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**MECHANICAL RELIABILITY AND THERMOELECTRICAL
STABILITY OF NOBLE-METAL THERMOCOUPLES AT 2600°F
TEMPERATURE AND DOSE RATES UP TO 10^{20} Nvt**

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FOREWORD

The test data and information presented herein were obtained by the writer, A. C. Dallman, while he was employed as a task leader by the Sensor Control Unit of the General Electric Company - Aircraft Nuclear Propulsion Department (GE-ANPD), Cincinnati 15, Ohio. Testing was accomplished under AEC contract AT(11-1)-171, Air Force Contract AF33(038)-21102, and Air Force Contract AF33(600)-38062. The GE-ANPD contracts were cancelled in April 1961, and the original text of this report was written after the author terminated his employment with the General Electric Company in May 1961. The initial draft of this report, presented to GE for editing and publishing in January 1962, was published by the Nuclear Materials and Propulsion Operation, Flight Propulsion Laboratory Department of GE as Informal Report GEMP-183. GEMP-183 was titled "Calibration Constancy of Noble-Metal Thermocouples Under High Temperature and Nuclear Radiation Exposure"; it was published 26 November 1962 and released March 1963.

The material is now being published as a Technical Documentary Report by the Research and Technology Division, Wright-Patterson Air Force Base, for the following reasons: To incorporate several corrections and to update the report wherever possible; to expand the Introduction; and to include several pages of recommendations.

The author wishes to thank Messrs. W. E. Niemuth and T. J. Hendricks. Mr. Niemuth, of the General Electric Nuclear Materials and Propulsion Operation edited the original report and secured its publication as the GEMP-183 report. Mr. Hendricks implemented a program for recalibrating thermocouples after they had been exposed to a high neutron flux level and had undergone thermal exposure. Under Mr. Hendrick's direction, a furnace facility, consisting of sheath stripping equipment, temperature controls, and a platinum resistance furnace, was installed inside a hot cell at the General Electric Idaho Test Site, and a technique for performing the recalibration operation remotely (by slave manipulator) was made operational. Mr. Hendricks is now employed by Edgerton, Germeshausen & Greer, Inc.

ABSTRACT

The initial purpose of this developmental effort was to obtain mechanical integrity from two-conductor platinum rhodium sheath-type thermocouples when such thermocouples are exposed to nuclear radiation at temperature levels of 2500° to 2600° F. The final phase of this program was to prevent unstable calibration of thermocouples during in-pile tests.

Predicted values of surface temperatures, calculated by careful analysis of heat-balance data (before evaluation facilities were available to recalibrate irradiated thermocouples), were determined to be several hundred degrees over measured values. After recalibration facilities became operational, the predicted values were determined to be several hundred degrees higher than measured values. Decalibration data obtained during the ICR-25 reactor test disclosed that the decalibration values ranged from -50° to -1450° F for identical, two-conductor, sheath-type MgO-insulated thermocouples that had been subjected to exposure conditions of 2600° F for 456 hours.

Reactor tests during the test program demonstrated that the following were accomplished:

1. Alumina Flame spray-coating of sheath-type noble-metal thermocouples prevented excessive loss of the noble-metal sheath and sensor wire by vaporization and redeposition on cooler surfaces in reactor tests at operating temperatures of 2500° F to 2600° F;
2. The use of an alumina-coated, noble-metal, bare-wire thermocouple (Span thermocouple) employing a controlled, maximum, feasible separation between sensor wire and/or an alumina-coated thermocouple constructed of two one-conductor coaxial, sheathed thermoelements (Duax thermocouple) insured that temperature information, which will not be affected by significant decalibration, can be obtained for a period of 1000 hours at a test temperature of 2600° F.

Publication of this technical documentary report does not constitute Air Force approval of the report's findings or conclusions. It is published only for the exchange and stimulation of ideas.

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ABBREVIATIONS AND DEFINITIONS

The following abbreviations and definitions are used in this report:

GE	The General Electric Company
TC	thermocouple
emf	thermoelectric potential
Pt	platinum
Rh	rhodium
10	pt - 10% Rh (10 Weight % Rh)
20	pt - 20% Rh (20 Weight % Rh)
40	pt - 40% Rh (40 Weight % Rh)
mil	.001 inch
ANPD	Aircraft Nuclear Propulsion Department of GE
MTR	Materials Test Reactor at National Reactor Testing Station, Idaho
ETR	Engineering Test Reactor at National Reactor Testing Station, Idaho
ORNL	Oak Ridge National Laboratories, Tennessee
ORR	Oak Ridge Research Reactor at ORNL
LITR	Low Intensity Test Reactor at ORNL
MgO	Magnesium Oxide
AL ₂ O ₃	Aluminum Oxide, or Alumina
ITS	Idaho Test Station of GE
RML	Radioactive Materials Laboratory at ITS
HTRE	heat transfer reactor experiment at ITS

