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TRANSIENT RADIATION EFFECTS ON THERMOCOUPLES

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ABSTRACT

Thermocouples of the type used in many power reactors were exposed to various levels of nuclear radiation in the AF-NETR reactor while immersed in a constant temperature medium. Measurements of voltage output obtained under these conditions indicate the existence of pronounced and reproducible transient changes in calibration of 15°F or more. The effects appear dependent on the level and possibly the rate of change of level of incident radiation. This magnitude of calibration change may be large enough to be of considerable significance in interpretation of in-pile thermocouple data.

INTRODUCTION

Thermocouples used in power reactors or elsewhere in radiation fields can be affected by radiation fields in several ways. It is well known that long-term irreversible changes in thermocouple calibrations can result from nvt-dependent transmutation of alloying agents in thermoelectric materials. Such effects have been studied experimentally by Kelly[1] and others and analyzed extensively by Browning and Miller,[2] who showed that chromel-alumel couples should be virtually unaffected by transmutation up to nvt values of 10^{20} cm^{-2}. Gamma heating has also long been recognized as a source of calibration error, which can usually be limited to less than a degree or two by appropriate selection of thermocouple geometry and placement.

Several authors[3,4] have postulated that nuclear radiation should also directly affect thermoelectric materials through ionization, production of interstitials and vacancies, and other solid state effects. Such radiation damage could be evidenced as relatively rapid, perhaps at least partially reversible, changes in calibration during irradiation. However, no conclusive observations of direct radiation effects have been reported in the literature, possibly because virtually all investigations of irradiation effects on thermocouples have employed post-irradiation calibrations which could not detect any transient radiation effects disappearing when irradiation ceased.

In the work reported herein, the existence of short-term radiation effects was investigated by observing thermocouple calibration changes during irradiation.

Experimental Procedure

The experimental technique used was to vary the level of radiation to which thermocouples were exposed while immersed in a constant temperature medium. Any observed changes in output voltage of the thermocouples under these conditions were thus indicative of direct radiation effects. As shown schematically in Fig. 1, sets of two nominally identical thermocouples were immersed in a bath of pure boiling water. The thermocouple leads were connected so that their respective thermoelectric emf's were opposed to each other (i.e., the emf's "buck" each other with the "hot" and "cold" junctions at the same temperature). By exposing only one of the two thermocouples so connected to high-level nuclear radiation, any direct radiation effects could be evidenced as changes from the nominally zero net emf of the "bucked" circuit. Use of this circuit with the compensated lead wires arranged symmetrically also helped to cancel out any extraneous voltages generated in leads and connectors due to inhomogeneities, cold-working effects, and temperature gradients from gamma heating.

An experimental apparatus, shown in Fig. 2, was constructed of aluminum with the constant temperature baths consisting of two interconnected tanks containing pure water brought to and maintained at boiling by electrical heaters. Water to replenish that boiled off was supplied automatically from the large reservoir shown on the right of the apparatus in Fig. 2. The connection between the tanks is provided so that both thermocouples would be immersed to equal depth in water of identical composition. Temperature gradients along the thermocouple sheaths, and small changes in boiling temperature due to pressure changes and possible accumulation of water impurities, would thus affect both junctions equally and be cancelled out in the net emf measured.

