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TUNGSTEN-RHENIUM ALLOY THERMOCOUPLES AND THEIR USE IN A UO_2 -FUELED REACTOR

by

E. J. Brooks and W. C. Kramer

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I. INTRODUCTION

Since publication of the initial design and test information on the BORAX-V fuel-rod thermocouples, further experimental work has been completed that provides additional performance characteristics of the thermocouples as originally designed.¹ Two new types of thermocouple construction have been tested at temperatures in excess of 2920°C in attempts at improving the initial design.

Measurements of central fuel temperatures in the reactor boiler fuel have been made with three fuel loadings ("B-2" boiling core, "CSH-1" central superheating core, and "PSH-1" peripheral superheating core). Some of these data are presented, along with results of fuel and thermocouple thermal-time-constant measurements and the testing of new experimental thermocouple designs.

II. DESCRIPTION

A. Boiling-fuel Thermocouple Rods for BORAX-V

Fuel-temperature measurements in BORAX-V were normally obtained from instrumented fuel assemblies containing five thermocouple rods per assembly. Provisions were also made for installation of thermocouple rods in place of standard fuel rods at any core location. This allowed temperature measurements without use of fully instrumented fuel assemblies. Construction of the thermocouple rods is shown in Fig. 1. Annular pellets were used to allow entrance of the thermocouple down the axis of the rod. Hole depth was varied by loading standard, cylindrical, fuel pellets accordingly. The hollow section in the top of the thermocouple rod was used to provide a fission-gas expansion space and to allow room for a thermocouple transition joint. The thermocouple rod was vacuum-baked and back-filled with helium at atmospheric pressure, and the final thermocouple seal at the top of the rod was made by induction-brazing with Nicro* brazing alloy (82% Au, 18% Ni).

*Trade name of Western Gold and Platinum Company.

ROD TYPE	DIM. A
V	30.48 cm
W	27.94 cm
X	25.40 cm
Y	22.86 cm
Z	20.32 cm

SECTION THRU ROD

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Fig. 1. Boiling-fuel Thermocouple Rod; BORAX-V

B. Thermocouple Construction

The standard thermocouples used throughout the BORAX-V in-vessel instrument program were of metal-sheathed, mineral-insulated construction. A compromise in size was necessary in the design of the thermocouples. It was desirable to have a small hole to minimize fuel loss, but the practical minimum size for sheath fabrication was found to be a nominal 0.160-cm OD. The thermocouples were constructed of two groups of materials to fit the environments inside the thermocouple-rod and the boiling-water regions of the reactor, respectively. The fission-gas expansion space provided a region in which to make the transition between materials. The thermocouple below this point consisted of W/W-26w/oRe conductors in BeO insulation and a tantalum sheath; above this point were conductors of Alloys 200* and 226* in Al_2O_3 insulation and Type 304 stainless-steel sheath. The transition joint consisted of an insulated junction and a mechanical joint (modified miniature tubing union). The union pieces were brazed after assembly to ensure a leak-proof joint.

C. Installation in Reactor

Completed thermocouple rods were installed in place of standard fuel rods in boiling-fuel assemblies similar to that shown in Fig. 2. In such an installation, the thermocouple rods were individually removable units. The other method of installation was as integral parts of instrumented boiling-fuel assemblies, where they were combined with other thermocouples and flowmeters to form removable instrumented fuel assemblies. An end view of such an assembly is seen in Fig. 3. The five fuel thermocouple leads can be seen in this view adjacent to the removable exit flowmeter.

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Fig. 2. Fuel Rod and Cutaway Section of Boiling-fuel Assembly; BORAX-V

ID-103-3607

Fig. 3. Instrumented Boiling-fuel Assembly; BORAX-V

*Trade names of Hoskins Manufacturing Company

III. EXPERIMENTAL MEASUREMENTS FOR THE BORAX-V PROJECT

A. Boiling-fuel Temperatures, In-reactor

The thermocouples described above have been used for measuring the UO_2 fuel temperatures during reactor power operation with three separate fuel loadings in the reactor. The first period of power operation with an all-boiling-fuel core designated as "core B-2" produced fuel temperatures up to 988°C . The temperature record of any thermocouple as a function of power appeared to be quite unpredictable, but related, somehow, to fuel burnup. Records of two representative thermocouples are shown

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Fig. 4. Boiling-fuel Temperature vs Cumulative Reactor Energy at Fixed Reactor Power Levels, Core B-2; BORAX-V

in Fig. 4. The indicated temperatures suggest early effects of thermal and/or irradiation damage to the fuel thermal conductivity.

Similar temperature records obtained during short-term power operation of the reactor with a central superheating core installed (designated as core CSH-1) showed little of the effects observed on the earlier core loading (see Figs. 5 and 6). Total fuel burnup in core CSH-1 was about $1/30$ that experienced in the B-2 core. The integrated neutron flux received by the thermocouples in either core loading was less than approximately 3.5×10^{18} nvt. No

calibration changes of any magnitude, on this account, were expected to be present, and the temperature records were expected to have an accuracy of 1.1 to 3.5% of indicated value, based primarily on the manufacturer's published data.^{2,3} Figures 8 through 10 of "Temperature, Its Measurement and Control in Science and Industry"³ indicate a perceptible transmutation change in composition of the W/W-26w/oRe thermocouple at an integrated thermal flux of about 10^{20} nvt. Displacement effects in the crystal structure are felt to be negligible and easily annealed out. The range of measurement accuracy, based on the manufacturer's published calibration standards,² was a result of the guarantee of $\pm 1\%$ of temperature over the range of 427 to 2315°C for the W/W-26w/oRe thermocouple wire, and a maximum error of 0.14 mV due to the extension wire-thermocouple wire junctions. The maximum combined error of 3.5% of temperature occurs at 427°C , and the minimum of 1.1% error at 2315°C .