ABBREVIATIONS AND DEFINITIONS (Cont'd)

ICR-X	ceramic sample No. X for RT-1 test hole in MTR (X indicates sample number)
99CR-X	ceramic sample No. X for 99 test hole in ETR (X indicates sample number)
ORC-X	ceramic sample No. X for ORR test hole (X indicates sample number)

INTRODUCTION

Noble-metal thermocouples are admittedly a dull subject because they were standardized and apparently perfected many years ago. Now, however, if one were dependent on them for controlling the operating temperature of a nuclear fuel-element performance test where the required temperature was 2600° F and an overshoot of 200° F would be disastrous to the test program, the discovery of a negative calibration vulnerability of 20 to 50 percent would quickly dispel any apathy one might have had for the reliability of these sensors. Since two-conductor, noble-metal, sheath-type thermocouples must operate in nuclear environments at high temperatures, they must be reliable mechanically and calibrations must be stable under the nuclear-environment conditions.

Exposure of two-conductor noble-metal sheath-type thermocouples to a nuclear environment is known to cause some decalibration to occur because of the transmutation of rhodium to palladium; however, the amount of error attributed to this cause does not reach serious proportions in dose rates up to 10^{20} nvt. The major factors governing mechanical integrity are electrical continuity and reliability of insulation.

The initial purpose of the work reported herein was to insure mechanical integrity of two-conductor platinum-rhodium, sheath-type thermocouples when such sensors were exposed to nuclear radiation at temperature levels of 2500° to 2600° F. The final phase of the program, which was destined to be one of major import, was to devise some means of preventing calibration instability of the thermocouple during in-pile tests. To achieve the purposes of the investigation, we required that a thermocouple be designed that would be mechanically reliable and thermoelectrically stable for 1000 hours at a temperature level of 2500° to 2700° F. In addition, the thermocouple was required to operate under temperature cycling conditions in a nuclear environment having dose rates up to 10^{20} nvt.

The developmental work on the desired thermocouples was accomplished as part of the now defunct Aircraft Nuclear Propulsion Development Program, operated by the General Electric Company at Cincinnati, Ohio. Figure 1, taken at the GE-Idaho Test Site installation at the National Reactor Test Station, shows the last full-scale nuclear power plant built by GE-ANPD. This was also the first A/C nuclear power plant to use a horizontally oriented core and the last of a series of HTRE power plants for which the temperature sensors described in this report were developed.

The thermocouple (TC) tests described in this report were conducted in test reactor facilities designed specifically for component testing. Four test reactor facilities were available to GE for testing components; however, all of these developmental tests were conducted in the Oak Ridge Research Reactor (ORR) in Tennessee and in the Materials Test Reactor (MTR) in Idaho. In nearly all cases, tests of thermocouples were conducted concurrently with ceramic fuel-element tests. A cross sectional view of a beryllium oxide (BeO) fuel element, composed of BeO tubes and rods, is shown in Figure 2. The first tier of six hexagonal members around the center rod contained hollow cylindrical centers, and these coaxial tubes were composite mixtures of BeO and nuclear fuel (U_2O_3). The outermost tier of BeO rods (shown as cross-hatched areas in Figure 2) and the BeO center rod were used for routing the thermocouple sensors. In fact, fuel elements were the major test article and, consequently, the number of experimental TC's that could be installed on a given sample was limited, since those installed could not be used for sample temperature control.

This report covers some of the operating experiences that led to the development of thermocouples that could withstand exposure to a high neutron flux level and a high temperature air flow, simultaneously. The test environment was achieved by installing the experimental thermocouples on ceramic fuel-element test samples. These samples were tested in the MTR and ORR test holes that had been leased by the General Electric Aircraft Nuclear Propulsion Department. Developmental tests, which were conducted in furnace facilities in a laboratory, are reported in reports XDC 60-12-28 (Reference 1) and APEX 746 (Reference 2) published by GE.

Figure 1. HTRE-3 Full-Scale Power Plant for Ground Test Experiments

Figure 2. Cross Section of Ceramic Nuclear Fuel Element

DESCRIPTION OF TESTS

As reported in Reference 1, the durability and mechanical reliability of platinum-rhodium thermocouples (both sheath and solid wire types) are improved considerably by applying a flame-sprayed coating of high-purity alumina to the hot zone portion of the thermocouple when it is to be operated in temperature ranges above 2300° F. The alumina coating process is performed basically as follows:

1. Degrease the thermocouple by using trichlorethylene solvent.
2. Uniformly roughen the surface to be flame sprayed by grit blasting with Metcolite F*, or equal.
3. Again degrease the thermocouple as in Step 1.
4. Flame spray (with Coors AD-100, McDanel AP-35, (or equal quality) alumina rod to a thickness of 2-1/2 to 3 mils by using a Metallizing Co. of America, Type R-2 metallizing gun (or equal). Give careful attention to details to assure satisfactory results.

