sleeve can then be positioned to cover the wire splice. Three sealing welds are needed (connecting the two halves of the splice sleeve, between the extension cable sheath and the splice sleeve, and between

the splice sleeve and the TC sheath). At this point, a vacuum/backfill process is used to fill the TC sheath with an inert gas. (To date, both helium and argon have been used to backfill.)

A specialized vacuum fixture had to be designed and developed to complete the vacuum/backfill process. As shown in Fig. 16, a five-way cross is at the heart of this fixture. The 2.54 cm OD cross is stainless steel and fitted with quick connect vacuum flanges on all five legs.



Fig. 16. Splice sleeve vacuum fixture.

To complete the vacuum/backfill, the backfill hole in the splice sleeve is centered in the fixture by inserting the TC straight through two legs of the cross. Quick connect vacuum flanges with integral Conax fittings are used to seal against the TC and extension cable sheaths. One leg of the cross is connected to an inert gas supply through an isolation valve. The leg opposite the inert gas supply is connected to a vacuum pump through another isolation valve. The remaining leg of the five-way cross is fitted with a glass view port. Vacuuming is completed with the valve on the inert gas supply closed and the valve on the vacuum pump open. After vacuuming to ~10<sup>-5</sup> torr, the vacuum isolation valve is closed (with the vacuum pump running); and the inert gas isolation valve is opened to pressurize the vacuum chamber and the loose assembly TC (through its backfill hole) with an inert gas at ~3 psig. With the inert gas pressure applied, a weld is then made through the view port with a laser welder to seal the backfill hole.

A test is currently underway to compare performance of loose assembly HTIR-TCs against swaged HTIR-TCs at high temperatures. The test is being conducted at 1500 °C for a minimum of 1000 hr. Results are expected to provide some insights into the potential benefits of the loose assembly design.

### IV. COST REDUCTION OPTIONS

Efforts to reduce HTIR-TC cost have focused on the application of improved fabrication techniques and the identification of appropriate extension cable for use with HTIR-TCs.

### IV.A. Automated Fabrication Techniques

Certain customers have required HTIR-TCs in very long lengths (up to 9 m). Swaging these TCs required as many as three people to feed, lubricate, rotate, and catch the instrument as it is discharged from the swager (see Fig. 17).



Fig. 17. Three people initially required to swage long TCs.

To reduce fabrication costs for these long TCs, a device was designed and developed at the INL to automate parts of the swaging process. The device includes provisions for lubricating, rotating, and feeding the TC as indicated in Fig. 18. This innovation has the added benefit of a very uniform feed rate that is difficult to achieve manually. The uniformity of the feed rate has been tied to improvements in sensor surface conditions. Although one individual is still needed, primarily to monitor the process, this development has substantially reduced HTIR-TC fabrication cost.

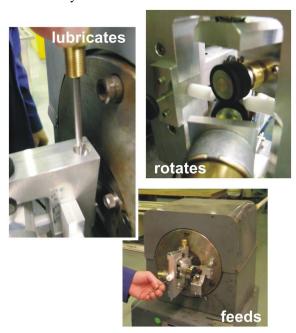


Fig. 18. Automated swaging reduced manpower by 2/3.

Like the swaging process, the calibration process initially used to produce HTIR-TCs was labor intensive and it was apparent that costs could be reduced with automation. The primary steps taken to reduce calibration costs are depicted in Fig. 19.



Fig. 19. Steps taken to reduce calibration costs (including use of multiple process tubes with inert gas flow control and implementation of programmable furnace control).

Specifically, the upper half of Fig. 19 illustrates the use of multiple process tubes within a tube furnace. This approach allows the simultaneous calibration of multiple TCs in a single furnace run, including the important potential for segregating TCs that may have sheath (material) incompatibilities at high temperatures. Critical control of inert gas flow into multiple tubes was addressed through development of a header fitted with adjustable needle valves. This ensures flow balance into each tube thereby protecting all TCs from oxidation.

Implementation of programmable furnace control is also depicted in Fig. 19. This allows the collection of data (through a computerized acquisition system) at many different user-specified temperatures (up to ~250 as currently configured) without operator attendance. Consequently, calibration can proceed for days in an automated mode, which enables cost-effective collection of very detailed data.

### IV.B. Extension Cable

HTIR-TC components are relatively expensive. To control cost, use of extension cable should always be considered in regions where the presence of cooler temperatures make this feasible. The identification of suitable HTIR-TC extension cable was of interest for that reason.

Previous research with various molybdenum and niobium alloys indicated that copper / nickel alloys may be suitable as extension cable. Unfortunately, molybdenum and niobium alloys previously studied are not exactly the same as materials used in HTIR-TCs. Consequently, INL/UI independently evaluated various copper / nickel alloys as reported in Reference 12.

