

Summary of Thermocouple Performance during Advanced Gas Reactor Fuel Irradiation Experiments in the Advanced Test Reactor and Out-of-Pile Thermocouple Testing in Support of Such Experiments

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Summary of Thermocouple Performance during Advanced Gas Reactor Fuel Irradiation Experiments in the Advanced Test Reactor and Out-of-Pile Thermocouple Testing in Support of Such Experiments

Abstract—High temperature gas reactor experiments create unique challenges for thermocouple-based temperature measurements. As a result of the interaction with neutrons, the thermoelements of the thermocouples undergo transmutation, which produces a time-dependent change in composition and, as a consequence, a time-dependent drift of the thermocouple signal. This drift is particularly severe for high temperature platinum-rhodium thermocouples (Types S, R, and B) and tungsten-rhenium thermocouples (Type C). For lower temperature applications, previous experiences with Type K thermocouples in nuclear reactors have shown that they are affected by neutron irradiation only to a limited extent. Similarly, Type N thermocouples are expected to be only slightly affected by neutron fluence. Currently, the use of these nickel-based thermocouples is limited when the temperature exceeds 1000°C due to drift related to phenomena other than nuclear irradiation. High rates of open-circuit failure are also typical. Over the past 10 years, three long-term Advanced Gas Reactor experiments have been conducted with measured temperatures ranging from 700°C–1200°C. A variety of standard Type N and specialty thermocouple designs have been used in these experiments with mixed results. A brief summary of thermocouple performance in these experiments is provided.

Most recently, out-of-pile testing has been conducted on a variety of Type N thermocouple designs at the following (nominal) temperatures and durations: 1150°C and 1200°C for 2,000 hours at each temperature, followed by 200 hours at 1250°C and 200 hours at 1300°C.

The standard Type N design utilizes high purity, crushed MgO insulation and an Inconel 600 sheath. Several variations on the standard Type N design were tested, including a Haynes 214 alloy sheath, spinel (MgAl_2O_4) insulation instead of MgO, a customized sheath developed at the University of Cambridge, and finally a loose assembly thermocouple with hard-fired alumina insulation and a molybdenum sheath. The most current version of the High Temperature Irradiation Resistant Thermocouple, based on molybdenum/niobium alloys and developed at Idaho National Laboratory, was also tested.

Index Terms—In-Pile Instrumentation, Material and Test Reactors, High-temperature Irradiation, Thermocouples, Thermocouple Testing.

I. INTRODUCTION

As part of the U.S. Department of Energy-sponsored Next Generation Nuclear Project, Idaho National Laboratory (INL) is currently conducting an experiment series designated the Advanced Gas Reactor (AGR) tristructural isotopic fuel experiments to qualify this fuel's irradiation performance for High Temperature Gas Reactor (HTGR) applications. The first experiment, AGR-1, was irradiated in the INL Advanced Test Reactor (ATR) from December 2006 to November 2009 [1]. The second experiment, AGR-2, was irradiated in the ATR from June 2010 to October 2013. The third experiment was designated AGR-3/4 and was irradiated in the ATR from December 2011 to April 2014 [2].

The fuel form for all three of these tests was similar. The fuel particles were coated with a thermosetting resin and pressed into cylindrical fuel compacts, which were then placed into graphite holders. Metal-sheathed thermocouples and gas lines were inserted into small holes drilled into the graphite holders. The graphite holders were sealed inside stainless steel capsules that served as the pressure boundary between the ATR coolant water and the inert gas environment of the capsules' interior. The thermocouples and gas lines passed through the top of each capsule head and were sealed by brazing. Each of the three tests consisted of six or more individual capsules welded together, with passages in the capsules above to allow instrumentation and gas lines from capsules below to pass through, as illustrated in Figs. 1 and 2.

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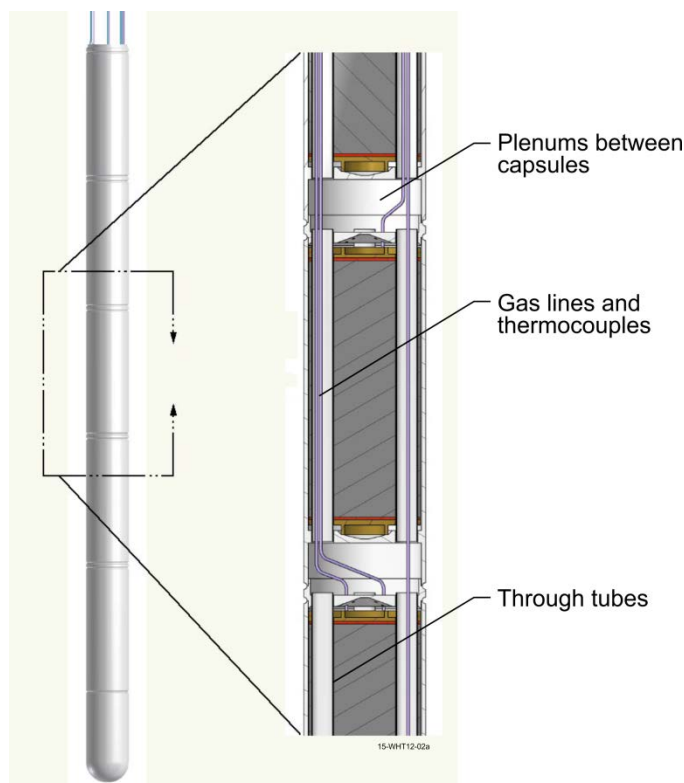


Fig 1. Elevation view of typical AGR fuel experiment.

The final test in the AGR series of experiments is denoted AGR-5/6/7 and is a combination of three experiments that were envisioned at the beginning of the program. There will be five separate capsules in the experiment train, and the conditions created in these capsules will serve as the formal fuel qualification irradiations (AGR-5/6) and the margin test (AGR-7). The purpose of the margin test is to demonstrate that there is a substantial margin between the highest fuel temperature in an operating HTGR and the temperature at which the fuel particle failure rate becomes unacceptable.