Sheath-type, 55-mil, two-wire, Pt-Rh(0-10) thermocouples were flame sprayed for in-pile test-sample installation in August 1960. Two of the coated TC's were tested for performance, and, on the basis of the test results, were considered satisfactory for installation in the hot zone of the ORC-4 sample. Both TC's were subjected to thermal shock tests and were tested to determine the minimum bend radius which could be tolerated before the ceramic coating began to crack or scale off. In the thermal shock tests, the temperature of the TC's was repeatedly dropped from 2600° F to room temperature in a few seconds. Twenty-four temperature cycling tests were conducted, and a 3/4-inch bend radius was achieved on each sample without incipient cracking of the alumina coating. The ORC-4 sample accumulated 600 hours of test time in the range of 2500° to 2600° F with no evidence of mechanical failure of the alumina-coated sheath-type TC's.

Figure 3 shows the center instrumentation rod of the ORC-4 sample after removal of the outer tier of fueled rods and very conclusively illustrates the value of alumina coating to prevent mechanical loss of TC's.

(a) Arrow 1 points to the sensor end of one of the alumina-coated thermocouples (TC #1) in the upper slot of the instrumented rod.

(b) Arrow 2 points to the sensor end of the other alumina-coated thermocouple (TC #2) in the lower slot of the center rod.

(c) In the same slot with, but underneath and beyond the end of TC #1, are the remains of an uncoated noble-metal sheath-type thermocouple (Arrow 3). Sheath material has been lost by the continuous surface vaporization of the platinum and rhodium metals. In the case of platinum such vaporization is associated with the formation of a volatile oxide (Reference 3) that begins to form at 1112° F (600° C) and disassociates at 2040° F (1115° C). A portion of the vaporized material has redeposited at the downstream end of the ceramic rod. Only the sensor wires and some of the MgO insulation remain, however, the thermocouple maintained electrical continuity.

*See Appendix I

(d) Two hard-fired alumina-insulated thermocouples are shown just above the instrumentation (Arrow 4) rod as they appeared after being pulled away from the rod. It can be seen that the entire sensor bead of each of these thermocouples has been lost due to vaporization of the thermocouple material.

Hard-fired alumina-insulated and unprotected platinum-rhodium sheath TC's were the types commonly used in all in-pile test samples prior to the initial installation of alumina-coated sheath thermocouples on sample ORC-4. Figure 3 convincingly demonstrates that the alumina-coated thermocouples have decisively better resistance to deterioration than the hard-fired alumina-insulated, or the uncoated sheath types.

After remote visual examination of the sample instrumentation, one of the coated thermocouples was stripped of its alumina coating so that the surface of the platinum alloy sheath could be inspected. There was no evidence of corrosion or deterioration of the metal surface. Additional tests of ceramic fuel elements (ORC 5, 6, and 7, MTR 1CR-24, 1CR-25, 1CR-26, and ETR 99CR-20) confirmed that flame-spray coating the platinum-rhodium TC's with alumina completely eliminated the serious loss of TC material due to vaporization of noble metals.

Prior to the existence of a post-irradiation-test recalibration facility, heat-transfer calculations indicated that the magnitude of decalibration was serious. Therefore, starting with tests ORC-4 and 1CR-23, corrections based on heat-transfer calculations were applied to TC data as a means of adjusting for decalibration. The heat-balance calculations gave expected surface temperatures for various locations in the test cartridge. These temperatures were correlated with initial "on test" readings of the TC's before decalibration had occurred. With continuing irradiation, there were changes in the relative values of temperatures obtained from the TC's, and these values, when compared to calculated temperatures, strongly indicated that decalibration had occurred. The formula used for the heat-balance calculations is given in Appendix II.

GE, in Reference 2, established that noble-metal volatilization, particularly that of rhodium, has produced detrimental thermoelectric effects in the 2500° to 2600° F range. Because of these effects, plans were made to determine how much decalibration was occurring during long-time reactor tests. This meant the development of a technique for recalibrating the TC after it had been exposed to both high neutron flux and high temperature, and the development of a facility for accomplishing this specialized recalibration work.