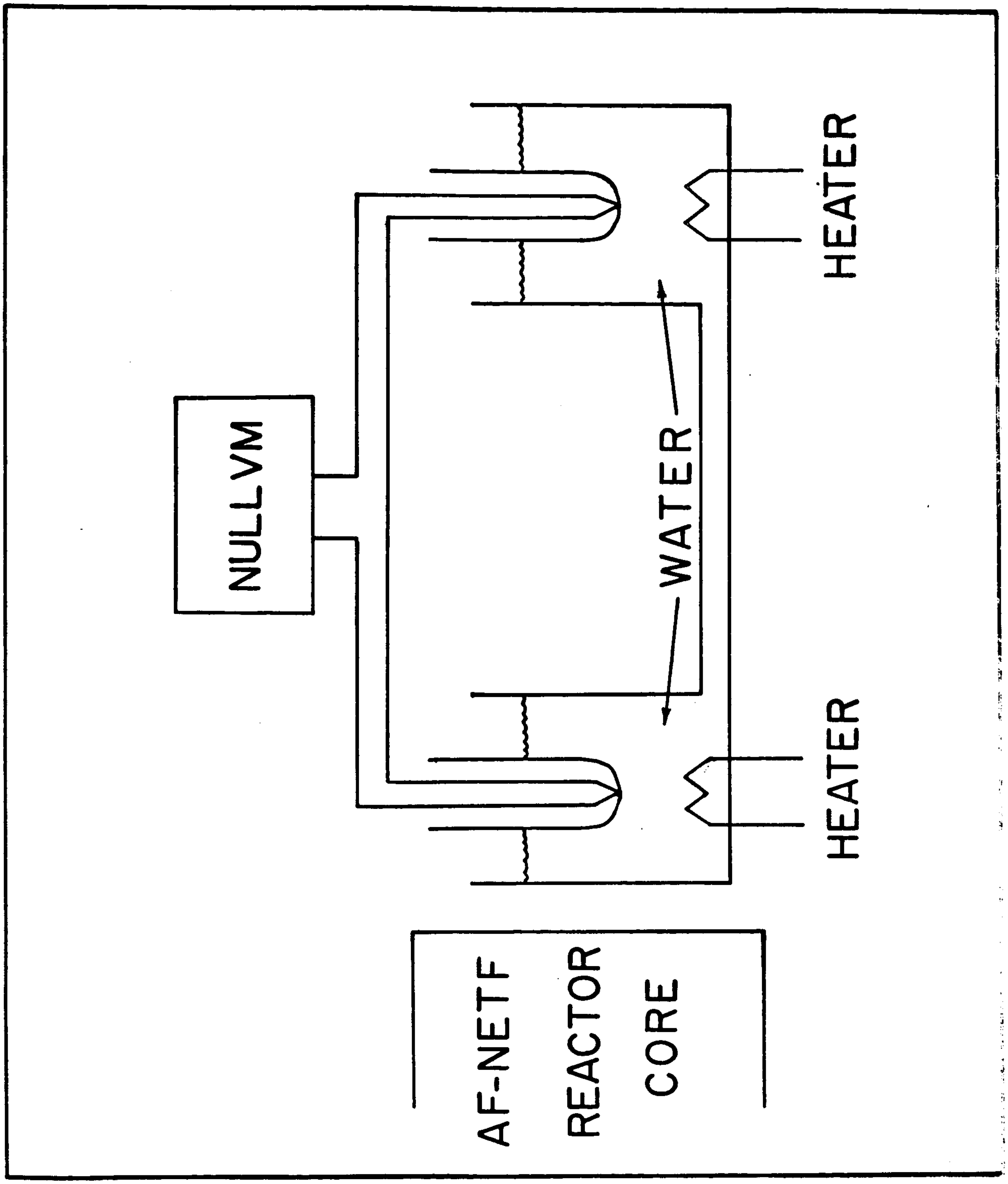

Fig. 1   Schematic Diagram of Experimental Technique.