Copper / nickel alloys that were evaluated are listed in Table IV. All of these alloys were selected from commercially available stock. The evaluation began by fabricating bare wire TCs consisting of pairings between KW-Mo and each of the copper / nickel alloys and pairings between Nb-1%Zr and each of the copper / nickel alloys. The resulting thermoelectric response for all bare wire TCs is shown in Figs. 20 and 21. These results are consistent with results from previous studies in that the thermoelectric response increases with increasing nickel content.

Table IV Copper / Nickel Alloys Evaluated

Wire	Vendora	Redraw Stock Chemical Analysis	Finished Wire Chemical Analysis		
Cu-3.5%Ni	1	3.50%Ni	3.6%Ni		
Cu-5.0%Ni	1	4.95%Ni	5.1%Ni		
Cu-6.5%Ni	1	6.49%Ni	6.6%Ni		
Cu-11%Ni	1	10.63%Ni	11%Ni		
Cu-22%Ni	1	23.83%Ni	22%Ni		
Cu-30%Ni	2	30.90%Ni	30%Ni		
99.9% Cu	1	NA	NA		
a All Vendor 1 alloys were "annealed" Cu-30%Ni from					

a. All Vendor 1 alloys were "annealed". Cu-30%Ni from Vendor 2 was "as drawn temper".

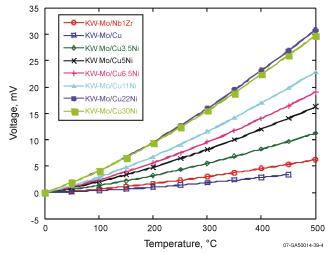


Fig. 20. Thermoelectric response for KW-Mo paired with various copper / nickel alloys.

Data from Figs. 20 and 21 were used to predict copper / nickel alloys combinations that could yield a thermoelectric response similar to a KW-Mo / Nb-1%Zr TC (at low temperatures). These predictions were useful in identifying copper / nickel combinations that merited further evaluation. Copper / nickel combinations consistent with the predictions were then used to fabricate bare wire TCs. The thermoelectric response of these bare wire TCs are shown in Figs. 22 and 23. As indicated, the measured results confirm that the thermoelectric response of the predicted copper / nickel combinations are comparable to the thermoelectric response of a KW-Mo / Nb-1%Zr TC.

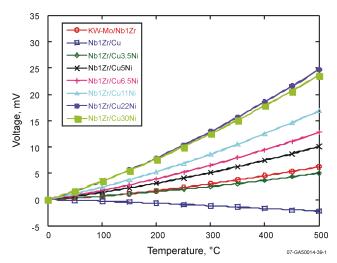


Fig. 21. Thermoelectric response for Nb-1%Zr paired with various copper / nickel alloys.

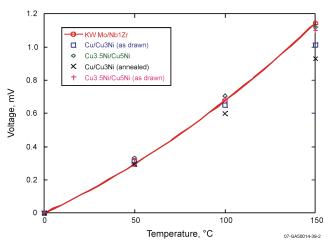


Fig. 22. Thermoelectric response for selected Cu / Ni alloys for temperatures up to 150  $^{\circ}\text{C}.$ 

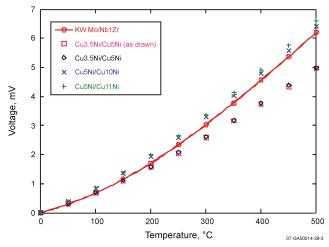


Fig. 23. Thermoelectric response for selected Cu / Ni alloys for temperatures up to 500  $^{\circ}\text{C}.$ 

Close evaluation of Figs. 22 and 23 indicate that the thermoelectric response of a Cu-3.5%Ni / Cu-5%Ni combination is the best match compared to the response of KW-Mo / Nb-1%Zr at temperatures up to 150 °C. Similarly, a Cu-5%Ni / Cu-10%Ni combination is the best match in the range of 150 to 500 °C. Although some refinement of these conclusions could develop through testing of custom alloys, the results provide the data needed to suggest suitable, commercially-available extension cable for minimizing the cost associated with using HTIR-TCs.

### V. SUMMARY

Several options have been identified to further improve recently-developed INL HTIR-TCs for in-pile testing. These options, which were evaluated through a joint INL/UI effort, have the potential to reduce costs and allow HTIR-TC use in higher temperature applications (up to at least 1800 °C). Options were considered to enhance HTIR-TC performance (through the use of specially formulated alloys, improved heat treatments, and the adoption of alternate geometries) and options were considered to reduce HTIR-TC costs (through automated fabrication techniques and the use of copper/nickel alloys as extension cable).