The technique developed for the program used a slave manipulator to perform the TC recalibration. The facility, consisting of sheath-stripping equipment, temperature controls, and a platinum furnace, was installed inside a hot cell of the RML at ITS.

MTR sample 1CR-25, instrumented entirely with uncoated sheath-type 0-10 55-mil outer diameter (O.D.) TC's, was the first sample for which facilities were available to accomplish recalibration of the TC immediately after post-test examination of the sample. On this sample, a considerable number of splice failures occurred, which resulted in shorting the sensor wires to the stainless steel splice protector. Therefore, in most instances, the TC was checked with the splice in place and the splice removed, especially in cases where a bad splice was suspected. This was done to prevent the splice condition from affecting the actual recalibration values.

Results of 1CR-25 TC recalibrations (tabulated in Table I) showed that all retestable TC's decalibrated excessively, and, in all cases, the decalibration varied from -150° to -1450° F in zones 3 and 4 (the hottest zones) where temperatures ranged from 2440° to 2610° F. The

recalibration temperature was about 2450° F. The exit air TC's had an indicated decalibration range of -130 to -170° F when recalibrated in their operating temperature range of approximately 1925° F.

The indicated magnitude of decalibration, shown in Table 1, was different for each TC and was often much greater than predicted values obtained by the use of thermodynamic calculations of the test conditions. The greater-than-nominal decalibration results obtained from exit-air TC's were attributed to exposure of their leads to high temperatures which caused rhodium mass transfer (References 1 and 5) and resulted in a change of emf output of TC's. The high temperatures at the lead wires were corrected by rerouting leads to the exit-air junctions and by using improved TC's on future samples.

The length of the temperature profile in the furnace used for recalibration was different from the length of the profile present during in-pile operation, and in-pile samples never maintained a constant temperature profile in operation. Consequently, according to information contained in Reference 1, if exposed thermocouples could not be retested in precisely the same temperature profile as existed during the recalibration process, measured errors of excessive magnitude indicate that serious decalibration has occurred, but did not accurately establish the magnitude of decalibration for each thermocouple. Decalibration data from post-irradiation evaluations did, however, show the same range of values as similar data taken under laboratory conditions for non-irradiated thermocouples.

Table 2 contains data from Table 45 of Reference 1 for TC specimens that were tested under laboratory-immersion test conditions for 542 hours at 2600° F. These data disclose that decalibration values ranged from -11° to -1063° F when MgO was used for the insulation material. Similar decalibration values were obtained during the ICR-25 reactor test (Table 1) in which the decalibration values ranged from -130° to -1450° F for identical, two-conductor, sheath-type, MgO-insulated TC's subjected to exposure conditions of 2600° F for 456 hours. A comparison of these two sets of data indicates that a constant-temperature-profile-recalibration of the test TC's which had been exposed to a nuclear reactor environment provides error data that closely approximates the error data obtained when TC specimens are tested under laboratory test conditions.

Additional calibration instability of nonmetal sheath-type TC's occurred when other instrumented samples were tested in the 2300° to 2700° F range. The decalibration data taken during the course of this work are presented in Tables 3, 4, and 5. The temperature listed in the tables is the temperature of the ceramic fuel in a limited area. The TC's were located so that they would measure the test-sample temperature profile as well as the exit air, consequently, not all of the TC's measured the maximum temperature shown in the tables.

- (a) Table 3 contains data from MTR sample 1 CR-23, which operated at test conditions of 2600° F for 212 hours.
- (b) Table 4 contains data from MTR test sample 1CR-24, which operated for 501 hours at a test temperature of 2600° F.
- (c) Table 5 contains data from ORC samples 4 and 5. Two of these TC's were alumina-coated and three were uncoated. ORC-4 test time was 600 hours at 2600° F, and ORC-5 test time was 920 hours at 2600° F.

Tables 1 through 4 contain data from unprotected sheath-type 0-10 55-mil O.D. TC's. Table 5 contains data from both alumina-coated and unprotected sheath-type 0-10 TC's that were removed from the test samples ORC 4 and 5, as noted above.

TABLE 1

POST-TEST RECALIBRATION DATA OF RETESTABLE THERMOCOUPLES
FROM MTR SAMPLE ICR-25

TEMPERATURE: 2600° F, TIME: 456 HOURS

(All Thermocouples Were Unprotected Sheath-Type 0-10 55-mil O.D.)

