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INSTRUMENTATION AND CONTROLS DIVISION

EFFECTS OF FAST-NEUTRON IRRADIATION ON SHEATHED CHROMEL/ALUMEL THERMOCOUPLES

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AUGUST 1976

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ABSTRACT

Stainless steel sheathed, magnesium oxide insulated, Chromel-P/Alumel thermocouples were tested by irradiation in the EBR-II reactor to determine their suitability for use in the Fast Test Reactor at the Hanford Engineering Development Laboratory. After exposure to high temperature (1200°F) and fast-flux irradiation (2×10^{22} nvt) in the reactor, there were small changes in the thermocouple characteristics. The measured error of these thermocouples was 2% at 1200°F. Thermoelectric inhomogeneities were generated in the Chromel and Alumel wires in opposite polarities with respect to unirradiated wires. These inhomogeneities were annealed at temperatures above 900°F. The outer diameter of the sheath increased no more than 1.2%. Irradiated thermocouples can be expected to perform satisfactorily in this environment and would survive many thermal cycles such as might occur in a reactor scram.

CONTENTS

1. INTRODUCTION

The metal-sheathed, Chromel/Alumel thermocouple was chosen for temperature sensing in nuclear reactors because it is a basic two-wire device that is rugged and corrosion resistant. Its response is fast and repeatable. Most of the existing information on reactor thermocouple performance is from thermal-neutron reactor applications. However, in liquid-metal fast breeder reactors (LMFBRs), temperature sensors are exposed to a large fast-neutron flux (10^{15} neutrons cm^{-2} sec^{-1}) in addition to a small (10^{11} neutrons cm^{-2} sec^{-1}) thermal-neutron flux.

ORNL has been developing temperature sensors for LMFBR applications, but the work reported herein is for specific application in the Fast Flux Test Facility (FFTF) at the Hanford Engineering Development Laboratory (HEDL). Thermocouples manufactured to ORNL specifications and selected by ORNL were fast-neutron irradiated in the EBR-II reactor at the National Reactor Test Site (NRTS) in Idaho, and, after irradiation, they were tested at ORNL and compared with similar nonirradiated thermocouples. The overall objective of this work was to develop sources of supply, manufacturing and fabrication techniques, and acceptance tests so that the FFTF reactor operators are assured that the thermocouples installed in the reactor are the most accurate and reliable temperature sensors available for that purpose.

Prior to the irradiation and the performance tests of the thermocouples reported herein, three major phases in the development sequence were completed:

1. Based on our previous experience in obtaining thermocouples for ORNL reactor programs, we prepared specification I.S. 509 for high reliability performance.[1]
2. The manufacturing and testing capabilities of the thermocouple industry were evaluated, and 31 typical thermocouples were tested for accelerated drift at 1600°F for 10,000 hr.[2]
3. Contracts were placed with thermocouple manufacturers whose product performance and fabrication capability indicated they could best meet the specification requirements. These thermocouples were acceptance tested by ORNL and HEDL. Of 212 thermocouples received, 99 of the 1/8-in.-OD, type 304L stainless steel sheathed, magnesium oxide insulated, Chromel-P/Alumel (type K) thermocouples were irradiated in the EBR-II reactor (Fig. 1). The sequence of testing is shown in Fig. 2.

To determine the effects of time and temperature on the sensors, 28 similar, type K thermocouples were tested at the same temperature profile and time as the irradiated thermocouples.

2. CONCLUSIONS

A major conclusion from this study is that 1/8-in.-OD, austenitic-stainless-steel-sheathed, MgO insulated, Chromel-P/Alumel (type K) thermocouple assemblies, manufactured and tested to high reliability requirements, can be expected to perform satisfactorily at 1200°F at a fast fluence up to 2×10^{22} nvt, thermal fluence of 2×10^{21} nvt, and in a gamma field of 10^{8} R/hr. The temperature indicated by an irradiated type K thermocouple may be as much as 15°F lower then the true temperature at 1200°F.