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Fig. 5. Indicated UO₂ Temperature vs Reactor Thermal Power, Boiling-fuel Thermocouple
Rods Nos. ZA-17, -18, -19, and -20, Core CSH-1B; BORAX-V

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Fig. 6. Indicated UO_2 Temperature vs Reactor Thermal Power, Boiling-fuel Thermocouple RocNo. YA-15, Core CSH-1B; BORAX-V

Operation with core PSH-1 (peripheral-superheater core configuration) provided a great deal more data on boiler fuel temperatures. Representative data are presented in Figs. 7, 8, and 9. The trend, again, was toward large indicated temperature changes in the fuel as a function of burnup.

In an attempt to verify the accuracy of thermocouples, two independent experiments were carried out on unirradiated thermocouple rods and thermocouples. One test was planned to measure the transient response of the fuel-thermocouple combination to a rapid temperature change both before and after irradiation. (Cracking of the fuel due to irradiation, thermal shock, etc., would theoretically lead to reduced thermal conductance of the fuel and jacket, and hence a time constant of larger value.) Pre- and post-irradiation thermal-transient measurements were made to evaluate fuel changes.

A second test to determine radiation damage (calibration change) to the thermocouple, involved irradiation in the EBR-I and BORAX-V reactors and hot-cell temperature calibrations.

Both groups of tests are described below.

B. Determination of Thermocouple-rod Thermal Time Constants

1. Tests on New Thermocouple Rods

The first experiments on new thermocouple rods were performed out-of-reactor and were made to establish the effective overall thermal time constant of the thermocouple-fuel system. These tests were conducted on three thermocouple rods by rapid insertion of each rod into a bath of heated NaK. Steps from room temperature to 260°C, and 454 to 260°C (nominal temperatures) were made in an argon atmosphere, and data were obtained for a total of nine "step tests." Signals from the internal thermocouple rod and a temporary external-jacket thermocouple were monitored by an oscillograph. The 63.2% time constants were determined from these recordings. Data are presented in Table I.⁴

The above data provide a representative indication of thermocouple output response to a thermal transient applied to the exterior of the thermocouple rod. To determine the time constant of the thermocouple in its environment (similar to a conventional thermocouple in its protecting well), and hence be able to calculate the true thermal time constant of the fuel-and-jacket combination, another series of transient tests was made. These tests were made by heating the thermocouple rods with a neutron pulse in the TREAT reactor. Reactor pulses with initial periods of 130 and 260 msec were used to heat the thermocouple rods, which were wrapped in aluminum-foil heat shields and mounted in an evacuated container. The

Fig. 7. Boiling-fuel Thermocouple Rod No. VA-2, Central Fuel Temperature vs Reactor Thermal Power and Burnup, Cores PSH-1A, B, and C; BORAX-V

Fig. 8. Boiling-fuel Thermocouple Rod No. ZA-19, Central Fuel Temperature vs Reactor Thermal Power and Burnup, Cores PSH-1A, B, and C; BORAX-V

Fig. 9. Comparison of Boiling-fuel-thermocouple-rod Central Temperatures for 4.95 w/o-enrichment, Rod No. ZA-20, vs 9.9 w/o-enrichment, Rod No. ZB-9, Cores PSH-1A, B, and C; BORAX-V

TABLE I. Thermal-time-constant Values for
"New" Boiling-fuel Thermocouple Rod

Thermocouple Rod Number	Run Number	Initial Fuel Temperature, °C	Final Fuel Temperature, °C	63.2% Total Time Constant, τ sec [†]
VA-2	2	459	262	8.0
VA-2	3	452	237	7.9
WA-8	1	22	246	11.1
WA-8	3	456	260	9.0
WA-8	4	463	257	10.3
VA-1	1	27	259	6.0
VA-1	2	458	264	7.2
VA-1	3	27	260	5.2
VA-1	4	459	263	6.2
Overall Average				7.9

[†]Total time constant for thermocouple rod and thermocouple (the time required for the output of the thermocouple to make 63.2% of the change produced by the applied temperature transient).

rapid heating of the thermocouple rods and slow subsequent cool-down provided 63.2% time constants on the thermocouples contained in thermocouple rods VA-1, VA-2, and WA-8 of 0.98, 0.9, and 1.5 sec, respectively.^{4,5} A time constant of 63.2%, based on the solution of a first-order differential equation, was chosen primarily because of its universal use, and not because the first-order equation was felt to fit the physical system. The same method was applied in all tests simply to measure changes. [Output = $f(1 - e^{-t/\tau})$, by definition.]

Using the overall average total time constant of 7.9 sec and the average thermocouple time constant of 1.12 sec (determined from above), a representative "new" thermocouple-rod thermal time constant of about 6.8 sec is obtained.*

Model studies of the reactor dynamics of boiling core B-2, BORAX-V, used a thermocouple-rod thermal time constant of 4.2 sec, which

*The simple operation of subtracting one time constant from the overall time constant is not theoretically correct because of the interrelated thermal constants of the fuel, jacket, gaps, and thermocouple. It is of practical use, however, and additional experimental information bearing on this subject is discussed in paragraphs B-2 and B-3 of this section.

was based on a constant fuel thermal conductivity of 6.2×10^{-3} cal/sec-cm-°C (1.5 Btu/hr-ft-°F).⁶ Overall thermocouple-rod thermal conductances were calculated on the basis of fuel-temperature measurements during operation with peripheral superheating core PSH-1, BORAX-V, and were found to range between 2.4×10^{-3} (0.57) and 9.1×10^{-3} cal/sec-cm-°C (2.20 Btu/hr-ft-°F), depending on reactor power and fuel burnup.⁷ These latter values extrapolate to a value of about 4.5×10^{-3} cal/sec-cm-°C (1.1 Btu/hr-ft-°F) at zero reactor power for an unirradiated fuel rod. Correction of the model for lower thermal conductivity would bring the model into closer agreement with measured values.