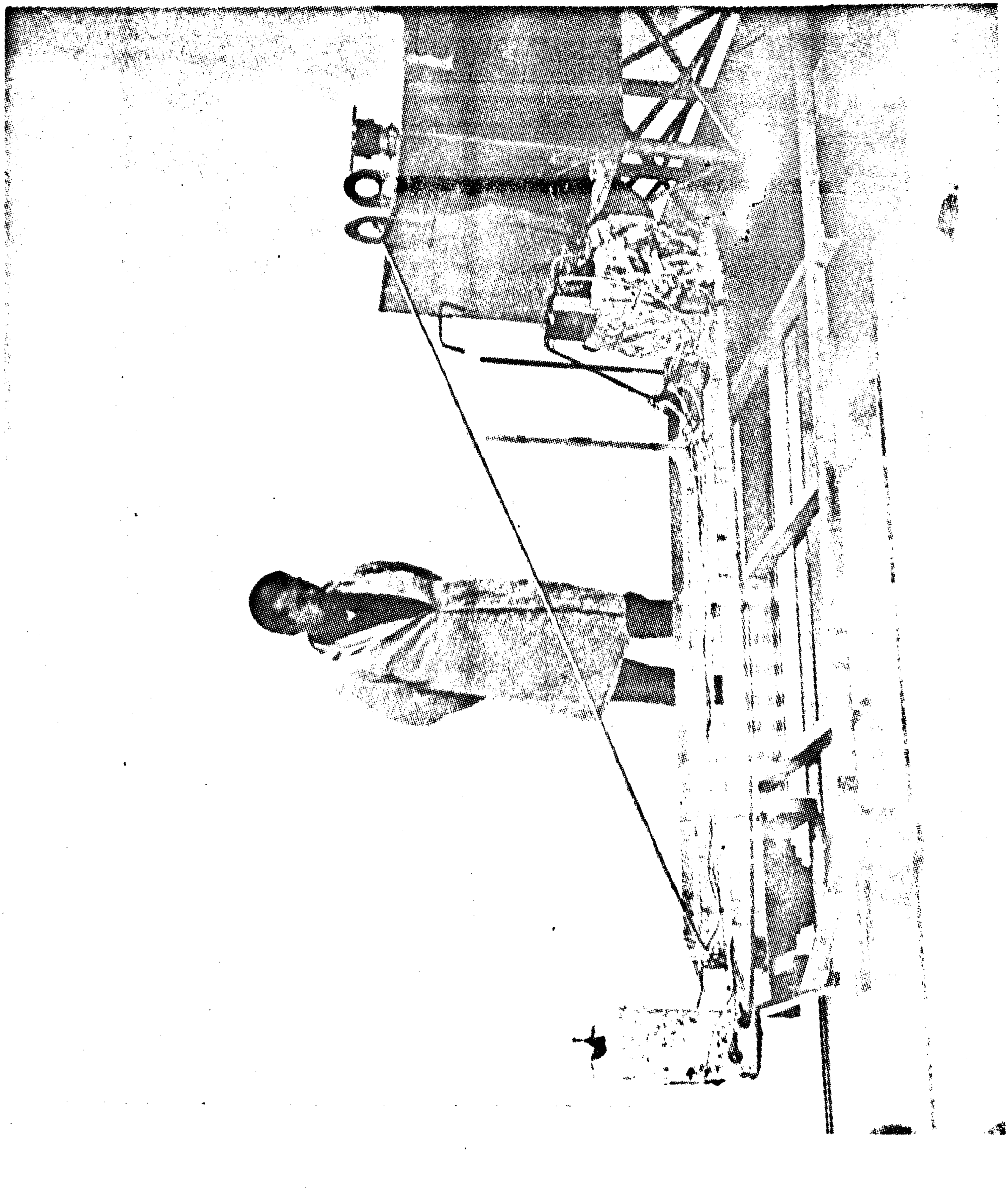

FIGURE 2--Apparatus Mounted on the NETF Cart

The experiment was also designed to minimize extraneous factors tending to affect thermocouple calibration. Chromel-alumel couples of the type commonly employed for monitoring reactor temperature were used for these experiments in order to minimize any calibration changes due to transmutation. Gamma heating effects were isolated by comparing results from thermocouples identical except for grounded and ungrounded junction construction with consequently different thermal conductivity and time response.

The entire assembly was placed in a test cell against one face of the Air Force Nuclear Engineering Test Facility (NETF) reactor at Wright-Patterson Air Force Base such that the thermocouples in the "front" bath (Fig. 1) were exposed to the maximum possible neutron flux levels. The "rear" tank was located about 9 feet from the reactor face and surrounded by 1/4 inch sheets of boral so that the flux levels experienced by the thermocouples in this tank (each of which was "bucked" against a nominally identical couple in the front tank) were much lower. The flux levels shown in Table I were measured at full power of 10 MW by activating gold and nickel wires attached to each of the tanks.

TABLE I.

Maximum Measured Neutron Flux Levels (neutrons/cm^2-sec)

	"Front" Bath	"Rear" Bath
Fast flux (>.85 mev)	6.5×10^{12}	2.4×10^{11}
Thermal flux	4.7×10^{11}	1.65×10^{10}

Gamma radiation levels were not measured in this experiment, but auxiliary experiments by NETF personnel indicate a gamma dose rate of 7×10^9 ergs/gm. C/hr in the vicinity of the experimental apparatus.

A total of 8 individual thermocouples with the characteristics shown in Table II were employed in the experiments reported here. All were commercially purchased chromel/alumel, magnesium oxide-insulated, Inconel-sheathed types certified to conform to MIL specifications for materials and calibration.

TABLE II.

Thermocouple Characteristics

Quantity	Junction Type	Sheath O.D., in.	Length, in.	Thermal Time Constant, sec.
4	Grounded	0.187	8.0	0.8
2	Ungrounded	0.187	8.0	2.15
2	Grounded	0.125	12.0	0.55

The thermocouple circuit output voltages were measured with a Model 419A Hewlett-Packard DC Null Voltmeter, whose specifications included: Input impedance $>$ 100,000 ohms; noise level $<$0.1 microvolt; drift $<$0.5 microvolt/day or $<$0.05 microvolt/°C; response time $<$1 sec; 60-cycle AC rejection $>$80 DB. A Hewlett-Packard model 3439A Digital Voltmeter with input impedance of 10 ohms and response time of 450 msec. was used to read the output of the Null Voltmeter to an accuracy of $\pm$ 0.01 millivolts.

RESULTS AND DISCUSSION

The thermocouples were exposed to radiation during a sequence of reactor operations involving 11 cycles (summarized in Table III) in which reactor power was rapidly increased, then held at constant power for a selected period of time followed by a manual scram to reduce power as quickly as possible.