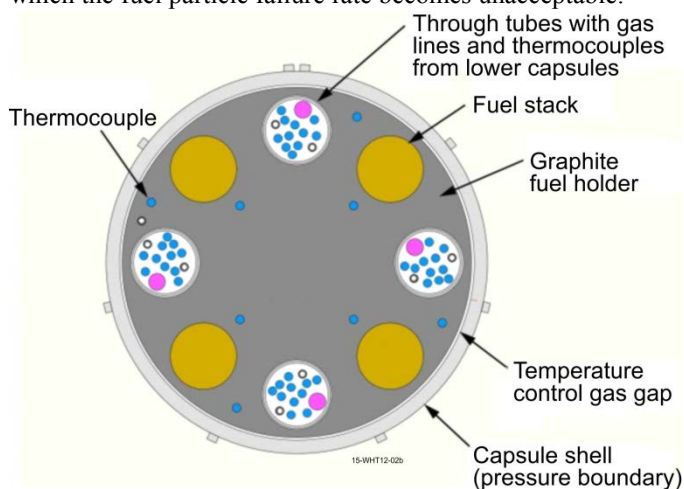


Fig 2. Cross-section view of AGR-5/6/7 fuel experiment.

Temperature measurement has been and continues to be an important but challenging aspect of these fuel performance tests. As a result of the interaction with neutrons, the

thermoelements of the thermocouples can undergo transmutation, which produces a time-dependent change in composition and, as a consequence, a time-dependent drift of the thermocouple signal. This drift is particularly severe for high temperature platinum-rhodium thermocouples (Types S, R, and B) and tungsten-rhenium thermocouples (Type C) [3, 4]. These thermocouples types, which are used routinely for high temperature measurements outside reactors, are only used in very special circumstances for reactor experiments. For lower temperature applications, previous experiences with Type K thermocouples in nuclear reactors have shown that they are affected by neutron irradiation only to a limited extent [5]. Similarly, Type N thermocouples are expected to be only slightly affected by neutron fluxes [6]. Currently, the use of these nickel-based thermocouples is limited when the temperature exceeds 1000°C due to drift related to phenomena other than nuclear irradiation. High rates of open-circuit failure are also typical.

The drift in Type N thermocouples has been traced to composition changes in the thermoelements due to elemental migration between the thermoelements, and between the metallic sheath and the thermoelements. Changes in thermocouple size, insulation composition, and sheath composition are being investigated to reduce drift in Type N thermocouples. Thermocouples based on low cross-section refractory thermoelements (molybdenum/niobium) are also being investigated. As an aid to final thermocouple selection for the AGR-5/6/7 experiment, a furnace test of various thermocouple designs, similar to one conducted at INL in 2005 [7], has been performed.

II. THERMOCOUPLE PERFORMANCE IN PAST AGR TESTS

A. AGR-1

Fig. 3 is a summary of the thermocouple failure trends during the three AGR experiments irradiated to date in ATR. The AGR-1 experiment saw an overall thermocouple failure rate of about 53% by the end of the irradiation period (approx. 620 full power days). Three different thermocouple types were used in this experiment. An INL-developed, doped molybdenum/niobium alloy thermocouple, with hafnia insulation (designated the High Temperature Irradiation Resistant-thermocouple [HTIR-TC]) [8], was installed in 11 locations (although three were broken during some phase of installation and startup). Of the eight 1.57 mm diameter HTIR-TC operating successfully at startup, five survived the entire irradiation period. These HTIR-TCs operated within a temperature range of 900°C–1200°C, the most severe environment for any of the TCs used, and had a better than average survival rate of 62%.

The second thermocouple type used in AGR-1 was a standard 1.57 mm diameter, Type N, crushed MgO insulation, Inconel 600 sheath design. This design was used in five locations, and only one survived the entire irradiation period. The operating temperature range was 750°C–900°C.

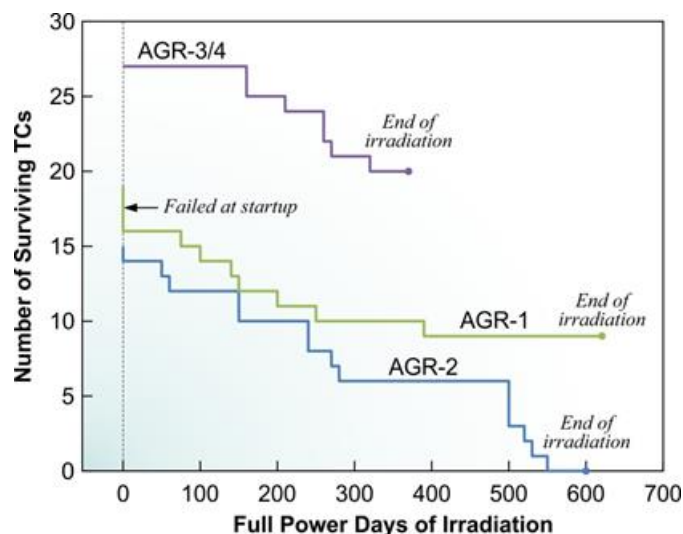


Fig. 3. Thermocouple failures as irradiation progressed for three AGR experiments.

The third thermocouple type used in AGR-1 was a 2.29 mm diameter, Type N, molybdenum sheath design with hard-fired, loose-pack alumina insulation. This design was used in three places, all in the top capsule, and all three survived the entire irradiation period. These thermocouples operated within a temperature range of 750°C–800°C. These thermocouples exhibited very good performance, but it was postulated that this may have been due to the lower operating temperature, larger size, and shorter heated length rather than an inherently superior design. Subsequent out-of-pile testing, described later, has been done to better understand this design.

B. AGR-2

Fig. 3 also shows the overall performance of the thermocouples used in AGR-2. In this case, there were a total of 15 thermocouples, and by the end of irradiation (600 days), all of them had failed. These thermocouples operated within a temperature range of 800°C–1050°C, with the majority operating around 950°C. For AGR-2, all of the thermocouples were of the same design: 2.0 mm diameter, Type N, crushed MgO insulation, niobium sheath. It was anticipated that the thicker thermoelements would result in better survivability than in AGR-1. However, this was not the case.