2. Tests on Irradiated Thermocouple Rods

A repetition of these transient tests was planned for thermocouple rods VA-1 and VA-2 after irradiation in the BORAX-V reactor. Failure of the thermocouple in boiling-fuel thermocouple rod VA-1 during irradiation in core PSH-1 and lack of adequately shielded facilities prevented the repetition of the complete group of transient tests. (The 427 to 260°C transient and TREAT irradiation were not repeated.) The thermal-time-constant test was repeated on fuel rod VA-2 by inserting the rod, from room-temperature conditions, into NaK at 260°C and recording the output from the fuel thermocouple. Total thermocouple-rod immersion time was measured by recording the signal from a continuously indicating, resistance level-detector, and was found to require 0.7 to 1.0 sec from the time of initial NaK contact.

The 63.2% total time constant was found to average 13.1 sec* on the basis of four test runs. This indicates a considerable change from the 7.9- and 8.0-sec values shown in Table I for the "new" condition of fuel rod VA-2.

3. Thermal-analog Model Studies of Thermocouple Rod

In an attempt to simulate the transient test findings, a thermal-analog model of the boiling-fuel thermocouple rod was set up and tested by use of an analog computer. Typical values of thermal resistances and heat capacities were tried in various combinations to get approximations of the measured time response for the boiling-fuel thermocouple rod. Various values of resistance and capacitances were used to study the relative effects of changes in resistance of the jacketing and "film" thermal resistance of fuel, and the thermocouple time constant. ("Film" is defined as the lack of wetting of the jacket.) Since the steady-state, in-reactor fuel temperature was not governed by heat-capacity and thermal-resistance values associated with the thermocouple, the primary effects studied on the model were those of variations in the fuel and jacketing parameters.

*From measured values of 11.5, 13.0, 13.5, and 14.5 sec.

The analog circuit representing the thermocouple rod is shown in Fig. 10. In Fig. 11, some results of the model study are compared with actual tests on the fuel rods. The relatively large initial delay in the fuel-thermocouple response (curves 1 and 2) is believed to be due primarily to a film resistance resulting from a lack of wetting of the stainless-steel fuel jacketing by the NaK. Time zero is taken as the point of initial contact of the bottom tip of the fuel rod with the NaK. Other studies made with the model are summarized in Fig. 12 and Table II.⁷

The transient tests in the NaK vessel with thermocouple rod VA-2 confirmed the in-reactor, static-temperature indications of an approximate 50% reduction in the thermal conductance of the fuel due to fuel burnup and operation at elevated temperature. The analog studies were limited, and the model and physical case were not matched exactly. Similar time-response curves and 63.2% time-constant values were obtained with the model. Furthermore, changes in the thermal resistance of the fuel are much more important to the overall time constant than changes in fuel jacketing or film resistances because of the small value of the ratio of jacket heat capacity to fuel heat capacity. (The overall time constant is directly related to the "RC" time-constant values of the sections of the thermocouple rod.)

C. Calibration Work

Two independent programs were carried out to determine the calibration stability of the W/W-26w/oRe thermocouples. The first test was made after operation of BORAX-V with the boiling core designated as B-2, and the second during operation with core PSH-1 in BORAX-V.

R_1 = Thermal resistance of jacketing

R_2 = Total thermal resistance of fuel and fuel-to-jacket gap

R_3 = Effective combined thermal resistance of fuel-to-thermocouple gap and thermocouple

C_1 = Total heat capacity of jacketing

C_2 = Total heat capacity of fuel

C_3 = Total heat capacity of thermocouple

E = Initial temperature differential between thermocouple and heat source
(model assumes a step increase in temperature)

V_{out} = Temperature of thermocouple

Fig. 10. Analog Thermal Model for Boiling-fuel Thermocouple Rod of BORAX-V

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Fig. 11. Computed and Measured Thermal Time Response of Boiling-fuel Thermocouple Rods; BORAX-V

ID-103-A3144

Fig. 12. Computed Thermal Time Response of
Boiling-fuel Thermocouple Rods; BORAX-V

TABLE II. Constants Used in Studies of Analog Thermal Model

Run No.	R_1	C_1	R_2	C_2	R_3	C_3	$\frac{R_2 C_2}{R_1 C_1}$	$\frac{R_2}{R_1}$	$\frac{C_2}{C_1}$	τ , sec
A	5	0.2	10	1	10	0.1	10	2	5	12.8
B	5	0.2	5	1	10	0.1	5	1	5	7.6
C	2	0.2	5	1	10	0.1	12.5	2.5	5	6.9
D	10	0.2	10	1	5	0.2	5	1	5	13.5
E	0.1	0.2	10	1	5	0.2	500	100	5	12.1

- NOTES: 1. Resistances in megohms, capacitors in μF .
2. Time constant, τ , is time required to make 63.2% of total change in temperature from time zero.
3. Based on original fuel design, the constants were calculated to be related by:

$$\frac{R_2 C_2}{R_1 C_1} = \frac{RC(\text{Fuel})}{RC(\text{Clad})} = 1050, \quad \frac{R_2}{R_1} = \frac{R(\text{Fuel})}{R(\text{Clad})} = 244,$$

$$\frac{C_2}{C_1} = \frac{C(\text{Fuel})}{C(\text{Clad})} = 4.3.$$

The effective time constant of the thermocouple is of the order of 0.5 to 1 sec (in the preirradiated condition).

In the first case, a new fuel-rod thermocouple and two Chromel-Alumel* thermocouples were mounted in the capsule shown in Fig. 13 and placed in a vacant fuel-rod space in the EBR-I reactor. Gamma and neutron heating, above coolant temperature, provided a maximum temperature of 366°C while in a fast flux of about 10^{14} nv. The thermocouples were exposed to six cycles of reactor startup and shutdown, and received an integrated fast-neutron exposure of approximately 2×10^{18} nvt. During four steady-state periods of reactor operation at 975 kW, the W/W-26w/oRe thermocouple attained an indicated temperature of $352 \pm 3^\circ\text{C}$, and the two Chromel-Alumel thermocouples indicated a temperature of $363 \pm 3^\circ\text{C}$. There was no apparent shift in calibration among the three thermocouples during the four maximum-power cycles of reactor operation.