TABLE III.

Radiation Exposure Cycles

Cycle No.	Power Level (MW)	Time at Max. Power(Min.)	Integrated Fluxtime at end of cycle, $cm^{-2} x 10^{-17}$	
			Fast	Thermal
1	10	115	0.45	0.032
2	10	55	0.66	0.048
3	1	58	0.69	0.049

TABLE III. (cont'd)

Radiation Exposure Cycles

Cycle No.	Power Level (MW)	Time at Max. Power(Min.)	Integrated Fluxtime at end of cycle, $cm^{-2}x10^{-17}$	
			Fast	Thermal
4	5	70	0.82	0.059
5	10	5	0.84	0.061
6	10	54	1.05	0.076
7	10	43	1.22	0.088
8	5	27	1.27	0.092
9	10	158	1.89	0.136
10	10	82	2.21	0.159
11	5	37	2.28	0.165

Characteristic voltage outputs measured for the several thermocouple sets are shown in Figs. 3 through 7 together with corresponding reactor power history. Figs. 3 and 4 give comparable results for grounded and ungrounded thermocouples with 0.187" O.D. sheaths with no prior irradiation exposure (cycles 1 and 2 of Table III). Figs. 5 and 6 show corresponding results for the same two thermocouple sets after several cycles of irradiation. Fig. 7 shows results obtained for additional grounded thermocouples with 0.125 in O.D. sheaths for the final two cycles of irradiation (cycles 10 and 11). In this instance, the two thermocouples (one in the front tank and one in the rear tank) were not connected in a bucked circuit. Instead, the output voltages were measured separately with respect to a common cold junction at ambient temperature.

From evaluation of the data obtained, the direct effects of radiation on the thermocouples studied can be summarized as follows:

1) Grounded and ungrounded thermocouples exhibited similar characteristics.

2) The output voltages for thermocouples with no prior irradiation exposure drifted downward by about 0.1 mv and stabilized about 45 min. after the reactor was brought to full power. (Figs. 3 and 4)