The reason for the comparatively poor performance of the AGR-2 thermocouples was probably a combination of three factors: higher temperatures, refractory metal sheaths, and handling stress. The niobium sheaths have a high affinity for oxygen. Even very minute quantities of oxygen or moisture present in the sweep gas, when integrated over thousands of hours, may have served to embrittle the sheaths and make the thermocouples more prone to failure when subjected to thermal expansion or handling. This experiment was subjected to substantial handling stress as the entire experiment test train was removed from its irradiation position and reinstalled two different times: once at about 200 days of irradiation and a second time at about 500 days of irradiation. The failures of the last six thermocouples seem to be directly correlated to the

second handling sequence.

C. AGR-3/4

The performance of the AGR-3/4 thermocouples is also illustrated in Fig. 3. This test used standard Type N thermocouples with crushed MgO insulation in an Inconel 600 sheath. There were 12 capsules in this test, and the bottom 11 used 1.0 mm diameter thermocouples. After 369 days of irradiation, only seven of the 24 thermocouples in the bottom 11 capsules failed, for an overall survival rate of 71%. These thermocouples operated in a temperature range of 600°C–1050°C, with the great majority operating at 600°C–800°C. The top capsule had three 2.0 mm, standard Type N thermocouples, two of which operated at 550°C and the third at 800°C, and all survived the entire test. The relatively good performance of the AGR-3/4 thermocouple set was probably due to the lower operating temperatures of most of the thermocouples and the shorter irradiation time.

III. AGR-5/6/7 DESIGN FEATURES

The final test in the AGR series of experiments is denoted AGR-5/6/7 and it is a combination of three experiments that were envisioned at the beginning of the program. The AGR-5/6/7 experiment is being designed so that some of the thermocouples will be placed in temperature zones hotter than any of the three previous experiments. About 25% of the thermocouples will operate in a temperature range of 1150°C–1300°C, and one thermocouple location has a projected temperature of 1400°C. The number and size of thermocouples in the previous three AGR experiments was limited by the space constraints of passages used to route the thermocouples through the experiment. The design of the AGR-5/6/7 capsules permits much larger passages. There will be room for at least ten 1.57 mm thermocouples per capsule. This will statistically increase the chances of retaining at least one operating thermocouple in each capsule at the end of irradiation. As explained above, some locations will operate at temperatures above 1200°C; however, each of the five AGR-5/6/7 capsules has locations for thermocouples at temperatures less than 900°C. Another favorable factor is that the irradiation period for AGR-5/6/7 will be on the order of 450–500 days versus the 600 day plus irradiation campaigns of AGR-1 and AGR-2.

IV. PREVIOUS OUT-OF-PILE TESTING

Although there is room for more thermocouples in the AGR-5/6/7 test, the higher temperature range for a significant fraction of the thermocouples greatly increases the chances for a high rate of failure for those in the hot zones. This concern is not new for the AGR experiment program.

During the initial design of the AGR-1 experiment (circa 2005), it was recognized that the combination of high temperature and a high neutron flux environment would be a challenge for any commercial thermocouple design. At temperatures above 1000°C, even the best base metal thermocouple type, Type N, is subject to drift and open circuit failure. At that time, several thermocouple vendors advertised

versions of Type N thermocouples that were less subject to drift and claimed these designs would survive for longer periods at high temperature. Also, in the 2005 timeframe, the HTIR-TC thermocouples had shown some early success. To test these various designs against each other, a 4,000-hour, 1200°C, out-of-pile furnace testing program was commissioned [7]. The results from that effort were: the worst Type N thermocouples drifted downward as much as 100°C, the best drifted about 40°C, and the HTIR-TC thermocouples drifted only about 20°C. It was expected that several of the thermocouples would experience open circuit failure during the test, but not a single thermocouple (out of more than 20) failed completely.

V. NEW OUT-OF-PILE FURNACE TESTING

A. Background and Overview

As an aid to the final thermocouple selection for AGR-5/6/7, a test similar to that conducted in 2005 has been performed. The results and discussion of this test constitute the balance of this paper.

The test was conducted in a tube furnace with the thermocouples embedded in a block of graphite, see Fig. 4 and Fig. 5. (Note that the Fig. 4 photo was taken near the end of the 1150°C phase of the test, after the furnace had been shut down and the internal assembly removed.) The test atmosphere was argon at flow rate of 250 cc/min. The oxygen concentration as measured at the outlet of the furnace was always less than 1 ppb. However, see Section VI for a further discussion of oxygen impurities in the furnace atmosphere.

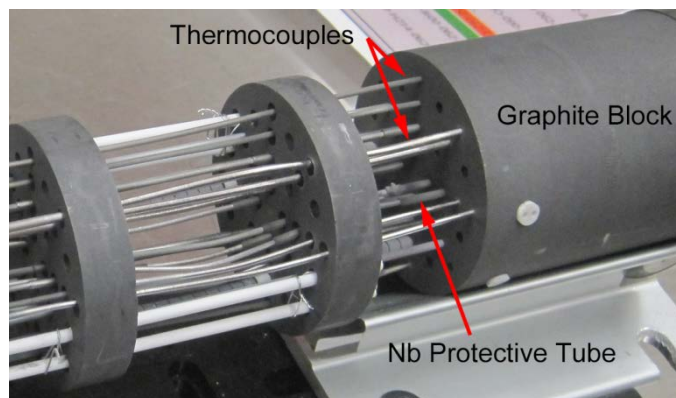


Fig. 4. In-furnace assembly after 1,800 hours at 1157°C. All thermocouple tips were inserted 50 mm into the graphite block and supported periodically by graphite spiders.