To verify the conclusion that the irradiation had no apparent effect on the performance of the thermocouples, the three thermocouples and fixture were placed in a muffle furnace in a hot cell and calibrated at temperatures up to 1092°C. Three temperature cycles were run from room temperature to 824, 1026, and 1092°C before the test was terminated by

*Trade name of Hoskins Manufacturing Company.

failure of the tantalum-sheathed W/W-26w/oRe thermocouple owing to loss of the argon purge. The calibration between the three thermocouples showed a maximum disagreement of 12°C over the range of 260 to 982°C , with no noticeable "drift" in calibration from one thermal cycle to another. It was concluded that the W/W-26w/oRe thermocouple retained its normal accuracy rating for the duration of all tests performed.

Fig. 13. Fixture for Determining Thermocouple Radiation Damage; EBR-I

The second calibration test confirmed the accuracy of the thermocouple in boiling-fuel thermocouple rod VA-2 by preoperational, in-reactor (BORAX-V), zero-power calibrations and by a postirradiation furnace calibration. This test was conducted by placing the irradiated boiling-fuel thermocouple rod VA-2, together with a new thermocouple rod, VA-3, and a Chromel-Alumel test thermocouple, in an argon-purged electric furnace located in a hot cell. A maximum temperature of 924°C was reached with the arrangement. As shown in Table III, no obvious error pattern was followed by either fuel-rod thermocouple.

The only known errors worth consideration are indicated by the manufacturer's standards for the thermocouples. For the Chromel-Alumel thermocouple, this is $\pm 2^{\circ}\text{C}$ for temperatures up to 277°C and $\pm 3/4\%$ of temperature for values greater than 277°C . Errors for the W/W-26w/oRe thermocouple are stated as $\pm 4^{\circ}\text{C}$ for temperatures up to 427°C and $\pm 1\%$ of temperature for values above 427°C . Consideration of these errors alone leads to the conclusion that no measurable calibration change was produced in the W/W-26w/oRe thermocouple after a neutron-flux exposure of approximately 2×10^{18} nvt in BORAX-V.

TABLE III. Temperature Intercalibration between W/W-26w/oRe
Thermocouple Rods and Chromel-Alumel Thermocouples

(Temperatures in °C)

Chromel-Alumel Thermocouple	Irradiated Thermocouple Rod VA-2	Nonirradiated Thermocouple Rod VA-3
24	31	29
94	31	29
147	152	140
216	226	219
267	277	266
309	321	313
367	376	368
427	435	431
491	496	493
539	544	543
312	318	314
533	539	537
594	595	596
645	644	646
705	704	710
760	760	763
822	819	824
871	871	877
924	-	929
815	815	822
705	703	713
591	587	596
482	478	488
368	367	373
270	270	274

IV. DESIGN AND TESTING OF IMPROVED THERMOCOUPLES

A. Thermocouple Performance Requirements

The thermocouples used to measure BORAX-V boiling-fuel temperatures would not be expected to indicate temperatures in the center of the UO_2 -fueled boiling-fuel thermocouple rods if the reactor were operated at maximum design power. A reaction between the BeO insulation and tantalum sheathing occurs at approximately 2200°C , and BeO melts at about 2550°C . Both these temperatures are below the maximum of 2760°C which would be expected under full-power conditions with flux peaking and low coolant flow. Improved sensors were designed and tested for measurement of temperatures to this upper limit.

In the selection of an improved design, several factors were considered. The materials comprising the thermocouple had to be chemically stable so that little or no interaction would occur between the sensing wires, the insulating material, and the sheathing. Metallurgical stability was required to the extent that at 2760°C the thermocouple materials would retain enough strength to enable the composite sensor to perform satisfactorily in a low-stress environment. A third consideration was electrical stability. Ideally, the junction of the sensing wires should produce a strong thermoelectric signal, increasing linearly with temperature to the maximum temperature expected. Insulating material should have high electrical resistivity at all temperatures to be experienced, thereby minimizing leakage of current to the sheathing and between the sensing wires.

B. Reference Designs

Of the refractory oxides, only ThO_2 has a melting point high enough to be considered as insulating material in a 2760°C thermocouple. The only other possible candidates were MgO , with a melting point of 2800°C , and HfO_2 , with a melting point of 2777°C . Both of these melting points are too close to the anticipated maximum service temperature to warrant consideration of either oxide. (MgO has also been reported to undergo a reaction with tungsten above approximately 2100°C .⁸)

Tungsten, rhenium, tantalum, and their alloys are the only metals having melting points above 2760°C . Although tantalum would not be expected to interact with ThO_2 ,⁸ tantalum was not considered for use in the new design as jacket material because of its high affinity for, and embrittlement by, oxygen. Tungsten in its pure form is quite difficult to work and becomes extremely brittle after subjection to high temperature. Rhenium does not have these limitations that plague tantalum and tungsten, but the high cost of this element would make thermocouples with rhenium sheathing very expensive. Since the alloy of W-26w/oRe is reasonably workable and substantially more ductile than tungsten after high-temperature heat treatment, this alloy was selected for reference sheathing.

Tungsten/tungsten-26w/oRe thermocouples have a high thermo-electric output, excellent sensitivity, and a reasonably linear emf-temperature relationship to temperatures in excess of 2760°C. The greatest deficiency of this thermocouple is the extreme brittleness of the tungsten leg. Rhenium in combination with tungsten-26w/oRe has been found to be unsuitable as a thermocouple for temperatures above 2200°C.⁹ Accordingly, W/W-26w/oRe was selected as the most satisfactory couple for such a high-temperature application.

The alternate of tungsten-5w/o rhenium, in place of the pure tungsten leg, was offered to prospective vendors because of their insistence that it was easier to fabricate.

Two purchase orders were placed with separate vendors for sample experimental thermocouples to be used for testing their characteristics. The first vendor furnished two thermocouples consisting of W/W-26w/oRe conductors in swaged ThO₂ and W-26w/oRe sheaths. Great difficulty was experienced in filling the order (which called for six pieces) because of wire breakage and sheath cracking, and the remainder of the order was cancelled. The second vendor supplied six pieces to a modified design: Hard-fired thoria insulation was used in very lightly swaged W-26w/oRe sheathing, and W-5w/oRe wire was substituted for the pure tungsten wire. These latter six pieces proved relatively easy to fabricate in comparison with the first group. A typical construction detail for a thermocouple is shown in Fig. 14.