TC No.	Location (Zone No.)	Temperature of Standard TC (° F)	Calibration at End of Test (° F)	Decalibration Deviation (° F)
1	4	2450	1000	-1450
4	4	2440	1440	-1000
6	3	2450	1225	-1225
7	3	2450	1750	-700
9	3	2430	1480	-950
10	3	2450	1865	-585
11	3	2440	1750	-690
12	3	2445	1985	-460
13	3	2450	1910	-540
14	3	2450	2325	-125
17	Exit Air	1770	1070	-700
19		2450	1870	-580
22		2440	2115	-325
23		2470	2230	-240
23		1930	1760	-170
24		2440	2270	-170
24		1930	1800	-130
25		2420	2250	-170
25		1950	1820	-130
26		2460	2240	-220
26		1940	1800	-140

NOTE

1. TC's 23, 24, 25, and 26 were recalibrated at surface temperature and at exit air temperature, therefore two readings are shown.
2. Temperatures in location 3 ranged from 2520° to 2610° F; in location 4, from 2440° to 2570° F. Exit air temperatures were about 1850° to 1900° F.

TABLE 2

THERMOCOUPLE SPECIMENS TESTED UNDER LABORATORY IMMERSION
 TEST CONDITIONS FOR 542 HOURS AT 2600° F
 (All Thermocouples Were Unprotected 55-mil Sheath-Type)

TC Number	Specimen Composition			Decalibration Deviation (° F)	Standard TC # S 21 (° F)
	% Rh Negative Leg	% Rh Positive Leg	Insulation Material		
1	0 ↓	10 ↓	MgO ↓	-29	2595
2				-45	2596
3				-19	2606
4				-266	2600
5				-425	2596
6				-21	2599
7				-11	2598
8			BeO ↓	-144	2608
9			Al ₂ O ₃ ↓	-95	2602
10				-615	2601
11			MgO ↓	-678	2602
12				-597	2600
13			Al ₂ O ₃ ↓	-75	2610
14				-60	2602
15				-457	2608
16				-270	2600
17				-239	2608
18				-291	2603
19				-205	2607
20	6 ↓	30 ↓	Al ₂ O ₃ ↓	-634	2596
21				-552	2599
22			MgO ↓	-2130	2592
23				-1063	2594
24			Al ₂ O ₃ ↓	-241	2597
25			Al ₂ O ₃ ↓	-1244	2598
26				-1260	2597
27	0 ↓	10 ↓	MgO ↓	-806	2604
28				-150	2606
29				-116	2604

NOTE: This data was obtained from Reference 1, Table 45

TABLE 3

POST-TEST RECALIBRATION DATA OF RETESTABLE THERMOCOUPLES
 FROM MTR SAMPLE ICR-23
 TEMPERATURE: 2600° F, TIME: 212 HOURS
 (All Thermocouples Were Unprotected Sheath-Type 0-10, 55-mil O.D.)

TC Number	Location (Zone No.)	Temperature of Standard TC (° F)	Calibration at End of Test (° F)	Decalibration Deviation (° F)
2	4	2450	2425	-25
5	4	2446	2421	-25
8	3	2455	2430	-25
11	3	2450	2435	-15
12	3	2455	2335	-120
12	3	2455	2330	-125
13	3	2465	2150	-315
14	3	2435	2405	-30
15	4	2445	2425	-20

NOTE: Temperatures in location 3 ranged from 2520° to 2610° F; in location 4, temperatures ranged from 2440° to 2570° F.

TABLE 4

POST-TEST RECALIBRATION DATA OF RETESTABLE THERMOCOUPLES
 FROM MTR SAMPLE 1CR-24
 TEMPERATURE: 2600° F, TIME: 501 HOURS
 (All Thermocouples Were Unprotected Sheath-Type 0-10 55-mil O.D.)

TC Number	Location (Zone No.)	Temperature of Standard TC (° F)	Calibration at End of Test (° F)	Decalibration Deviation (° F)
19	Exit Air	2465	2460	-5
20	" "	2465	2430	-35
13	3	2480	2475	-5
11	2	2475	2455	-20
14	3	2485	2445	-40
15	3	2455	2445	-10
8	2	2485	2485	0
16	4	2465	2460	-5
10	3	2460	2374	-86

NOTE: Temperatures in location 3 ranged from 2520° to 2610° F; temperatures in location 4 ranged from 2440° to 2570° F.