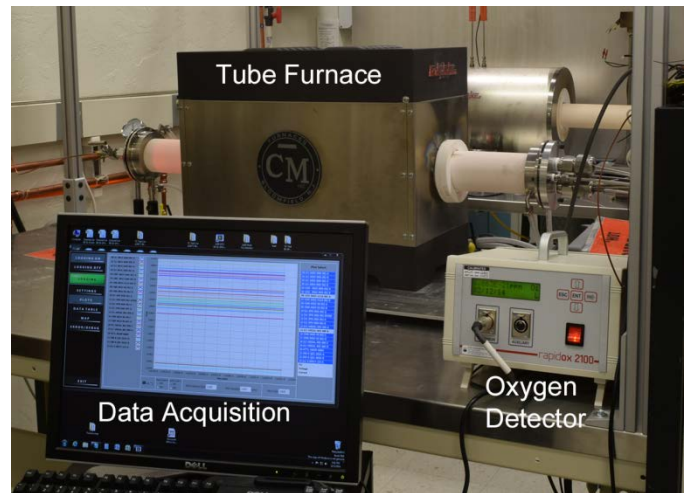


Fig. 5. Experiment setup showing a tube furnace in operation, the data acquisition system, and an oxygen detector.

In addition to varying the size from 1.57 mm to 2.34 mm, the following variations on a standard Type N design were tested. The test used a:

- 1) Haynes 214 alloy sheath rather than Inconel 600
- 2) Spinel insulation (MgAl_2O_4) rather than MgO insulation
- 3) Special sheath material developed at Cambridge University [9] rather than Inconel 600
- 4) Loose assembly with molybdenum sheath, similar to the design used in the AGR-1 top capsule, as described above.

A current version of the HTIR-TC thermocouple design was also included in the test.

The furnace test was conducted as follows: 2,060 hours at 1157°C followed by 2,006 hours at 1207°C, 201 hours at 1258°C, and 266 hours at 1301°C (with a 77 hour excursion at the beginning of this period to 1311°C). Figs. 6–13 provide comparisons of the thermocouple designs being evaluated compared to two representative “standard construction” Type N thermocouples from two different manufacturers: “M1” and “M2.” The standard construction thermocouples were 1.57 mm in diameter and utilized Inconel 600 sheaths and high purity MgO insulation. In these figures, the open circles that appear at the end of some of the data traces indicate the point at which the sensor exhibited signs of open circuit failure. During the test, the furnace was shut down 10 different times and allowed to return to room temperature (this simulated reactor outages, which appear to contribute to thermocouple failure). In two cases, the test assembly was pulled from the furnace, examined, and additional thermocouples were added. In the interest of clarity, these temperature cycles have been removed from the plots shown in Figs. 6–13.

Stability of the furnace temperature was verified by comparing the furnace control thermocouple (Type B) reading to a Type S thermocouple placed in the graphite block near the thermocouples being tested.

B. Size Variation

Fig. 6 illustrates that there was little benefit obtained by increasing the thermocouple size from 1.57 mm to 2.34 mm for the 1157°C test period (this confirms results obtained by [10]). However, the larger size appeared to offer notably reduced drift at 1207°C. Note that these larger-sized thermocouples did not survive the 1258°C test period.

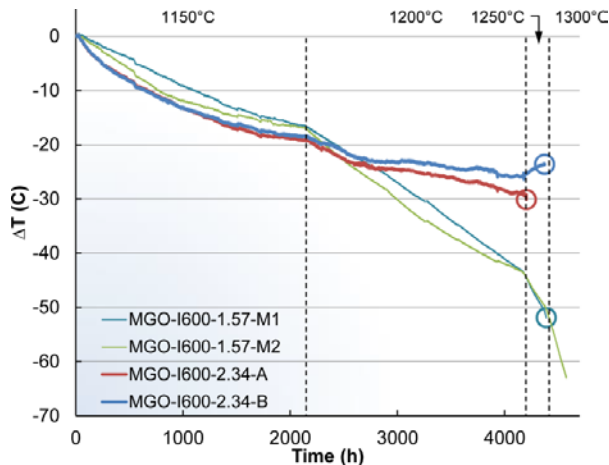


Fig. 6. Drift of 2.34 mm OD (outer diameter) standard Type N thermocouples compared to 1.57 mm OD standard Type N thermocouples.

C. Haynes 214 Alloy Sheath

Haynes 214 is an alloy that is designed to be highly oxidation and carburization resistant. Fig. 7 shows that the thermocouples with the Haynes 214 sheaths exhibited a small but consistent improvement in drift rate for both the 1157°C and 1207°C temperature ranges compared to the standard construction thermocouples. The H214 sheathed thermocouple, designated “A,” was surrounded by a niobium protective sleeve 0.25 mm thick. The presence of a niobium sleeve, or lack thereof, did not seem to make any appreciable difference. Similar conclusions were reached by [10]. Note that neither of these two thermocouples survived beyond the 1258°C portion of the experiment.

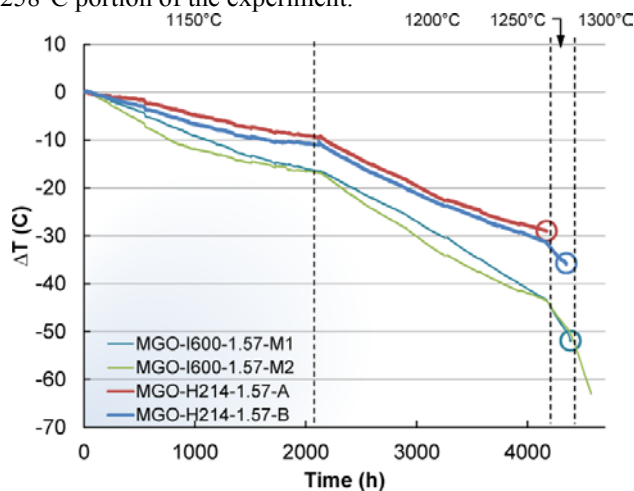


Fig. 7. Drift of 1.57 mm OD Type N thermocouples with Haynes 214 alloy sheaths compared to 1.57 OD mm standard Type N thermocouples.