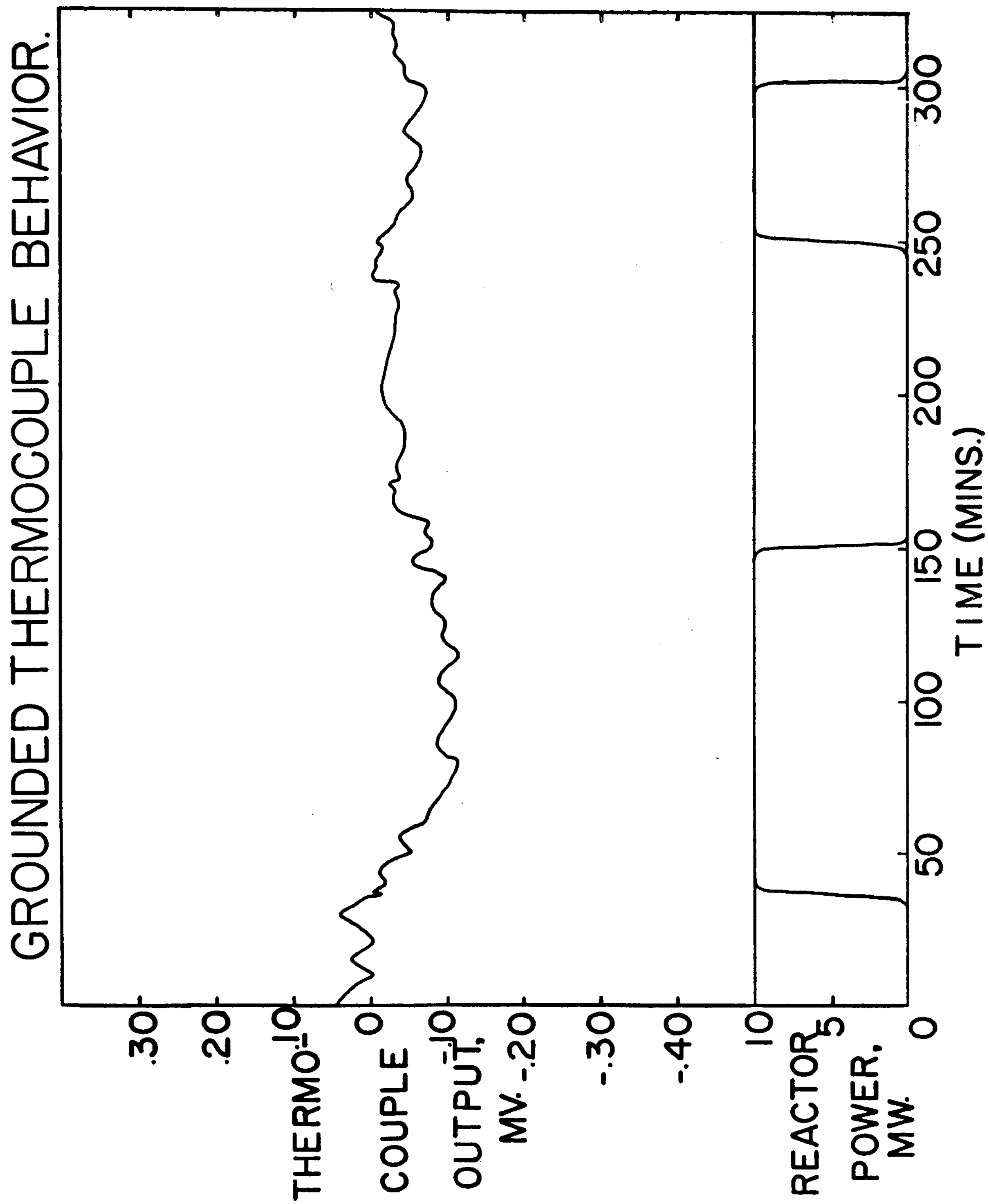

Fig. 3 Radiation-Induced Voltage Characteristic of Grounded Thermocouple Set with No Prior Irradiation History.