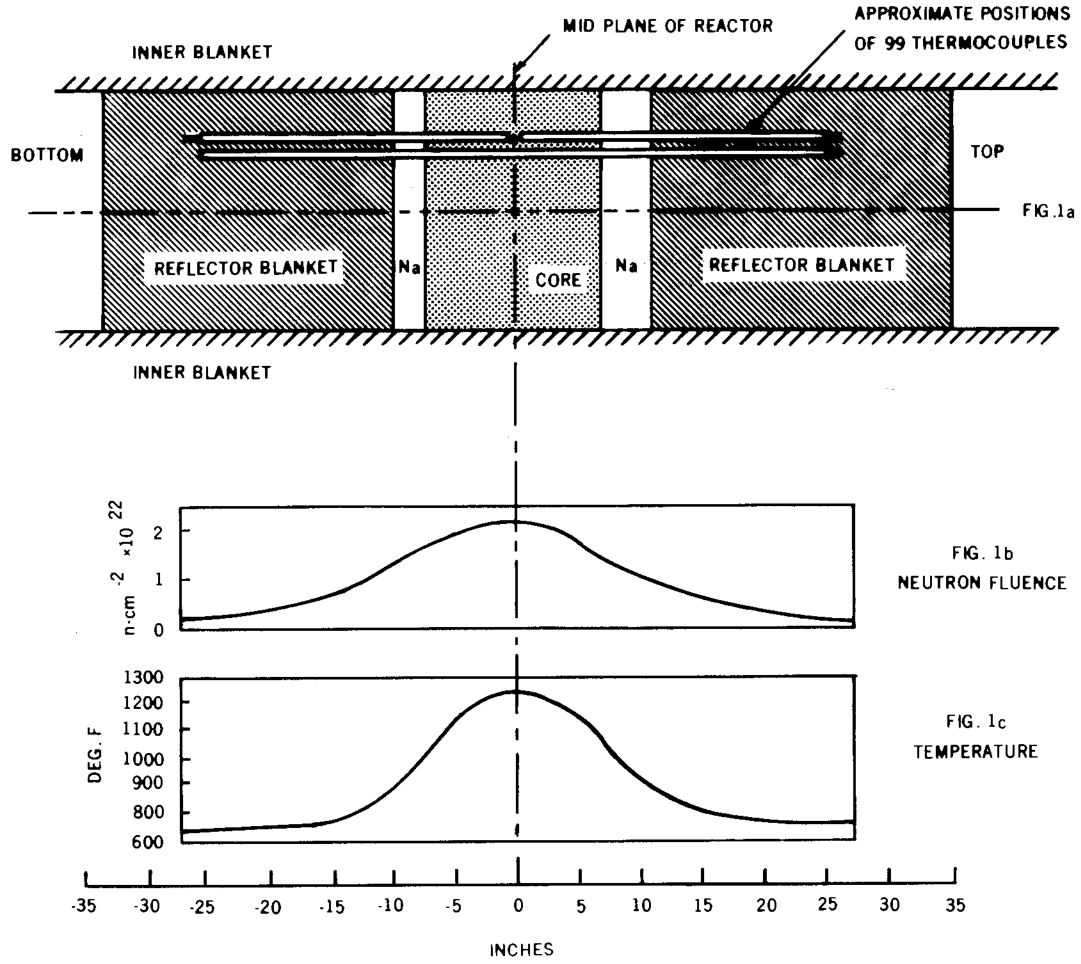

Fig. 1. Sketch of EBR-II reactor arrangement and position of thermocouples. The 26-in.-long thermocouples were placed end to end, with the hot junctions at the midplane of the reactor core. The hot junction of the 52-in.-long thermocouples were in the reflector blanket outside the core.