D. Spinel Insulation

Spinel insulation has been considered for thermocouples for nuclear use for at least 20 years [Patent WO1993005520 A1]. Spinel is less reactive with the thermoelements than MgO but, like MgO, causes less damage to the thermoelements during the drawing process compared to alumina, which has harder grains. Figs. 8 and 9 show the results of substituting Spinel insulation for the standard MgO insulation for two different sizes of thermocouples. As shown in Fig. 8, the 1.57 mm Spinel-insulated thermocouple provided only marginal improvement over 1.57 mm standard Type N. The 2.34 mm Spinel-insulated thermocouple performed better than a 2.34 mm standard construction Type N during the 1157°C phase of the experiment (compare Fig. 6), but overall the drift was about the same at the end of the 1258°C phase. Based on these observations, Spinel insulation might be a slight improvement over standard MgO insulation.

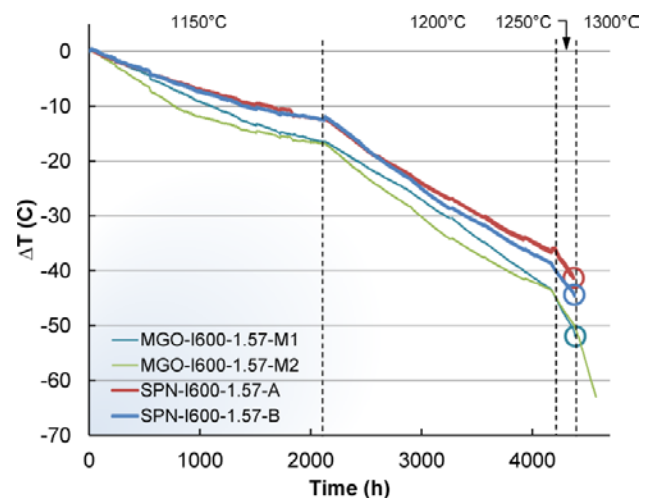


Fig. 8. Drift of 1.57 mm OD Type N thermocouples with Spinel insulation compared to 1.57 mm OD standard Type N thermocouples.

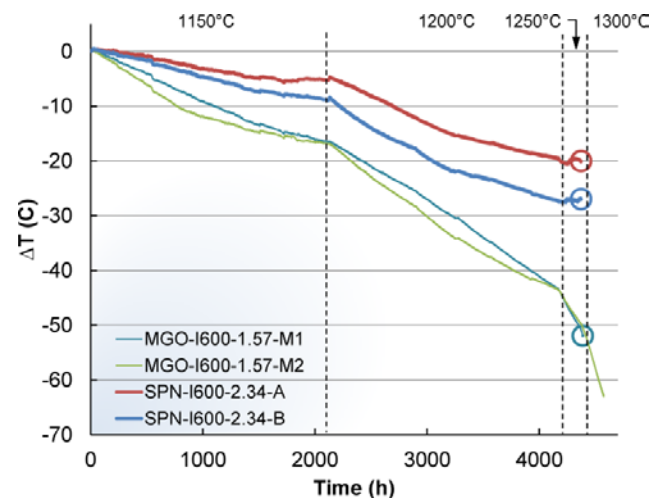


Fig. 9. Drift of 2.34 mm OD Type N thermocouples with Spinel insulation compared to 1.57 mm OD standard Type N thermocouples.

E. Cambridge Special Alloy Sheath

Fig. 10 illustrates the performance of two Cambridge special sheath thermocouples against the two standard construction Type N thermocouples. Both of these thermocouples were surrounded by niobium protective sleeves. In this case, the performance of the Cambridge design is clearly superior to the standard construction thermocouples. After 2,060 hours at 1157°C, the special sheath Cambridge thermocouples had drifted an average of only 4°C, and after an additional 2,000 hours at 1207°C, the total drift was about 15°C. In spite of a temperature excursion above 1300°C and some impure gas, both thermocouples provided temperature signals to the end of the test, although the drift became much more pronounced during the 1300°C phase of the test. This may have been due to a degradation of the sheath from oxygen contamination of the purge gas (see Section VI).

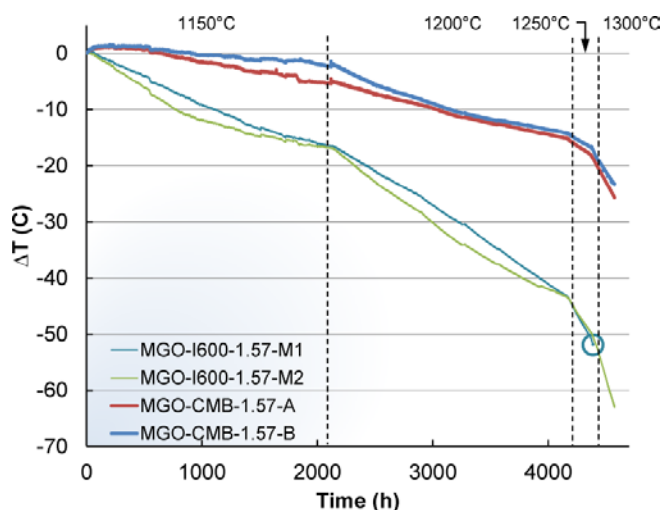


Fig. 10. Drift of 1.57 mm OD Type N thermocouples with Cambridge special alloy sheaths compared to 1.57 mm OD standard Type N thermocouples.

F. Loose Assembly, Hard-Fired Insulation, Molybdenum Sheath

Fig. 11 provides a comparison of two loose assembly, hard-fired alumina insulated, molybdenum-sheathed Type N thermocouples against the two standard Type N thermocouples. These thermocouples are 2.29 mm OD with 0.25 mm thermoelements. These loose assembly thermocouples are clearly superior to the standard design in terms of drift. Interestingly, these thermocouples seem to have a break-in period lasting 400 to 600 hours, during which time they drift upward. After that period, the drift is very small and downward. Because of this unusual behavior, Fig. 12 was generated to provide additional insight into the performance of this thermocouple design. It shows the absolute temperature reading of these two loose assembly thermocouples compared to a reference grade Type S thermocouple that was placed in the graphite block. During the 1157°C period, the loose assembly thermocouples started below the reference temperature and then drifted above it. However, after the first temperature increase, the surviving loose assembly thermocouple (HRDAL-MO-2.29-A) provided a temperature

reading nearly identical to the Type S reference thermocouple until the end of the test. Of the 20 plus thermocouples that started the test, HRDAL-MO-2.29-A was the only one that both survived and read within 10°C of the reference Type S at the end of the test.