ID-103-A3384

SHEATH--0.157 ± 0.008 cm OD--W-26% Re 0.025 ± 0.005 cm Wall

WIRES--#30 AWG W-5% Re or W, and W-26% Re

INSULATION--THORIA--99.94% Purity, Hard Fired

Fig. 14. Idealized Hot-junction Construction for W/Re Thermocouple (Longitudinal Section); BORAX-V

BX5-6-2189-A

Fig. 15. Thermocouple Calibration Furnace; BORAX-V

C. Test Results on Swaged Thermocouples

The two W/W-26w/oRe thermocouple samples of swaged construction were tested in an electric resistance furnace heated by a graphite heating element. A tantalum container, concentric with the heating element, held the thermocouples. A schematic diagram of the furnace arrangement is shown in Fig. 15. Thermocouples were normally tested in the argon atmosphere of the tantalum tube, but were exposed to helium when a leak in the tube occurred. Calibration was carried out in two separate test series as indicated in Tables IV and V and in Fig. 16. Each calibration point required approximately 15 to 20 min to ensure temperature stability.

TABLE IV. Temperature-millivolt Equivalents for
Tungsten/Tungsten-26w/o Rhenium Thermocouples
Nos. 1 and 2; First Cycle

Temperature, °C			Millivolts*	
Chromel-Alumel Thermocouple	Uncorrected Pyrometer	Corrected Pyrometer	W/W-Re No. 1	W/W-Re No. 2
19.5			-0.014	-0.014
611			6.62	6.41
967	970	970	13.69	13.48
	1516	1552	25.31	25.22
	1585	1627	26.57	26.54
	1699	1755	28.57	28.63
	1793	1849	30.08	30.25
	1896	1957	31.97	32.23
	2026	2088	33.74	34.15
	2126	2188	35.00	35.47
	2225	2290	36.11	36.54
	2340	2410	36.81	37.47
	2471	2685	39.53	38.97**
	2480	2551	37.77	38.30
	2572	2793	40.81	40.48**
	2707	2954	41.29	41.58**
	2335	2407	36.90	37.42
	2221	2288	36.30	36.48
	2132	2193	35.31	35.34
	2004	2071	33.48	33.43
	1993	1999	32.35	32.29
	1804	2140	29.79	29.70
	1713	1769	28.06	27.94
	1599	1641	25.96	25.81
	1471	1507	23.43	23.27
	960	960	12.55	12.31

*24.5°C reference junction for W/W-Re thermocouple.

**Data from separate test.

TABLE V. Temperature-millivolt Equivalents for Tungsten/Tungsten-26w/o Rhenium Thermocouples Nos. 1 and 2; Second Cycle

Temperature, °C			Millivolts*	
Chromel-Alumel Thermocouple	Uncorrected Pyrometer	Corrected Pyrometer	W/W-Re No. 1	W/W-Re No. 2
	960	960	12.55	12.31
	1519	1555	24.34	24.36
	1599	1641	25.95	25.81
	1719	1774	28.32	28.19
	1796	1851	29.77	29.66
	1929	1991	32.41	32.33
	2013	2080	33.76	33.69
	2125	2185	35.33	35.33
	2226	2290	36.44	36.60
	2335	2410	37.13	37.47
	2455	2525	37.80	38.17
	2475	2478	39.04	38.45**
	2545	2770	39.86	39.34**
	2690	2935	41.08	40.66**
	2338	2407	37.16	37.48
	2232	2300	36.38	36.62
	2135	2195	35.39	35.51
	2025	2090	33.82	33.81
	1938	1999	32.48	32.39
	1796	1851	29.58	29.45
	1702	1757	27.94	27.76
	1596	1638	25.85	25.66
	1507	1543	24.15	23.95
	982	982	13.03	12.80
22.4			-0.005	-0.005

*24.5°C reference junction for W/W-Re thermocouples.

**Data from separate test.

Fig. 16.

Calibration of Two W/W-26w/oRe Thermocouples in Argon and Helium Atmospheres; BORAX-V

Note that the performance curve (Fig. 16) falls considerably below the curve determined by the manufacturer's published data.

During the calibration of the two thermocouples, a 10-cm length at the tip end of each sensor was within the hot zone of the heating furnace. In attempting to obtain calibration points at and near 2760°C, temperatures were incorrectly determined by means of an optical pyrometer. Actual temperatures were high enough above 2760°C to cause incipient melting of the thermocouple hot junctions. Metallographic examination of one of the thermocouples gave no indication of reaction between the components of the thermocouple.

Figure 17 is a transverse section taken 1.3 cm from the tip of the thermocouple. Melted sheathing and fusion of wires to the sheathing are clearly indicated. A longitudinal section taken 1.3 cm from the tip is

38728

85X

Fig. 17. Transverse Section at 1.3 cm from Tip (Hot Junction) of W/W-26Re Thermocouple No. 1, Showing Evidence of Incipient Melting of Sheathing and Sensing Wires

shown in Fig. 18. The W-26w/oRe sheath and conducting wire are fully recrystallized, as expected, and the tungsten wire (partially polished away in this view) is extremely large-grained. At 12.5 cm from the tip, a point that was outside the furnace hot zone, the sheath and wires display a worked structure, which is characteristic of all the remaining length of the thermocouple. Figure 19 is a longitudinal section showing the structure at this point. In this photomicrograph, a crack is seen to run longitudinally along the one wire. Both wires in this thermocouple were found to be cracked in this manner.