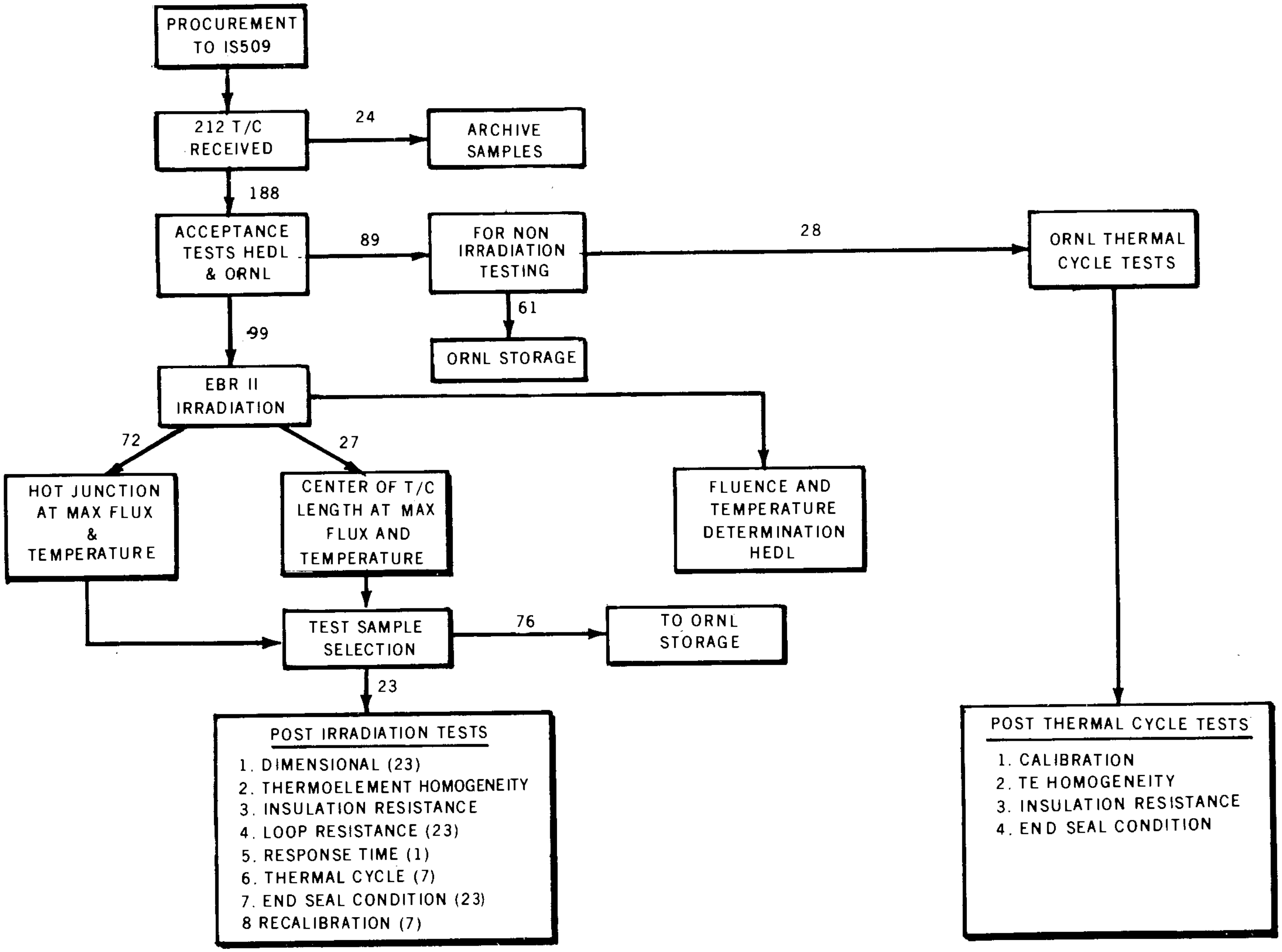

Fig. 2. Flow diagram of test.

Other conclusions are as follows:

1. An error in accuracy of no more than 2% will be caused by an inhomogeneity induced in the Alumel thermoelectric conductor by the radiation. The inhomogeneity is removable by annealing to temperatures above 900°F.
2. The time response of the thermocouple assembly and the thermocouple loop resistance will not change appreciably.
3. The diameter can be expected to increase up to 1.2%.
4. The sensor should survive 3000 thermal transients of 122°F/sec from 1250 to 750°F. This is equivalent to one reactor scram per day for 9 years.

3. RESULTS AND DISCUSSION

3.1 Sheath Dimensions

The specified diameter of the thermocouple was 0.125 in. After the 99 thermocouples had been irradiated, the diameter of 11 thermocouples was measured. Swelling was detected, amounting, in some cases, to an increased diameter slightly over 1%.

3.2 Calibration

Three types of calibrations were performed: (1) acceptance tests of thermocouples purchased from three suppliers, (2) tests of thermocouples that had been irradiated and heated in the EBR-II, and (3) tests of nonirradiated thermocouples that had been subjected to the same time-at-temperature exposure as the irradiated thermocouples. The third test was performed to separate heat effects from irradiation effects.[3]

Test results indicate that nonirradiated sensors in a fixed temperature gradient exhibited a positive error trend. We attribute this trend to short-ranged ordering effects.[4] Irradiated thermocouples exhibited errors in the negative direction, and we attribute this trend to fast-neutron damage.