Evaluations of specially formulated alloys indicate that molybdenum/low niobium alloys and niobium/low molybdenum alloys tested offer no advantages relative to the KW-Mo / Nb-1%Zr thermoelements identified in the original HTIR-TC design. Furthermore, slight improvements in thermoelectric resolution provided by an ODS-Mo / Nb-1%Zr combination are not sufficient to offset the lower cost of commercially available KW-Mo wire. Consequently, KW-Mo / Nb1%Zr thermoelements appear to offer the best performance achievable at this time.

Results from heat treatment investigations indicate that the heat treatment temperature may be more important to HTIR-TC stability than the heat treatment duration. Results also suggest that some attention should be given to the heat treatment duration because the thermoelectric response tends to decrease as treatment duration increases. These suggestions may be limited to a relatively narrow range of application temperatures (between ~1300 °C and ~1500 °C). Consequently, additional evaluation may be warranted to allow for more general application of these results.

INL/UI evaluations confirm that TC stability and reliability tend to increase with thermoelement diameter. That result should be considered (along with cost and the potential effects on response time) in the specification of a HTIR-TC for any given application. Potential advantages of a loose assembly TC configuration led to the initiation of tests that will ultimately allow performance comparisons to swaged designs. Results from these tests, which are being conducted at 1500 °C for a minimum of 1000 hr, should provide additional guidance for HTIR-TC specification.

Devices and procedures have been developed at INL to automate swaging and heat treating procedures associated with HTIR-TC fabrication. In addition, investigations have been completed to identify suitable extension cable for use with HTIR-TCs. These efforts have led to significant cost reductions, which should promote consideration of the selection of this instrument in demanding high temperature irradiation conditions.

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### **REFERENCES**

- 1. J. L. Rempe, D. L. Knudson, K. G. Condie, and S. C. Wilkins, "Thermocouples for High Temperature In-Pile Testing," *Nuclear Technology*, **156**, **No. 3**, p. 320 (2006).
- E. D. Zysk and A. R. Robertson, "Newer Thermocouple Materials," presented at the *Fifth Symposium on Temperature*, Washington, DC (1971); published in H. H. Plumb (Ed.-in-Chief), *Temperature: Its Measurement and Control in Science and Industry*, Vol. 4, Part 3, p. 1697, American Institute of Physics, New York (1972).
- 3. S. C. Wilkins, "Low Cross-Section Mo-Nb Thermocouples for Nuclear Applications: The State-of-the-Art," *Fifth Symposium on Space Nuclear Power Systems*, Albuquerque, NM (1988).
- 4. R. C. Knight and D. L. Greenslade, "Irradiation Testing of a Niobium-Molybdenum Developmental Thermocouple," *Westinghouse Hanford Company Technical Report WHC-SA-1256-FP* (1991); presented at the *Ninth Symposium on Space Nuclear Power Systems*, Albuquerque, NM (1992).

- 5. S. C. Wilkins, "Characterization and Materials-Compatibility Tests of Molybdenum-Niobium Thermocouples," *Temperature: Its Measurement and Control in Science and Industry*, Toronto, Ontario, Canada (1992).
- 6. D. A. Prokoshkin and E. V. Vasil 'eva, *Alloys of Niobium*, S. Monson, Jerusalem, Israel (1964).
- R. Schley and G. Metauer, "Thermocouples for Measurements under Conditions of High Temperature and Nuclear Radiation," J. F. Schooley (Ed.-in-Chief), Temperature: Its Measurement and Control in Science and Industry, Vol. 5, Part 2, p. 1109, American Institute of Physics, New York (1982).
- 8. G. M. Ludtka, T. G. Kollie, R. L. Anderson, and W. H. Christie, "Performance of Chromel versus Alumel and Nicrosil versus Nisil Thermocouple Assemblies in Vacuum and Argon Between 1000 °C and 1200 °C," *Industrial Temperature Measurement Conference* (1984).
- 9. Pratt & Whitney Aircraft, "Development of High Temperature Sensors," Pratt & Whitney Report PWAC-339 (1961).
- S. Fanciulllo, "Thermocouple Development, Lithium-Cooled Reactor Experiment," Pratt & Whitney Report PWAC-422 (1964).
- 11. C. Davoine, M. Sardin, G. Metauer, and M. Gantois, "Etude de la compensation entre 0 C to 300 C des thermocouples Mo<sup>+</sup>/Nb<sup>-</sup>," Research Report (1974).
- J. E. Daw, J. L. Rempe, D. L. Knudson, S. C. Wilkins, and J. C. Crepeau, "Extension Wire for High Temperature Irradiation Resistant Thermocouples," *Measurement Science and Technology*, MS 264021, accepted January 2008.