Fig. 18 Longitudinal Section at 1.3 cm from Tip (Hot Junction) of W/W-26Re Thermocouple No. 1, Showing Large-grained Structure of Sheathing and Sensing Wires after Calibration to 2760°C

Fig. 19. Longitudinal Section at 12.5 cm from Tip (Hot Junction) of W/W-26Re Thermocouple No. 1, Showing Worked Structure of Sheathing and Crack in Sensing Wire

D. Test Results on Lightly Swaged Thermocouples

In a second group of six thermocouples, insulated with hard-fired thoria, only light swaging was used to reduce the size of the sheathing. In this case, W-5w/oRe/W-26w/oRe conductor alloys were used as thermocouples. It was hoped that a W-5w/oRe alloy leg would be less brittle than a tungsten leg. It has been reported, however, that the use of a W-5w/oRe alloy in place of the positive tungsten leg of the thermocouple offers no advantage, so far as the brittleness problem is concerned, for service at temperatures above 2200°C.¹¹ The thermocouples were calibrated during two furnace cycles up to 2760°C, by use of the equipment described before. Data from testing are presented in Figs. 20 and 21, and in Tables VI through IX. A much greater change in output between cycles occurred, compared to the change in output between cycles on the two W/W-26w/oRe thermocouples. Although the reason for this change is not known, there are at least three definite possibilities. During the first cycle, the W-5w/oRe and W-26w/oRe legs may have realloyed at the junction to form a different thermocouple, causing an altered second-cycle output. This type of occurrence has been observed with thermocouples having Pt-Rh alloys in

each leg, when used at their upper recommended temperature limits.¹² A suggestion that such a phenomenon may have occurred may be inferred from the appearance of a longitudinal section through the junction end of thermocouple No. 2. In Fig. 22a, one leg of the thermocouple (the other leg is missing) is seen to be metallurgically bonded to the nose plug. Bonding must have occurred during testing, since the legs were not welded to the sheath during fabrication of the thermocouple. Presumably, a diffusion-bonding (and realloying) occurred during the first cycle. This factor is not considered as good a probability as those discussed later, when it is considered that calibration instability was not experienced on the first two samples, which were subjected to even more severe conditions during test.

BX5-7-2069-6

Fig. 20. Calibration of W-5w/oRe vs W-26w/oRe Thermocouples Nos. 1, 2, and 3 in Argon Atmosphere; BORAX -V

BX5-7-2142-C

Fig. 21. Calibration of W-5w/oRe vs W-26w/oRe Thermocouples Nos. 30, 31, and 32 in Argon Atmosphere; BORAX-V

TABLE VI. Temperature-millivolt Equivalents for W-5w/oRe-
W-26w/oRe Thermocouples Nos. 1, 2, and 3; First Cycle

Temperature, °C			Millivolts*		
Chromel-Alumel Thermocouple	Uncorrected Pyrometer	Corrected Pyrometer	T.C. No. 1	T.C. No. 2	T.C. No. 3
19			-0.05	-0.05	-0.05
599			9.58	9.61	9.40
987	988	988	17.33	17.12	17.08
	1482	1532	26.25	26.15	26.10
	1599	1663	28.20	28.10	28.09
	1705	1779	29.86	29.77	29.81
	1779	1860	31.02	30.95	31.10
	1882	1971	32.42	32.41	32.42
	1988	2085	33.70	33.77	33.66
	2110	2210	35.15	35.37	35.03
	2215	2320	36.21	36.45	35.92
	2315	2425	37.05	37.24	36.61
	2420	2530	37.21	37.79	37.19
	2545	2665	38.29	38.22	38.13
	2625	2750	38.54	38.54	38.67
	2505	2625	37.84	37.83	37.83
	2410	2520	37.14	37.21	37.16
	2312	2423	36.24	36.52	36.34
	2210	2315	35.50	35.85	35.56
	2120	2220	34.90	35.15	34.76
	1982	2075	33.49	33.54	33.21
	1892	1982	32.42	32.49	32.13
	1788	1868	30.70	30.79	30.43
	1699	1774	29.46	29.57	29.21
	1610	1674	28.27	28.38	28.02
	1496	1546	26.38	26.49	26.17
	996	996	16.75	16.77	16.51

*24.5°C reference junction for sample thermocouples.

TABLE VII. Temperature-millivolt Equivalents for W-5w/oRe-
W-26w/oRe Thermocouples Nos. 1, 2, and 3; Second Cycle

Temperature, °C			Millivolts*		
Chromel-Alumel Thermocouple	Uncorrected Pyrometer	Corrected Pyrometer	T.C. No. 1	T.C. No. 2	T.C. No. 3
	996	996	16.75	16.77	16.51
	1487	1538	25.93	25.98	25.61
	1601	1665	27.82	27.88	27.48
	1671	1746	29.07	29.13	28.75
	1780	1860	30.65	30.70	30.32
	1868	1983	32.40	32.40	32.01
	1999	2095	33.80	33.79	33.36
	2125	2222	35.29	35.27	34.68
	2210	2314	36.08	36.11	35.42
	2312	2423	36.84	36.94	36.18
	2535	2540	37.63	37.78	37.05
	2525	2695	38.18	38.31	37.68
	2632	2760	38.15	38.60	37.94
	2540	2655	37.39	38.05	37.34
	2435	2548	36.50	37.35	36.66
	2330	2442	35.66	36.62	35.98
	2228	2328	34.88	35.85	35.25
	2110	2208	34.09	34.87	34.33
	1987	2080	33.06	33.72	33.24
	1888	1978	32.11	32.62	32.18
	1783	1863	30.58	30.88	30.48
	1685	1759	29.27	29.52	29.10
	1585	1649	27.87	28.09	27.68
	1480	1529	25.71	25.93	25.57
	982	982	16.22	16.53	16.29
66			-0.04	-0.04	-0.04

*24.5°C reference junction for sample thermocouples.