TABLE 5

POST-TEST RECALIBRATION DATA OF RETESTABLE THERMOCOUPLES
FROM ORC SAMPLES 4 AND 5 -- TEMPERATURE: 2600° F

ORC Sample 4 Test Time: 600 Hours ORC Sample 5 Test Time: 920 Hours

ORC Sample Number	Location	Temperature of Standard TC (° F)	Calibration at End of Test (° F)	Decalibration (° F)	Sheath
4	1C	2465	2395	-70	Alumina coated
5	1C	2445	2340	-105	"
4	1B	2460	2420	-40	Unprotected Sheath
5	1B	2450	2390	-60	"
5	2B	2440	2300	-140	"

GE report (Reference 2) contains data which indicate that the threshold temperature of decalibration is very sensitive to the pack density of the insulation used in the construction of a sheath-type TC. It was observed that in most cases granular MgO is a better insulating material than granular Al_2O_3 . This is attributed to the better pack-density properties of MgO (especially after long exposure to high temperature) and is indicated in Table 2. Table 2 shows that in the tests seven Al_2O_3 -insulated sheath-type TC's decalibrated from -552° to -2130°F , while nineteen MgO-insulated sheath-type TC's decalibrated from -11° to -1063°F . A comparison of the data listed in Table 1 with the data in Tables 4 and 5 shows wide discrepancies in the amount of measured decalibration of TC's from these tests, but all TC's listed in these tables were MgO-insulated. These wide variations in total decalibration values were believed to be caused by corresponding wide variations in pack density of the MgO insulation material, as reported in Reference 2.

The data in Table 5 show that, while the alumina coating of sheath-type TC's is very effective in extending the mechanical life of the TC's, the coating has little effect on the calibration stability. Decalibration results principally because of vaporization and mass transfer of rhodium from the platinum-rhodium sheath and thermoelement to the pure platinum thermoelement.

To reduce the decalibration of noble-metal TC's, GE made installations that utilized alumina flame-sprayed on bare sensor wires and a maximum feasible separation of the two types of thermoelements. The construction of these "Span" TC's is shown in Figure 4. Figure 5 shows a typical routing scheme which maintains the separation of the sensor wires. In Figure 6, the separation between sensor elements that exists in a 55-mil O.D. sheath-type TC and the separation that can be obtained in a typical installation of a Span TC on a BeO instrumentation rod are compared. The ultimate design incorporating this technique for preventing mass transfer of rhodium is a butt-welded TC which has the sensor wires leaving the junction in opposite directions and uses the alumina flame-spray coating.

Four 13-mil butt-welded Span TC's were installed in the hot zones of test sample ORC-6. This sample was irradiated for 500 hours at 2000°F and then irradiated for 400 hours at a nominal 2600°F . During the latter period, surface-temperature readings from TC's in the hot zones ranged from 2530° to 2600°F . Unfortunately, the irradiation cartridge was designed so that the splices of sensor wire to lead wire were stressed during the longitudinal motions of the sample that were necessitated by reactor refueling operations. The stress caused loss of lead wire continuity and consequent excessive TC failure. One of the hot-zone Span TC's failed after 495 hours, a second failed after 566 hours; however, the remaining two hot-zone TC's operated without serious decalibration for the life of the test. One 20-mil Duax TC was installed in the hot zone of the ORC-6 sample, and it failed after 225 hours of reactor operation at 2600°F . See Figure 7 for a Duax TC description.

Design changes were made to improve the reliability of the splices in the next irradiation cartridge, ORC-7. The sample was instrumented with seven 20-mil alumina-coated Span TC's and two 38-mil alumina-coated Duax TC's to monitor hot-zone temperatures. Both Duax TC's and six of the seven Span TC's maintained electrical continuity during the entire 1000-hour test period at a 2600°F nominal surface temperature for the fuel elements. Figure 8 is a plot of TC readings taken during the test period.

The heat balance in the test sample was carefully and continuously analyzed during the testing period to determine whether serious decalibration was occurring. The heat-balance data from coolant flow and temperature rise enabled testing personnel to predict the expected surface temperature. Figure 8 shows a comparison of measured values for two of

the TC's with values of expected surface temperature obtained from the analysis. The data of Figure 9 shows that little, if any, decalibration of hot-zone TC's was evidenced in the test period. Figure 10 shows the predicted longitudinal temperature shift due to reactor flux shift upon refueling. Figure 11 shows measured temperatures before and after refueling. The agreement here, with all TC's following the predicted shift, shown by Figure 10, is quite unusual based on experience from previous tests and indicates that TC behavior for this test was by far the best ever achieved. In earlier tests, predicted values of surface temperature obtained by this same method generally increased several hundred degrees Fahrenheit over measured values. This strongly indicated decalibration of the TC. Therefore, the instrumentation performance obtained in the ORC-7 test was of decisive value, since this was the first 1000-hour test that was conducted without the handicap of serious TC decalibration.

In further reactor tests, the samples were instrumented with both Span and Duax TC's, however, the tests were terminated short of the 1000 hours of test ORC 7. Eventually, reactor testing was ended by termination of the Aircraft Nuclear Propulsion program and, consequently, the ORC 7 test is the only example of long-term, high-temperature performance of these thermocouple types. Although the data are somewhat sparse, it is believed that very significant improvement has been attained for TC performance in reactor environment.