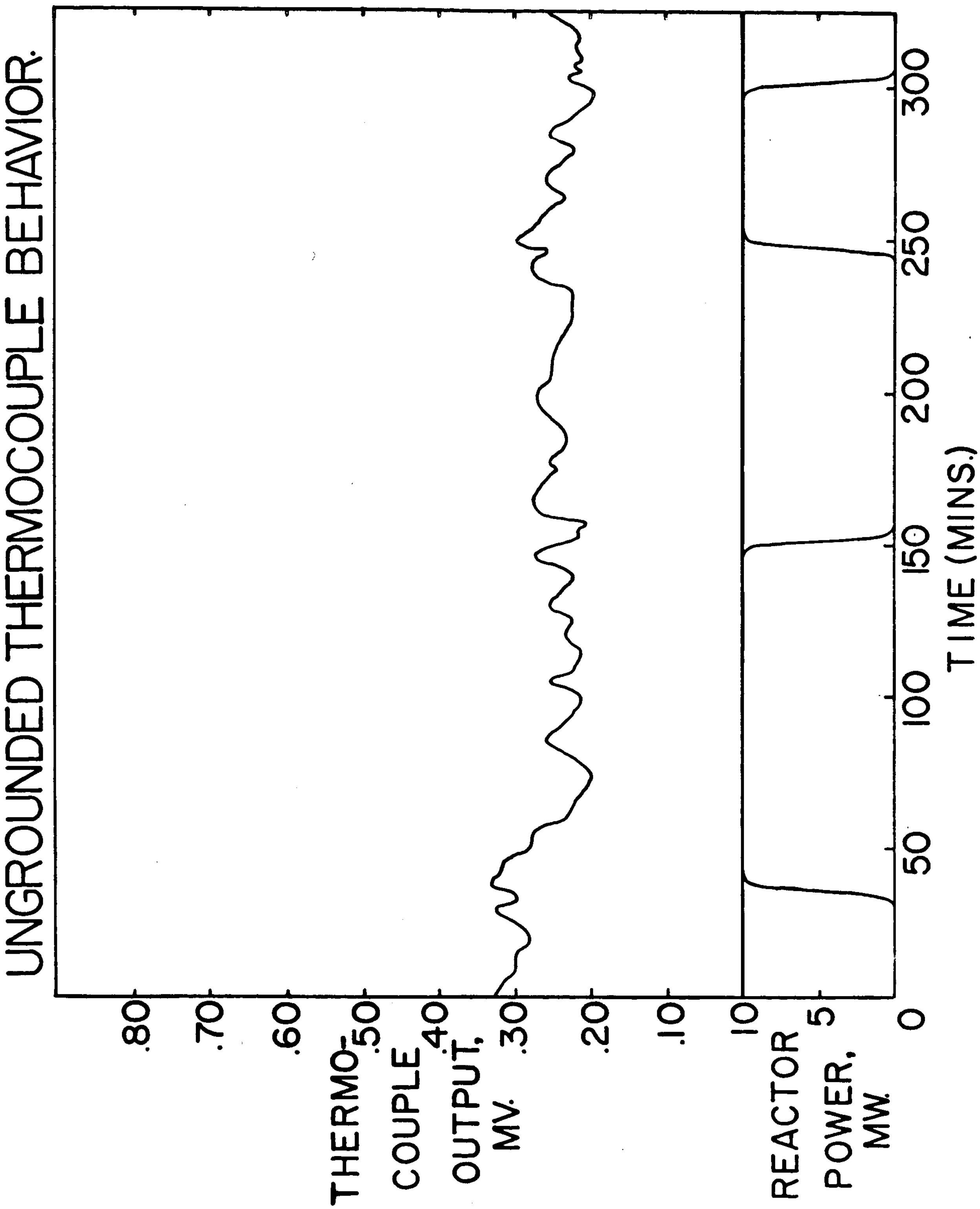

Fig. 4 Radiation-Induced Voltage Characteristic of Ungrounded Thermocouple Set with No Prior Irradiation History.

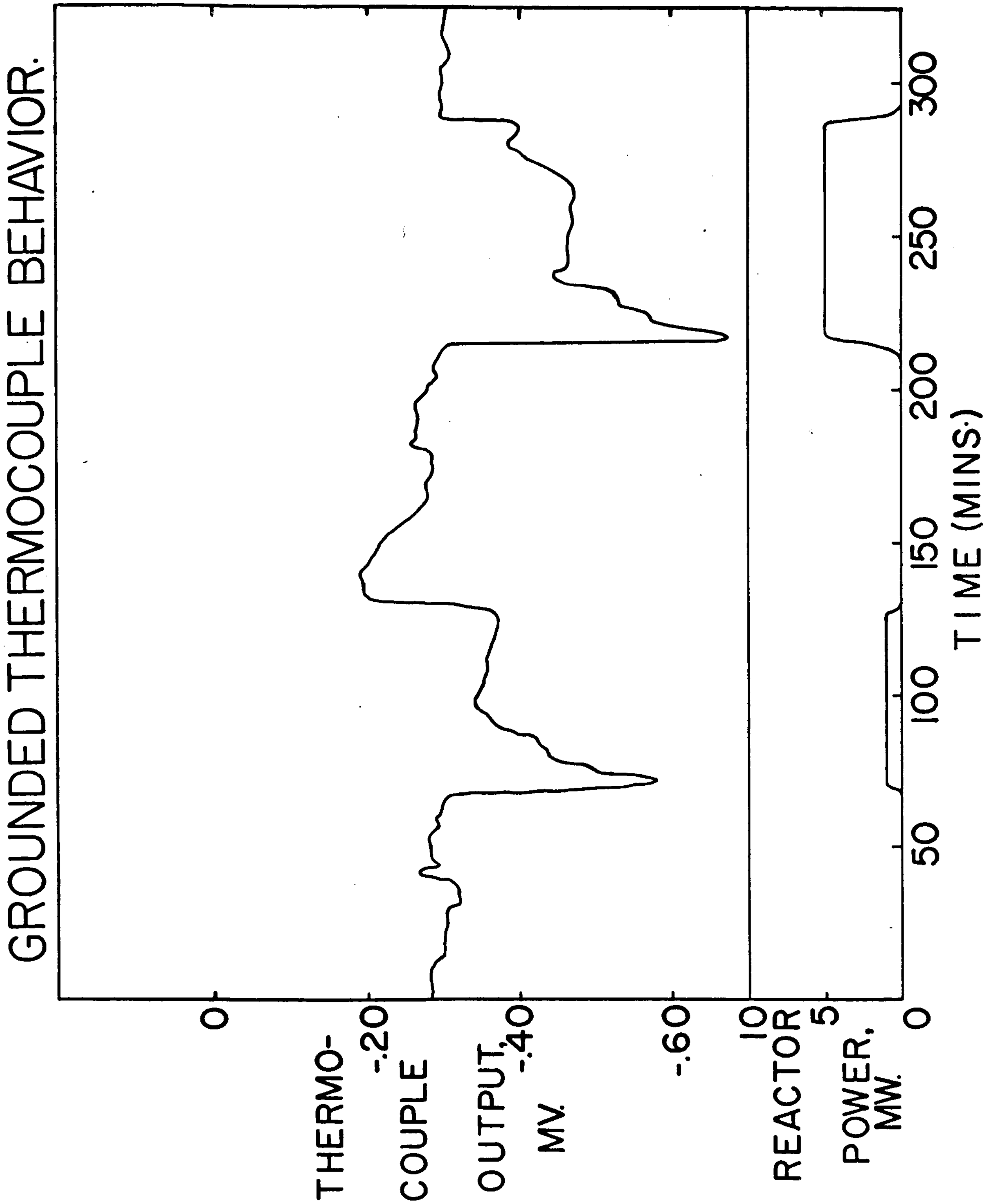

Fig. 5 Radiation-Induced Voltage Characteristic of Grounded Thermocouple Set after Several Cycles of Irradiation Exposure.