TABLE VIII. Temperature-millivolt Equivalents for W-5w/oRe-
W-26w/oRe Thermocouples Nos. 30, 31, and 32; First Cycle

Temperature, °C			Millivolts*		
Chromel-Alumel Thermocouple	Uncorrected Pyrometer	Corrected Pyrometer	T.C. No. 30	T.C. No. 31	T.C. No. 32
19			-0.05	-0.05	-0.05
575			9.23	8.97	9.12
971	974	974	16.77	16.68	16.74
	1485	1536	26.23	26.13	26.15
	1608	1676	28.22	28.11	28.12
	1690	1773	29.38	29.27	29.26
	1782	1862	30.67	30.56	30.51
	1870	1985	32.43	32.32	32.27
	1997	2093	33.71	33.58	33.55
	2118	2215	35.30	35.20	35.18
	2210	2314	36.25	36.15	36.09
	2314	2425	37.02	37.00	36.91
	2537	2542	37.66	37.61	37.59
	2525	2650	38.05	37.89	38.04
	2622	2750	38.42	38.26	38.43
	2530	2700	37.85	37.73	37.88
	2430	2545	37.19	37.09	37.25
	2320	2430	36.50	36.40	36.58
	2215	2315	35.81	35.71	35.86
	2110	2208	35.05	34.95	34.97
	2001	2097	33.87	33.77	33.75
	1965	2010	32.73	32.61	32.57
	1785	1864	30.84	30.73	30.70
	1750	1788	29.76	29.64	29.60
	1588	1677	27.93	27.79	27.76
	1496	1546	26.47	26.30	26.30
	985	985	16.89	16.59	16.64

*24.5°C reference junction for sample thermocouples.

TABLE IX. Temperature-millivolt Equivalents for W-5w/oRe-
W-26w/oRe Thermocouples Nos. 30, 31, and 32; Second Cycle

Temperature, °C			Millivolts*		
Chromel-Alumel Thermocouple	Uncorrected Pyrometer	Corrected Pyrometer	T.C. No. 30	T.C. No. 31	T.C. No. 32
987	989	989	16.90	16.87	16.94
	1507	1557	26.58	26.59	26.64
	1610	1674	28.39	28.38	28.41
	1708	1782	29.51	29.50	29.53
	1818	1899	31.25	31.23	31.27
	1878	1995	32.57	32.55	32.59
	2008	2108	34.14	34.11	34.15
	2118	2215	35.38	35.32	35.38
	2195	2300	36.19	36.11	36.22
	2330	2545	37.10	37.04	37.20
	2418	2525	37.56	37.55	37.66
	2505	2625	37.98	38.01	38.06
	2612	2735	38.44	38.57	38.50
	2510	2630	37.81	37.92	37.88
	2435	2550	37.24	37.35	37.29
	2340	2450	36.71	36.83	36.79
	2235	2340	35.84	35.91	35.90
	2129	2225	34.97	34.98	35.03
	1990	2085	33.63	33.61	33.66
	1883	1971	32.30	32.28	32.33
	1788	1869	31.28	31.28	31.32
	1705	1780	29.43	29.42	29.46
	1591	1655	28.36	28.36	28.42
	1494	1543	26.53	26.51	26.57
	1005	1005	17.00	17.01	17.11
70			-0.04	-0.04	-0.04

*24.5°C reference junction for sample thermocouples.

38758

(a)

33X

38759

(b)

125X

Fig. 22. Longitudinal Section at Tip (Hot Junction) of W-5Re-W-26Re Thermocouple No. 2, Showing (a) One Leg of Couple Metallurgically Bonded to Nose Plug, and (b) Porosity in Leg and Nose Plug

A second explanation that may be offered for the output change between cycles is based on the assumption that incomplete homogeneity of the W-Re alloys was achieved during the fabrication of thermocouple legs by using powder-metallurgy techniques. With this assumption, it would be expected that subsequent heat treatments to 2760°C would bring about completion of the sintering process and thorough homogenization of the alloys.¹¹

Assuming no other variables, out of the thermocouple would be expected to remain constant from cycle to cycle, only after homogenization is complete. The porosity that is evident in both parts of Fig. 22 is perhaps indicative of the agglomeration of gaseous inclusions during testing and final sintering at 2760°C.

The thermoelectric properties of an incompletely homogenized alloy leg would be defined by the continuous metal matrix, and the output of the thermocouple would be a function of the properties of this matrix. (In this case, the continuous matrix would most likely be a tungsten-low-rhenium-content alloy, with a rhenium-rich second phase dispersed throughout the matrix.)

A third explanation for the output change might be attributed to improper relief of the effects of cold-working of the conductors.¹³

As had been found in the case of the W/W-26w/oRe thermocouples, no interaction between components of the W-5w/oRe/W-26w/oRe thermocouples was evidenced as a result of testing up to 2760°C. The transverse section seen in Fig. 23, taken 6.4 cm from the tip of thermocouple No. 31, illustrates

38766 66.3X
Fig. 23. Transverse Section at 6.4 cm from Tip (Hot Junction) of W-5Re/W-26Re Thermocouple No. 31, Showing Large-grained Structure of Sheathing and Sensing Wires after Calibration to 2760°C

the typical large-grain structure of sheath and legs after testing. At 20.3 cm from the tip of thermocouple No. 2, a point outside the furnace hot zone, the transverse section is as shown in Fig. 24.

Fig. 24. Transverse Section at 20.3 cm from Tip (Hot Junction) of W-5Re/W-26Re Thermocouple No. 2, Showing Worked Structure and Cracks in Sheathing and Sensing Wires

Cracks were commonly found in the legs of all thermocouples that were metallographically examined. These cracks were most likely introduced during fabrication of the sensor legs, since they were found throughout the lengths of the thermocouples, from the cold ends to the hot junctions. Figure 25 is a longitudinal section at 20.3 cm from the tip of thermocouple No. 31, showing a severely cracked sensor leg.

Such a flaw, in itself, may not be sufficient to impair the sensing ability of the thermocouple. However, defects of this nature are prone to propagation, especially if they are within the hot-zone portion of the

thermocouple. Complete fracture of the leg, seen in Fig. 26, a longitudinal section at 5 cm from the tip of thermocouple No. 31, was probably caused by aggravation of a fabrication defect. The brittle nature of the fracture points out the lack of ductility of the sensor leg after testing at high temperatures. In this case, fracturing may have occurred during handling after testing or during sectioning for examination. The important point is that sensors of this type are subject to low-stress failure. Accordingly, their usage should be limited to essentially stress-free environments.