The Chromel-P/Alumel thermocouples for irradiation testing were purchased to ORNL Specification I.S. 509, which requires 0.38% accuracy in meeting the standard emf-temperature relationship given in NBS Monograph 125. When the 188 thermocouples were calibrated, about one-third met this accuracy requirement at all calibration points from 530 to 1400°F. The remainder showed errors as much as 0.55%. The average error of those thermocouples meeting the accuracy requirement was 0.07% (0.85°F) at 1200°F, the design operating temperature of the Fast Test Reactor (FTR). On subsequent calibration, the error increased due to effects of short-ranged ordering (a rearrangement of the chromium atoms in the Chromel wire).

Seven thermocouples that had been irradiated in the EBR-II were calibrated in a hot cell at ORNL, with their irradiated portion in the temperature gradient of the calibration furnace. The results from these calibration tests showed a negative error as much as 2% (25°F) at 1200°F, as shown in Fig. 3. More thermocouples could not be tested because of the cost associated with hot cell testing.

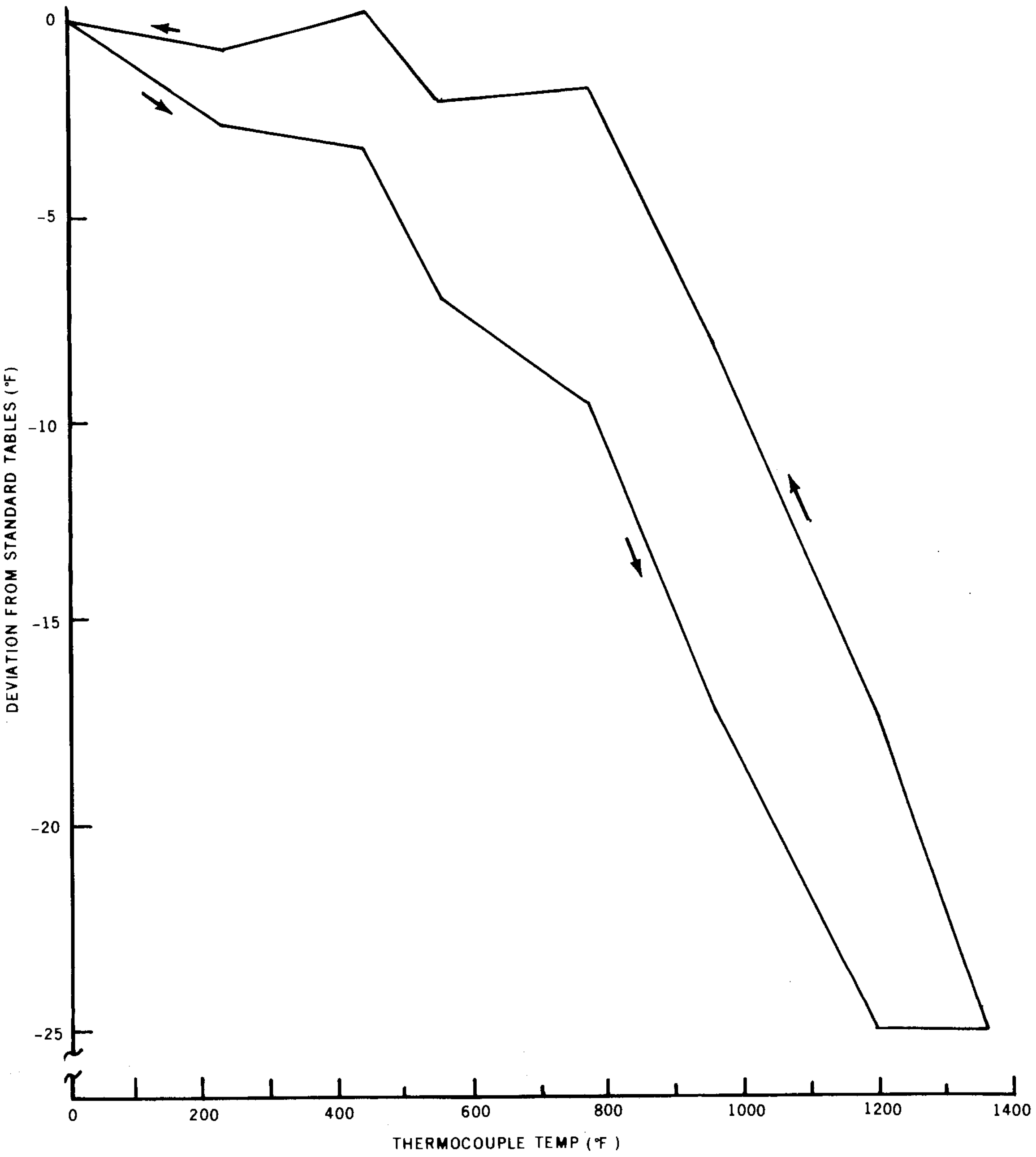