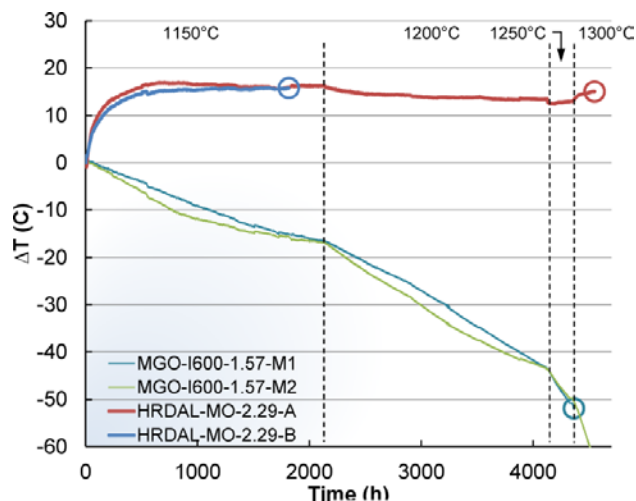


Fig. 11. Drift of 2.29 mm OD, Type N, loose assembly thermocouples with hard-fired alumina insulation and molybdenum sheaths compared to the 1.57 mm OD standard Type N thermocouples.

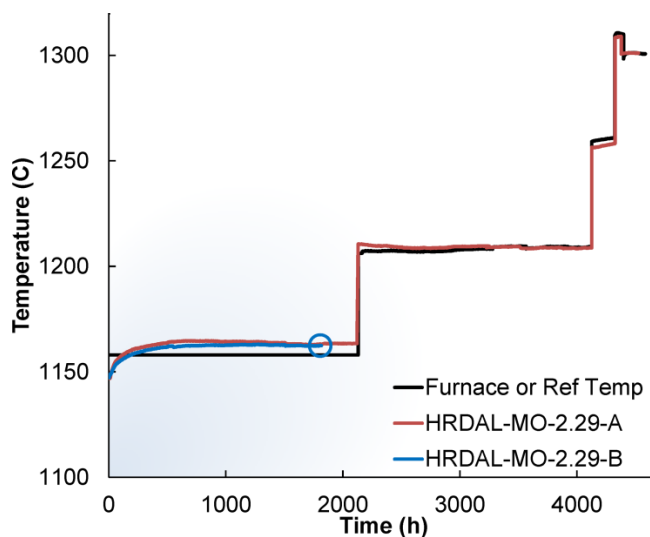


Fig. 12. Comparison of temperature readings from 2.29 mm OD, Type N, loose assembly thermocouples with the Type S reference thermocouple.

During the early phase of the test, it was clear that these loose assembly thermocouples were performing well, and so two more were procured and installed at about hour 3,500 of the test. The performance of this second pair of thermocouples is shown in Figs. 13 and 14. This pair of thermocouples also exhibited very low drift and a good match to the Type S reference thermocouple (within 10°C). However, both experienced open circuit failure prior to the end of the test (as did HRDAL-MO-2.29-B). The reason for these open circuit failures is probably related to differential expansion between the refractory molybdenum sheath and the high nickel alloy

Type N thermoelements. For this test, the heated length of the thermocouples was about 50 cm and ran at temperatures between 400°C and 1200°C. This resulted in an axial differential expansion of more than 3 mm between the sheath and the thermoelements. These loose assembly thermocouples were built to try and accommodate this expansion by including a hollow space in the insulators beyond the junction for the thermoelements to grow into. However, the thermoelement wires sometimes hang up along their length as they slide back and forth inside the hard-fired alumina insulators, which causes a fracture to occur.

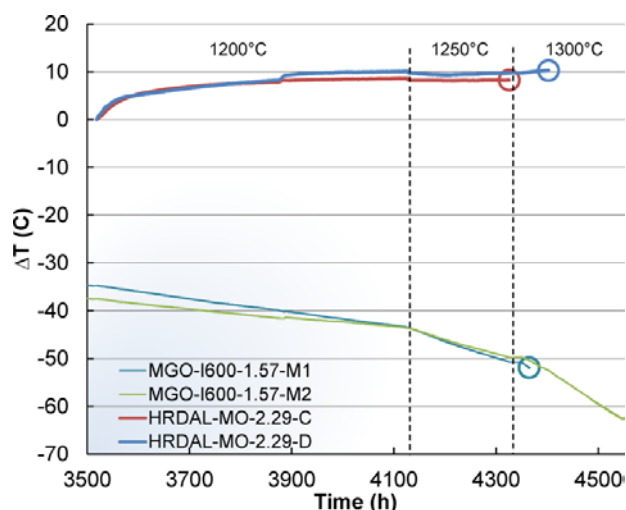


Fig. 13. The drift of a second set of 2.29 mm, OD Type N, loose assembly thermocouples with hard-fired alumina insulation and molybdenum sheaths compared to 1.57 mm OD standard Type N thermocouples.

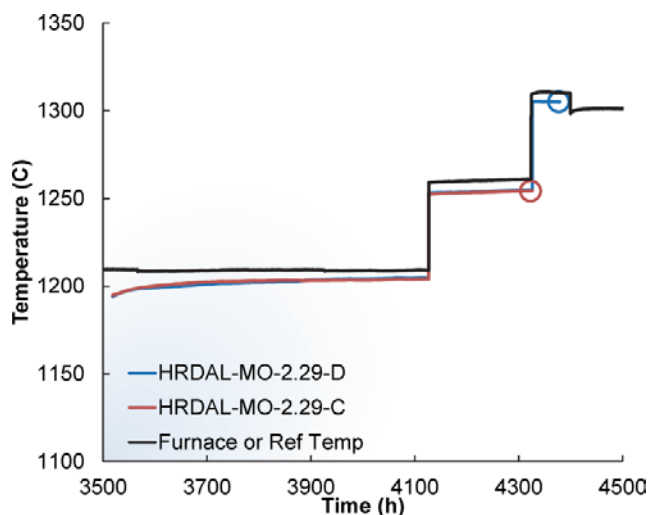


Fig. 14. Comparison of temperature readings from a second set of 2.29 mm OD, Type N loose assembly thermocouples with Type S reference thermocouple.