Fig. 25. Longitudinal Section at 20.3 cm from Tip (Hot Junction) of W-5Re/W-26Re Thermocouple No. 31, Showing Severely Cracked Sensing Leg

Fig. 26. Longitudinal Section at 5 cm from Tip (Hot Junction) of W-5Re/W-26Re Thermocouple No. 31, Showing Brittle Fracture in Sensing Leg

V. CONCLUSIONS

A. Suitability of the Two Latest Thermocouple Designs Tested

1. Calibration Stability

Thermocouples of W/W-26w/oRe and W-5w/oRe/W-26w/oRe, with ThO₂ insulation and W-26w/oRe sheathing, have been calibrated up to 2760°C. In comparison with calibration data on the W/W-26w/oRe thermocouples, the change in output between the two cycles on the W-5w/oRe/W-26w/oRe thermocouples was quite large. Although the reason for this change is not known, three explanations have been suggested: (1) The W-5w/oRe and W-26w/oRe legs realloyed at their junction to form a different thermocouple during the first cycle; (2) The powder-metallurgy-fabricated alloy thermocouple legs were incompletely sintered and homogenized until heating to 2760°C during the first calibration cycle; or (3) the alloys were incompletely heat-treated to relieve cold-working stresses. In any case, output during the second calibration cycle would be different, but much less difference in output would be expected between the second cycle and any additional cycles.

2. Chemical and Structural Stability

There was no apparent chemical interaction between components of these thermocouples during two calibration cycles up to 2760°C. Radial and longitudinal cracking, believed to have been introduced during fabrication, was found in varying degrees in all metallographic sections through the sensing legs of the thermocouples. The portions of the W and W-5w/oRe legs that had been cycled to 2760°C displayed increased brittleness, and it is quite likely that stresses of even a minor nature would cause propagation of fabrication defects and ultimate fracturing of sensing legs.

3. Ease of Fabrication and Cost Considerations

A unique type of hot junction was incorporated into the design of these thermocouples to obviate a welded junction that would have been inherently brittle and fragile. This appears to be a successful method of fabricating the junction and should aid in preventing premature failures during thermocouple installation or low-temperature operation. Cost of this special junction is negligible compared to the general high cost of the thermocouple (a cost stemming from the difficulties associated with fabricating tungsten materials) and the expensive rhenium component.

The sample thermocouples made with ThO₂ and W-26w/oRe sheaths definitely will withstand operating temperatures of 2760°C, although their added cost over Ta-BeO construction must be justified by

the need to measure the added 556°C range of temperatures. Further work is also required to support measurements in this upper range, and is described in the following paragraphs.

B. Suggestions for Improvements

1. Metal Ductility and Homogeneity

It has been observed that most of the difficulties associated with the experimental W-Re alloy thermocouples are related to the brittleness of the metal constituents. Consideration should be given to other, more ductile, high-temperature alloys that could be substituted for some of the sensor constituents. The advantages to be gained from increased component ductility include (1) greater ease in fabrication, (2) decreased frequency of fabrication defects, (3) reduced production costs, and (4) increased reliability of performance.

Some rhenium-base alloys have been found to have good thermoelectric properties and fair ductility for high-temperature thermocouple applications. Specifically, alloys of rhenium with up to 10 w/o ruthenium have been found to be satisfactory up to at least 2480°C.¹¹ Perhaps an alloy of this type as the positive element in combination with a negative W-26w/oRe leg would provide a more ductile thermocouple for applications up to 2760°C. Unfortunately, of the refractory metals, only tungsten, rhenium, and tantalum have melting points above 2760°C; therefore an alloy of any of these with any other element will have a lower melting point. The number of alloys with good thermoelectric properties and melting points above 2760°C is undoubtedly quite small.

Fabrication techniques for the sensing legs of the thermocouples, whether by powder metallurgy or by other methods, should be adjusted to ensure that sintering and/or homogenization of the alloys are complete. If this is accomplished, one source of output instability will have been eliminated. Proper attention must also be given to proper stress relief of conductors in the completed thermocouple. Further studies should be made on the role of crystal size and stability and their effects on thermoelectric stability.

2. Junction Metallography

Perhaps additional calibration cycles to 2760°C would aid in establishing the exact cause of the change in output that occurred in the W-5w/oRe/W-26w/oRe couples. If the increment of change decreased or vanished in subsequent cycles, it would indicate that at least one of the explanations offered for this phenomenon is correct. To determine if one or more factors were important would require calibration of sensors in

which either the hot-junction realloying had progressed to an equilibrium stage, or sintering and homogenization of the sensing legs were known to be completed, or residual stresses were thoroughly annealed.

C. Proper Application and Calibration

In addition to simple furnace calibrations, which establish basic performance characteristics of the thermocouples, additional factors must be considered to ensure accuracy of measurement in the final application, as described below.

Insulation resistance remains as the primary limitation to making accurate temperature measurements much above 1650°C, assuming that the above problems of calibration, stability, and ductility can be conquered. The greatly reduced electrical resistivity of all known metal oxides at temperatures above about 1650°C leads to measurement errors mainly influenced by (1) length of thermocouple in the hot transition zone, (2) magnitude of the temperature gradient from hot to cold junctions, and (3) magnitude of the hot-junction temperature. Nonhomogeneity of wire also contributes errors due to factors (1) and (2), but this normally would not be as serious as the errors due to lowered insulation resistance. The result of lowered insulation resistance is that of averaging of local temperatures along the thermocouple with those at the hot junction. If the thermoelectric emf and insulation resistivity are accurately known as functions of temperature, the three above factors can be incorporated in a calculation that determines the net thermocouple output voltage. Such calculations, backed by calibrations under simulated operating conditions, should allow W-5w/oRe/W-26w/oRe or W/W-26w/oRe thermocouples to be applied to measurements of temperatures in the region of 2760°C.

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