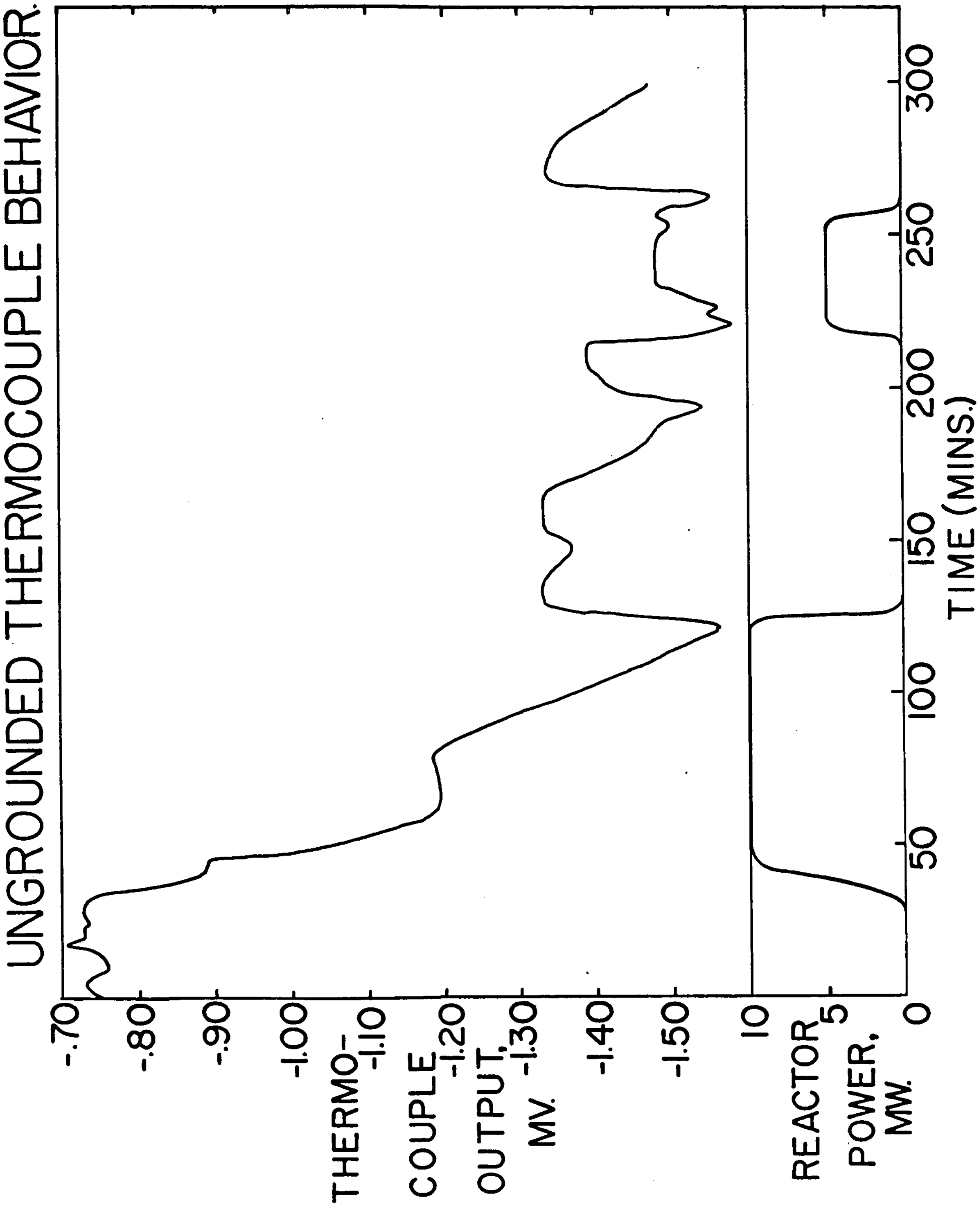

Fig. 6 Radiation-Induced Voltage Characteristic of Ungrounded Thermocouple Set after Several Cycles of Irradiation Exposure.