Fig. 3. Typical calibration curves of a thermocouple after irradiation with associated heating in the reactor. The errors after irradiation are much larger and in the opposite direction from those in Fig. 4. The curves indicate short-ranged ordering before irradiation and annealing after.

The control group of 28 thermocouples, subjected to similar time-temperature conditions as those encountered by the irradiated thermocouples, was also calibrated. The calibrations showed a positive error of 0.2% (2.5°F) at 1200°F due to the effects of short-ranged ordering. After the thermocouples were heated to 1427°F, the calibrations at lower temperatures showed hysteresis with reduced error, as shown in Fig. 4.

3.3 Ordering Effects

The thermoelectric wire used for this experiment was Chromel-P/Alumel, which corresponds to ISA type K. This thermoelectric material is available with two accuracy limits:

	Accuracy to 530°F	Accuracy above 530°F
Standard Grade	±4°F	±3/4%
Premium Grade	±2°F	±3/8%

The thermocouples for this experiment were premium grade.

Chromel-P/Alumel thermoelectric conductors are high-nickel alloys. Their principal constituents (nominal percentages) are as follows:

	Nickel	Chromium	Manganese	Silicon	Aluminum
Chromel-P	90	10	-	-	-
Alumel	94	-	3	1	2

In other related work,[4] it was determined that calibration errors are attributable to short-ranged ordering of the Chromel conductor.