Another potential drawback of the molybdenum sheath loose assembly design is that the sheaths have limited flexibility. This drawback can be partially overcome by making only a short portion of the sensor from the loose assembly design, then installing a sealed transition followed

by a long length of standard crushed insulation cable with a ductile sheath material, as shown in Fig. 15. In this design, the molybdenum tip is relatively stiff, but the rest of the sensor is as ductile as a standard, crushed mineral-insulated thermocouple. This design could make sense for many high temperature reactor experiments where only a small part of the experiment volume operates at temperatures above 1000°C.

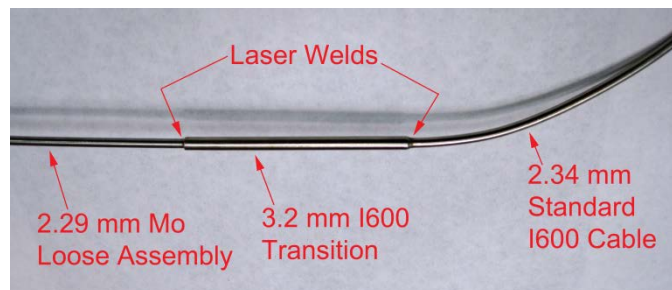


Fig. 15. Construction details for a 2.29 mm OD, Type N, loose assembly thermocouple with hard-fired alumina insulation and a molybdenum sheath.

Loose assembly designs with molybdenum sheaths are typically not rated for bending, partly because molybdenum is more brittle than typical thermocouple sheath materials (although modern manufacturing methods produce molybdenum tubing with elongation in excess of 10%). Furthermore, with the loose assembly design, bending causes fractures in the insulators and puts the thermoelement wires in tension. However, in practice, some limited bending is possible. Bend tests were performed to see if there would be enough flexibility to allow this design to be incorporated into the specific geometry of the AGR-5/6/7 experiment. Fig. 16 illustrates that the molybdenum end of this design can be bent into an offset configuration, and the 8 mm offset shown is sufficient to meet the requirements of the AGR-5/6/7 experiment.

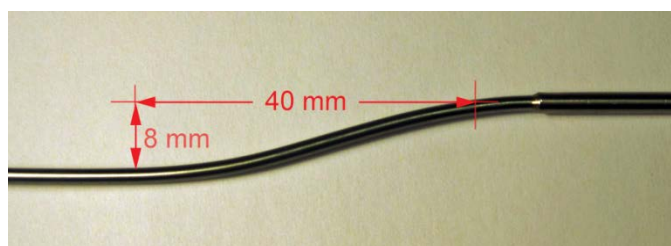


Fig. 16. The offset bend in a loose assembly, molybdenum-sheathed thermocouple.

The conclusion of the foregoing is that the molybdenum-sheathed, loose assembly design is drift resistant, and can be assembled into the experiment, but the high failure rate observed in this furnace test is still a concern. Two modifications have been identified to reduce the failure rate. (1) Reduce the length of the loose assembly portion of the probe to minimize the overall differential thermal expansion. The immersion depth into the fuel holder is typically on the order of 10 cm, and the rest of the thermocouple cable experiences temperatures less than 900°C. Reducing the loose assembly portion to about 15 cm would likely increase the life.

(2) Consider making the sheath from nickel 200/201, which would provide a coefficient of thermal expansion close to that of the thermoelements. Nickel 200/201 has the further advantage of a melt temperature substantially above Inconel 600 (1453°C versus ~1350°C) and well above the melt temperature of the thermoelements.

G. HTIR-TC

For more than 10 years, INL has been involved in the development of high temperature, drift resistant thermocouples based on molybdenum/niobium alloys. These specialty refractory thermocouples are designated HTIR-TCs. As discussed previously, these instruments have performed reasonably well in both out-of-pile and in-pile tests. Two examples of this design were included in this furnace test. These thermocouples were 1.57 mm OD with a Nb/1% Zr sheath and 0.25 mm thermoelement wires. Unfortunately, as seen in Fig. 17, one of these thermocouples (NB05) failed early in the test, after only 200 hours. These thermocouples were fabricated by drawing rather than by the swaging process used previously, and this premature failure may have been related to this change in the fabrication process. The second (NB04) demonstrated very good drift resistance before failing at the beginning of the 1300°C portion of the test. As will be discussed later, this failure was likely due to higher oxygen levels in the furnace purge gas during the latter part of the test rather than any inherent defect in the sensor itself.

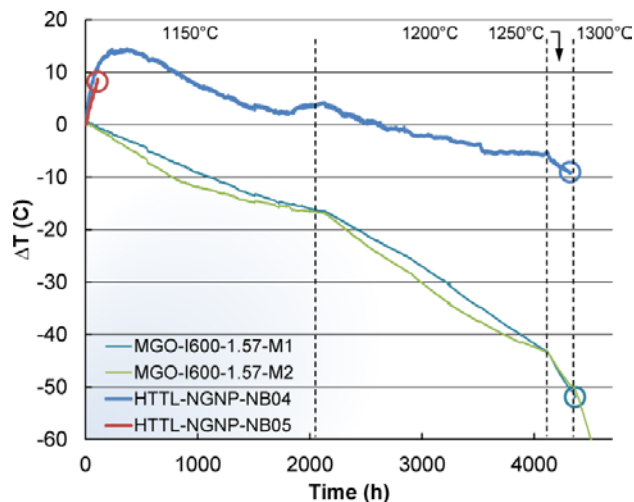


Fig. 17. Drift of 1.57 mm OD HTIR-TCs compared to 1.57 mm standard Type N thermocouples.

Fig. 18 shows a comparison of the HTIR-TCs to the furnace or Type S reference temperature. Each HTIR-TC is calibrated individually after a high temperature conditioning treatment. In this case, the heat treatment may have not been sufficiently long as the HTIR-TC reading ended up being about 50°C below the reference temperature during the 1207°C portion of the test.