Figure 3. Instrumented Center BeO Rod From ORC-4 Cartridge, After Test

Figure 4. Cross Section of Span-Type Thermocouple

Figure 5. BeO Hexagonal Rod Instrumented With Span Thermocouples

Figure 6. Comparison of 55-mil Duplex Thermocouple With 20-mil Span Thermocouple

Figure 7. Cross Section of Duax Thermocouple Thermoelements. The Duax Thermocouple is Constructed of Two One-Conductor Coaxial Sheath Thermoelements. The Sheath of Positive Thermoelement is Made of Same Material As Positive Element, and Sheath of Negative Element is Made of Same Material as the Negative Element. Each Thermoelement is Fused Separately Into a Single Capped Junction and Then the Two Fused Junctions are put Together to Form a Sensor Junction.

Figure 8. Temperature Test History of ORC-7 Test Cartridge. 1000 Hours at 2600°F.

Figure 9. Recorded Vs Predicted Temperatures of ORC-7 Test Cartridge Test

Figure 10. Predicted Temperature Profile

Figure 11. ORC-7 Temperature Profile Data

CONCLUSIONS

Unprotected two-conductor noble-metal sheathed TC's are susceptible to both mechanical loss and serious decalibration in temperature environments above 2300° F. Experimental results obtained from reactor testing show that alumina spray-coating of thermoelements or sheaths provides excellent protection against the vaporization and ultimate deterioration of these elements. Furthermore, the test experience indicates that alumina of very high purity is not required for the coating.

Serious decalibration of TC's can be prevented by the separation of dissimilar sensor wires and sheaths. Both coated Span-type thermocouples and coated Duax thermocouples have been utilized with essentially no decalibration in reactor tests up to 1000 hours at temperatures to 2600° F.

In Figure 12, the ductility of 20-mil Duax and Span TC's is evaluated by comparing the cross sectional areas of the ceramic and metallic materials used in their construction. Sensor elements having identical O.D.'s were compared, and an alumina-coated Duax of the 20-mil O.D. size was found to have a cross sectional area of only 35.5 percent metal. Whereas, a coated 20-mil O.D. Span-type TC had a cross sectional area of 56.5 percent metal. Therefore, the ductility of the Span TC is significantly greater than that of the Duax type, since ceramic materials have very poor ductility, especially at room temperature. Ductility refers to the ability of a material, or structure, to absorb energy inelastically without failure, therefore, the greater the ductility the greater the resistance to failure. This strength advantage appeared obvious during required handling operation preparatory to, and during, flame spraying of the alumina coating on Duax and Span TC's.

In general, there are some instrumentation requirements which are better satisfied by the Duax TC design, while other requirements are better satisfied by the Span design. Therefore, the choice of TC design should be dictated by the instrumentation requirements. The Duax type is probably the more sophisticated of the two designs and is to be preferred when routing space is not seriously limited.

Exposure of two-conductor noble-metal sheathed TC's to a nuclear environment causes some decalibration by transmutation (References 4 and 6) of rhodium to palladium, but the error does not reach significant proportions in dose rates up to 10^{20} nvt. In other words, the major portion of the decalibration encountered was caused by mass transfer of rhodium (Reference 11).

RECOMMENDATIONS

The only multi-stage test cartridge instrumented with Span TC's during the test period covered by this report was a four-stage MTR sample, the ICR-26, which had four Span TC's installed on it. The test temperature for this sample was 2700° F, but it was taken off test after 96 hours of test time due to failure of all the exit air couples, which were all 2-conductor sheath-type TC's. However, all four of the Span TC's were reported operating satisfactorily at the time the sample was taken off test.

To the best of the writer's knowledge, no post-test of TC recalibration data is available for the multi-stage sample from which to ascertain the structural integrity of Span TC when subjected to more critical mechanical load factors peculiar to multi-stage sample tests. However, if it were necessary to use a more rugged TC than the Span type, a combination Span and Duax type TC is recommended. This TC would have coaxial sheath-type sensor wires with the sheath material identical to the sensor wire it surrounds, like the Duax TC, except that the sensor wires would be stripped of their sheath from the sensor junction to a point about 1-1/2 to 1-1/4 inches away from the junction (this area is identical in construction to the Span TC shown in cross section in Figure 3). This is one of several construction techniques which satisfies the basic design objective for a TC which, in an electrical sense, has a fully floating sheath surrounding each sensor wire. Of course, both the bare wire junction of this TC and that portion of the sheath section which is exposed to the hot-zone temperature should be coated with alumina, and the sensor wires to the hot junction must be so routed that they have maximum separation, like a common Span TC. Since in an electrical sense, the sheaths of this combination type TC would be floating independently, the following advantages would be gained:

1. Splices to the lead wires could be made less complex and thereby more reliable, since the sheaths could safely make full electrical contact (or be brazed together) in the splice construction design;
2. Sheaths could be permitted to make physical contact with each other or with sheaths of other TC's without disturbing the emf output of the TC;
3. Cost of the TC material would be less, since commercial grade platinum and platinum-rhodium sheaths could be used;
4. Fabrication work would be less tedious and complex; therefore, overall instrumentation costs would be reduced;
5. The size of the coaxial wire could be set at 33 ± 1 mil O.D. so that a $0.015 + \frac{.001}{-.000}$ inch O.D. sensor wire could be utilized and still provide an adequately rugged TC and be competitive dimensionally with the 55-mil sheath-type Duplex thermocouple;
6. This TC would have application for an installation through the walls of a high-temperature metal-capsule type of sample, when it is desired to braze or weld the sheath to the capsule walls at the point of insertion to provide a sealed assembly;
7. The sensor junction of butt-welded or common Span-type TC's would also be simpler to fabricate;

RECOMMENDATIONS (Cont'd)

8. The mechanical ruggedness (ability to withstand vibration and impact) of the sheath type (Reference 1) TC would be duplicated.

The McDanel* AP-35 alumina flame-spray rod is apparently equal to, or better than, the Coors* AL-100 rod for TC application, since approximately 90 percent of the ORC-7 test TC's were coated with McDanel AP-35 alumina because of a shortage of the recommended material. The good results of the ORC-7 test indicate that the high-purity aluminas, which are more expensive and much more difficult to flame spray, need not be used to protect the noble metal TC's. Since these observations were also made in Reference 2, the lower-purity aluminas are recommended for TC coatings.

The maximum operating potential of the Span-type TC design can be estimated with reasonable accuracy, and it is conservatively estimated to be 3200° F. Since an 80% - 20% rhodium vs 60% platinum - 40% rhodium thermocouple (Reference 7 and 9) is suitable for continuous use at a maximum of 3200° F, it appears feasible to apply the Span TC design to this noble metal TC range and thereby obtain reliable information on operation at 3200° F for 1000 hours. It follows, therefore, that a TC with excellent mechanical integrity and temperature-information reliability of 3200° F maximum operating temperature should be readily available without resorting to exotic TC materials, such as molybdenum and rhenium, which must be protected by an inert atmosphere or a hermetically sealed coating. Additional development tests should be conducted to develop a 3200° F TC employing the same design principles as used for either the Duax or the Span TC. Either flame-sprayed alumina or plasma-sprayed MgO (for the higher temperature ranges) should be satisfactory for use as the insulation and protective coating.

* See Appendix I

Figure 12. Comparison of Ductility of Duax and Span Thermocouples, 20-mil O.D. Size. By Calculating Areas of Metal and Ceramic in the Two Designs, it was Found That the Cross Sectional Area of the Duax is 35.6% Metal; the Span is 56.3% Metal. After Grit Blasting the Outer Surface Preparatory to Alumina Spraying, the Cross Sectional Area of the Duax is 36.7% Ceramic; Whereas the Span is 100% Metal.

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APPENDIX I

COMMERCIAL ITEMS

The commercial items called out in this report are:

Type R-2 Metallizing Gun

Manufacturer: Metallizing Company of America (METCO)
1181 Prospect Avenue
Westbury, New York

Coors AD-100 Flame Spray Rod

Manufacturer: Coors Porcelain Company
600 Ninth Street
Golden, Colorado

McDanel AP-35 Flame Spray Rod

Manufacturer: McDanel Refractory Porcelain Company
Beaver Falls, Pennsylvania

APPENDIX II

HEAT-BALANCE CALCULATION METHOD

The test is controlled on indicated exit air temperature after "on test" conditions are established. The surface temperature (T_s) for any TC location at any time can be calculated from the following relation:

$$T_s = \left(\frac{W}{W_o} \right)^{0.2} \times \left(\frac{T_{so} - T_{in_o}}{T_{ex_o} - T_{in_o}} \right) \times (T_{ex} - T_{in}) + T_{in}$$

where

T_s = surface temperature

T_{in} = inlet air temperature

T_{ex} = exit air temperature

W = air weight flow

Subscript o = indicated values at the "on test" base point.

From this relation, it can be seen that if the exit air temperature corresponding to the hottest fuel channel is held constant and the specified inlet air temperature is maintained, the maximum fuel tube temperature will remain very close to the desired "on test" value.