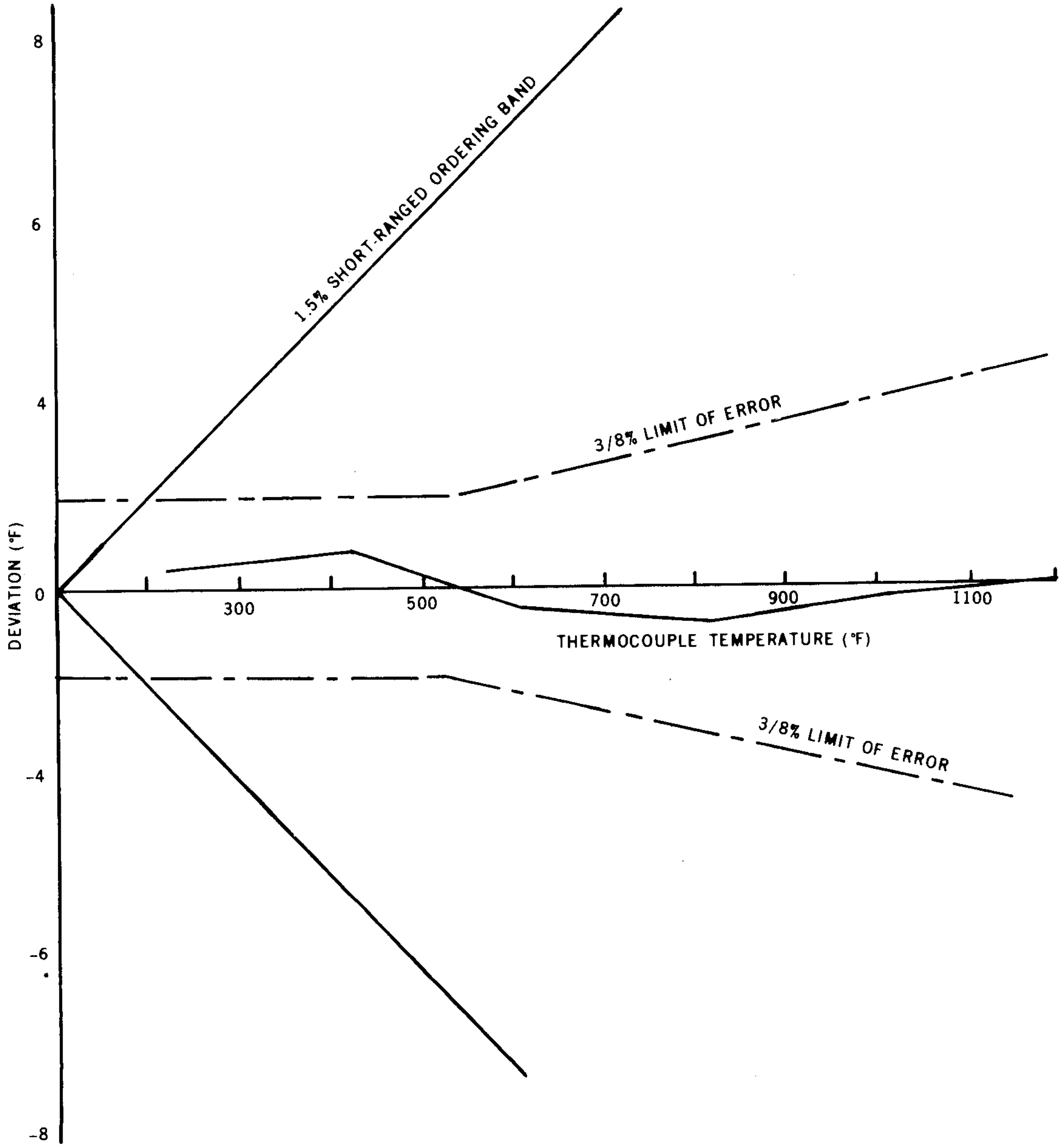

Fig. 4. Calibration of unirradiated thermocouple.

During the manufacturing process, this wire is annealed, and the chromium and nickel molecules are uniformly distributed. In calibration or use of these thermocouples at temperatures between 800 and 1400°F, there is a selective reorientation of the chromium and nickel molecules, such that a nonuniform condition, defined as "short-ranged ordering," occurs.

The effect of the ordering on the thermoelectric power of the Chromel wire is of little consequence if the ordered section is not in a temperature gradient, but if the ordered thermoelectric material is located in a temperature gradient, temperature errors up to about 1.5% can occur.

This ordering effect has caused us to seriously question the ISA-assigned accuracy limits of ±0.38% above 530°F for premium-grade material. Although it is possible to obtain this accuracy on the initial heating of fully annealed wire, any subsequent heat exposure may cause an error up to 1.5% if the heated wire is placed in a sharp temperature gradient. Figure 4 indicates the calibration limits of premium-grade thermocouple wire, the 1.5% possible positive error due to ordering, and the first calibration of typical sheathed thermocouple assembly.

3.4 Characterization of Irradiated Thermocouples

3.4.1 Inhomogeneity Tests

Calibration of the irradiated thermocouples showed a negative error opposite to the positive errors expected from exposure to time and temperature alone. Since the thermocouples were irradiated nonuniformly, that is, different portions of the length of the thermocouple received different neutron fluences and experienced different temperatures, inhomogeneity tests were performed in a hot cell to determine the location of the heat- and radiation-affected regions. These tests showed that in the region where the thermocouple was hotter than about 900°F the radiation damage was annealed out. Where the thermocouple was in a cooler region, the radiation damage accumulated and affected the thermoelectric power of the wires in an opposite and unequal manner.

An inhomogeneity test was applied to typical 52-in.-long and 26-in.-long thermocouples. A 52-in.-long irradiated thermocouple and a previously unheated thermocouple were lowered together into the sharp temperature gradient of a salt bath at 300°F, and the difference of emf of these two sensors was plotted as a function of insertion depth. The results of this test are shown in Fig. 5(e). Two regions of this curve (A and E) show a positive error of about 250 μV (11°F), attributed to short-ranged ordering. Region A is in the first 13 in. from the measuring end, and region E is the balance of the thermocouple length past 39 in. Region C did not show any inhomogeneity.