Overall, the HTIR-TC performance did not meet expectations. These thermocouples are needed to measure the hottest locations in the AGR-5/6/7 experiment, especially in the AGR-7 margin test capsule. As such, another furnace test

has been commissioned to test both swaged and drawn versions as well as a longer and hotter heat treatment.

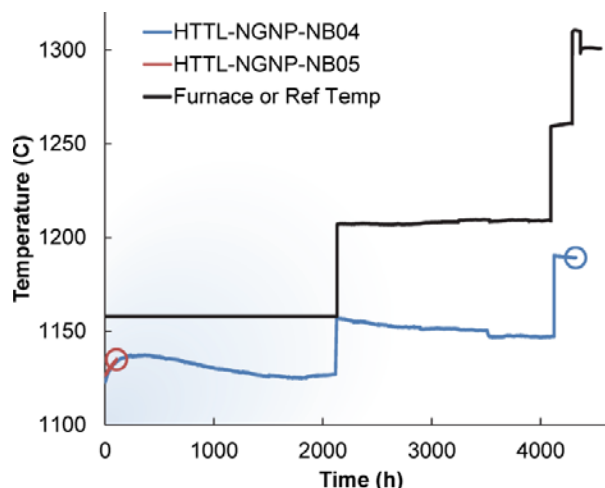


Fig. 18. Comparison of temperature readings from 1.57 mm OD HTIR-TCs with a Type S reference thermocouple.

VI. REASONS FOR OPEN CIRCUIT FAILURES

Unlike the out-of-pile test undertaken in 2005 [7], many of the thermocouples experienced open circuit failure, but most of these may be attributed to suspected gas impurities during the 1300°C portion of the test as well as a small temperature excursion above 1300°C (test actually ran at 1311°C for a few days), which drove the Inconel 600 sheathed thermocouples very near to their melting points. In hindsight, the 1300°C portion of the test provided little additional insight as nearly all of the Inconel-sheathed thermocouples either completely melted or became so soft as to sag and fall apart. The Cambridge special sheath thermocouples continued to provide signals through to the end of the test, but their niobium protection sleeves were virtually destroyed (see Fig. 19 “CMB-A”). The molybdenum-sheathed thermocouples appeared nearly unaffected (see Fig. 19 “HRDAL-C”) but upon further examination were brittle with virtually nil ductility. The niobium-sheathed HTIR-TCs were essentially destroyed by oxygen impurities (see Fig. 19 “NB04”).

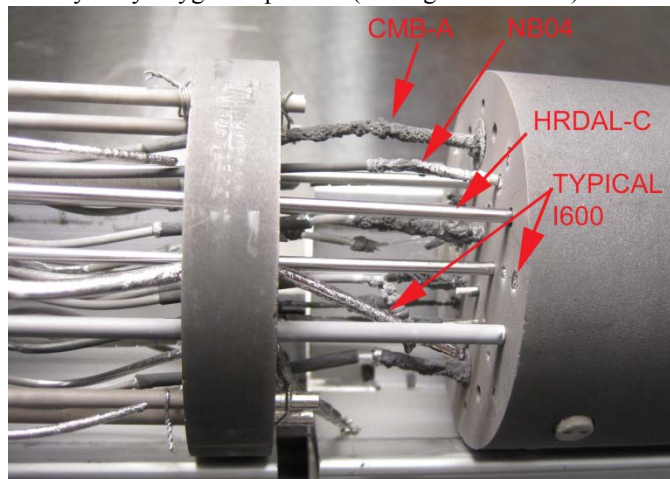


Fig. 19. Test assembly after completion of all heating cycles.

During the entire test program, outlet gas oxygen readings were off-scale low (less than 1.0 ppb). After the fact, investigations on the empty furnace at room temperature revealed that the inlet gas had an O₂ concentration of about 2 ppm, and the outlet gas ran as high as 18 ppm O₂, possibly due to a small furnace leak. Apparently, the high temperature metals and graphite gettered the oxygen before the gas stream left the furnace, resulting in the very low oxygen readings on the outlet gas when the furnace was at temperature. It is believed that these higher oxygen levels were associated with the latter portion of the test program because the thermocouple sheaths showed only moderate signs of degradation during the 1150°C phase (see Fig. 4).

The suspected oxygen contamination of the furnace purge gas was not altogether negative. As discussed in Section II.B, the gases used to control high temperature reactor experiments typically have a slight oxygen impurity, and when integrated over 10,000 hours or more, refractory materials will show signs of attack. It was evident that the molybdenum-sheathed thermocouples performed much better than the niobium sheaths, which leads to the conclusion that molybdenum may be a better protective sleeve material than niobium.

VII. CONCLUSION

Thermocouples for the final AGR fuel experiment, denoted AGR-5/6/7, will be required to measure temperatures higher than any of the previous three AGR fuel experiments. Based on the experience from these three previous experiments and previous out-of-pile testing, a new furnace test was planned and conducted with a variety of Type N thermocouple designs as well as the INL-developed HTIR-TC design. The variations on the standard Type N design were as follows:

1. Increase in size from 1.57 mm OD to 2.34 mm OD
2. Substitute Haynes 214 alloy sheath for Inconel 600
3. Substitute spinel insulation (MgAl₂O₄) for MgO insulation
4. Substitute special sheath material developed at Cambridge University for Inconel 600
5. Incorporate a loose assembly design with hard-fired alumina insulators and molybdenum sheath.

The first three variations produced only a limited improvement in drift rate versus standard 1.57 mm Type N

thermocouples. The special sheath material developed at Cambridge University reduced the drift rate markedly, and this thermocouple design offers the advantage of being just as flexible and physically robust as a standard Type N thermocouple. This Cambridge design appears well suited to operating in the 1000°C–1200°C range (and perhaps higher). The loose assembly design shows promise for being drift resistant but needs further development to prevent open circuit failure when cycled repeatedly. The HTIR-TC design again demonstrated that it exhibits low drift; however, the particular batch supplied for this test suffered from calibration inaccuracy, and an early open circuit failure. Another furnace test is being commissioned to test variations on the HTIR-TC design.

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