Two distinct, symmetrical, negative peaks, approximately 140 μV (6°F), have maxima at 17 in. and 35 in. from the measuring junction end of the 52-in.-long thermocouple. The 7-in. section of the thermocouple sheath around the negative peak (region D) was heated to 1800°F. An inhomogeneity scan revealed that the negative peak was removed [Fig. 5(f)]. It is postulated that the void formation effect of fast-neutron irradiation creates an effect similar to cold work.[5]

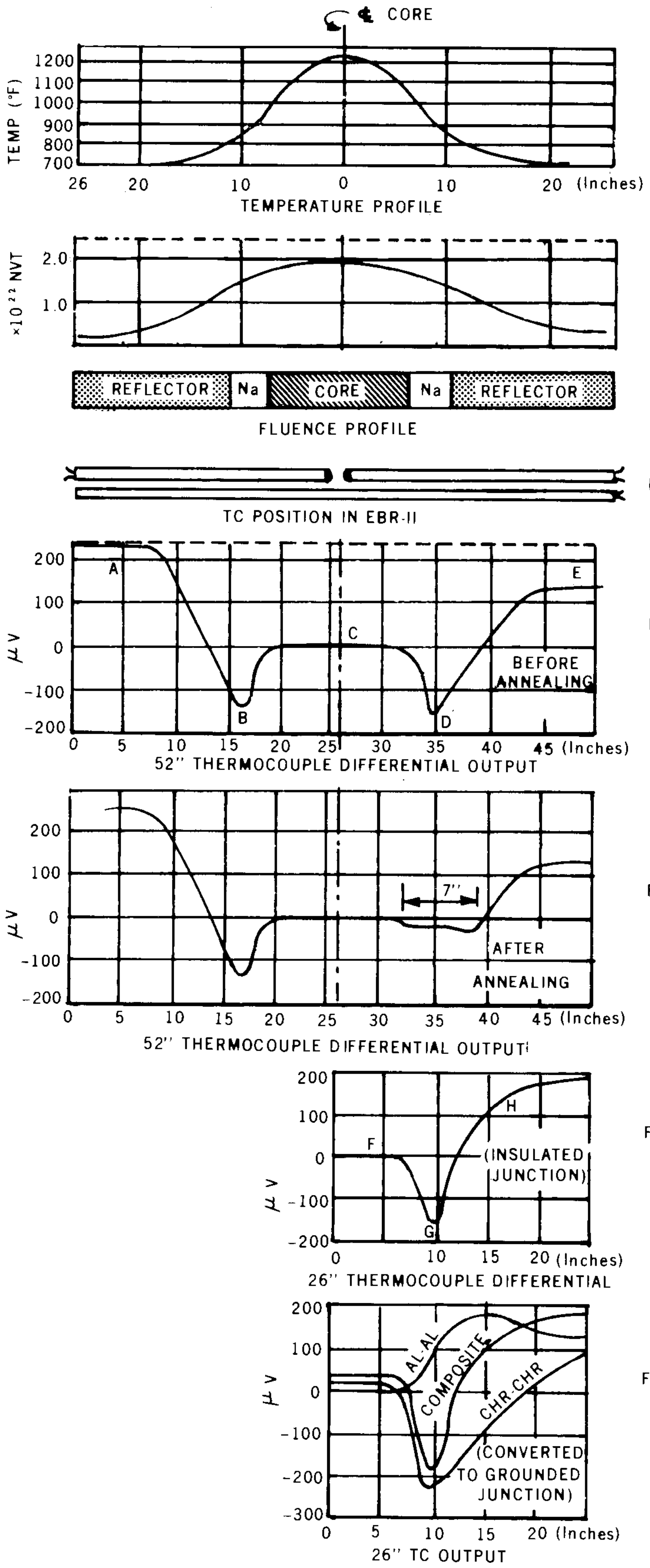

FIG. 5a NEUTRON FLUENCE PROFILE IN EBR-2

FIG. 5b TEMPERATURE PROFILE IN EBR-2

FIG. 5c REACTOR ARRANGEMENT IN EBR-2

FIG. 5d POSITION OF THERMOCOUPLES IN EBR-2

FIG. 5e 52" THERMOCOUPLE WITH DOUBLE INHOMOGENEITY PEAK

FIG. 5f INHOMOGENEITY PEAK REMOVED BY ANNEALING

FIG. 5g 26" THERMOCOUPLE WITH SINGLE INHOMOGENEITY PEAK

FIG. 5h COMPOSITE, SHOWING INHOMOGENEITY OF EACH WIRE

Fig. 5. EBR-II fluence, temperature, and sensor performance.

A 26-in.-long thermocouple which had its measuring junction at the core centerline was also analyzed. In this case [Fig. 5(f)], the same negative peak is shown in region G, about 10 in. from the core centerline, and an error of 200 μV due to short-ranged ordering caused by reactor heating was observed at the reference junction end.

Since this type of thermoelectric error is similar to that of the 52-in.-long thermocouple, the contribution of each thermoelectric wire to the observed error was investigated. The 26-in. thermocouple was converted from an insulated junction to a grounded junction assembly, and an inhomogeneity test was performed. The standard for this test was a previously unheated, grounded junction thermocouple connected differentially to determine the voltage contribution of the irradiated wires. The results [Fig. 5(h)] show an ordering effect in the Chromel wire, caused by heating during calibration; some of this effect was annealed out at about 10 in. from the measuring junction.

When the measurement of the emf of the standard was compared to that of the irradiated Alumel wire, a positive voltage signal, up to ∿180 μV, was observed in region G about 7 in. from the measuring junction. This was due to the short-ranged ordering effect of the Chromel wire. The maximum emf occurred in the gap between the core and the reflector, where the fluence was about 1.5×10^{22} nvt, the temperature was about 850°F, and the temperature gradient was ∿40°F/in. The irradiated Alumel wire did not exhibit the ordered-type error observed in the Chromel wire. Previous tests indicated that when the Alumel conductor was placed in a heated zone, the short-ranged ordering phenomenon observed in high nickel-chromium alloys did not occur. It appears that the error in the irradiated thermoelectric conductor is related to the embrittlement of the alloy in a fast-neutron field.

3.4.2 Electrical Insulation Resistance and Reference End Seals

The electrical insulation resistance (wire to sheath) of 19 of the 23 irradiated thermocouples was below the minimum specified value of 500 MΩ-ft for unirradiated thermocouples at room temperature. Ten thermocouples measured less than 0.05 MΩ-ft. The glass reference end seals showed damage and discoloration, caused by some combination of neutrons, gamma rays, and rough handling.

The insulation resistances increased substantially when the thermocouples were heated during calibration. This indicates that the low resistances were caused either by moisture absorbed through the damaged end seals or by surface contamination of the thermocouple sheath.

3.4.3 Thermal Cycling

Seven of the irradiated thermocouples were subjected to thermal cycling tests,[6] heating at 30°F/sec and cooling at 100°F/sec between 200°F and 1400°F. All of the thermoelements survived more than 3200 cycles of heating and cooling. FFTF specifications require thermocouples to survive 3000 cycles of cooling at 100°F/sec over this temperature range.

3.4.4 Loop Resistance

Loop resistances of 23 irradiated thermocouples were measured. There was no significant change from original values.

3.4.5 Response Time

Response time is defined as the time required for the emf output of a thermocouple to reach 63% of the final output resulting from a step change in temperature.[7]

The response time of the thermocouples was measured during acceptance tests before irradiation. The average response time was 678 msec; the response time of 73 of the 93 thermocouples was less than the 900 msec maximum requirement. The response time of 17 thermocouples exceeded the maximum, ranging from 937 to 1260 msec.

The average response time of the irradiated thermocouples was 755 msec, indicating that no significant changes had occurred.

3.4.6 Summary of Results

Table 1 briefly summarizes the test results discussed in the preceding sections.

Table 1. Results from tests of irradiated vs heat-treated thermocouples

	Test	Thermocouples Heated in Furnace	Thermocouples Irradiated and Heated in Reactor
1.	Sheath Diameter		Increased ∿1.2%
2.	Calibration, T[a]	0.5 to 2% high at >530°F	2% low at 1380°F
3.	Calibration hysteresis	Larger errors after initial heating, indicating short-ranged ordering	Smaller errors in calibration after heating, indicating some annealing of radiation damage
4.	Thermoelement homogeneity	emf to +60 μV[b]	Negative emf in region of high fluence, T <1000°F. No inhomogeneity in region of highest fluence, T >1000°F. Positive emf in region of low fluence, T <750°F.
5.	Annealing		Inhomogeneities removed by heating to >1800°F for 7.5 min or >1200°F for a few hours
6.	Radiation damage to Chromel and Alumel		+emf in Alumel, -emf in Chromel (30% larger)
7.	Insulation resistance	>330 MΩ-ft after heating	>330 MΩ-ft after irradiation, some <0.03 MΩ-ft
8.	Loop resistance		No change
9.	Thermal cycling survival		Tested 3200 cycles with no failures
10.	Response time		No change

[a] T = temperature.

[b] 1°F is ≃ 22 μV.

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