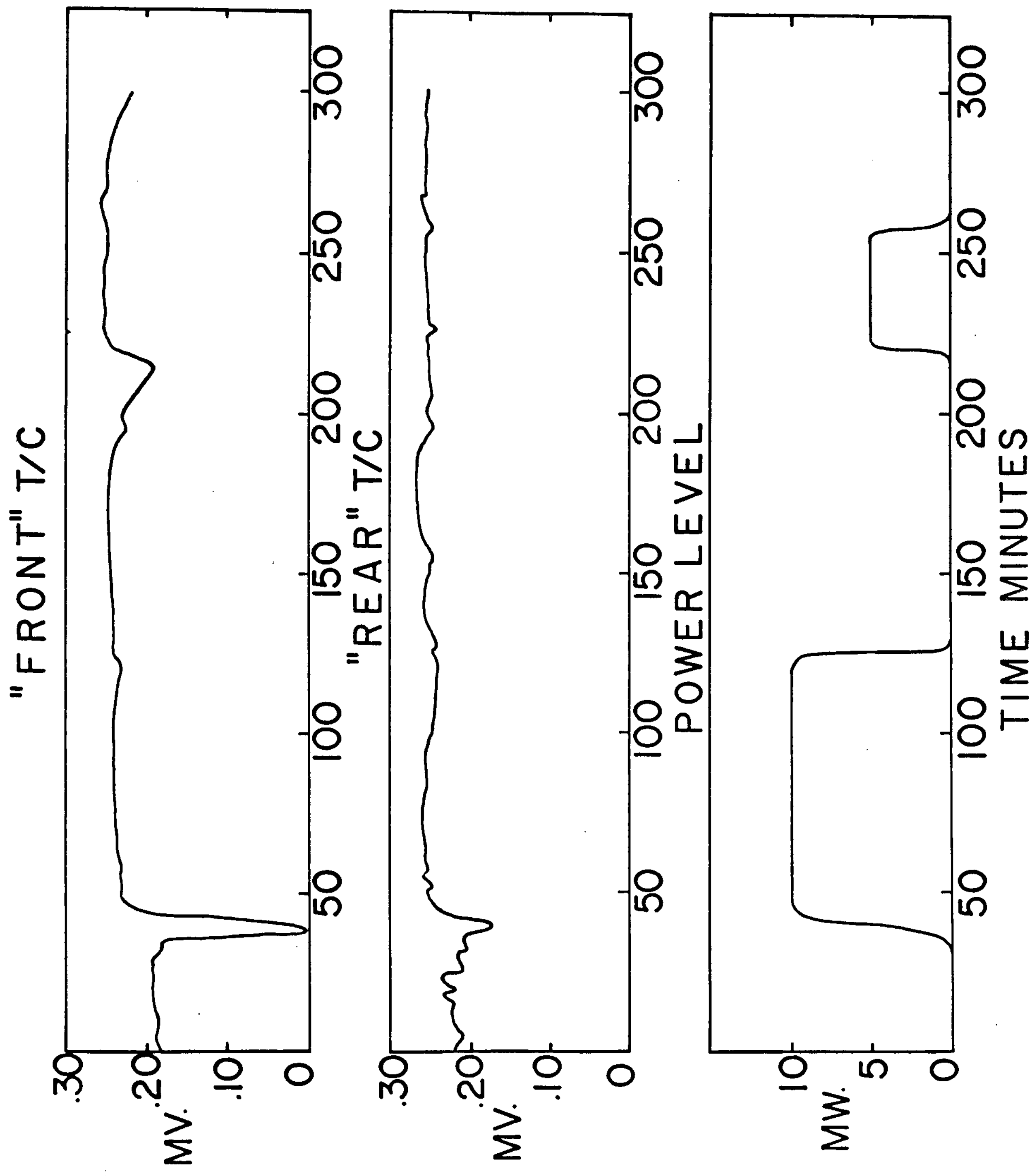

Fig. 7 Radiation-Induced Voltage Characteristics of Additional Grounded Thermocouples Measured Separately at Maximum Irradiation Exposure.

3) In subsequent cycles, rapid decreases in output voltage of upwards of 0.3 mv. occurred over periods of several minutes as maximum reactor power was reached, and a rapid increase to voltages sometimes exceeding initially measured values occurred when the reactor was shut down by manual scram (Figures 5 and 6).
4) The rapid changes noted above also occurred for thermocouples of different geometries (Fig. 7).
5) The rapid transients observed are reproducible and appear to be proportional to the power (and flux) levels attained because smaller transients were noted for the thermocouples in the "rear" tank (which experienced lower flux levels) and for both "front" and "rear" thermocouples when the reactor power was brought to 5 MW instead of 10 MW (Fig. 7).
6) There is some evidence that the rapid transients may respond to direction and magnitude of the change in reactor power level rather than to absolute reactor power (Figs. 3 and 4).

In evaluating these results, it is important to recognize that the observed effects are not attributable to transmutation in the thermoelectric materials because low cross section thermoelectric materials were used and because transmutation could not result in such rapid and reversible effects. Further, extraneous de-calibrating effects due to cold-working and inhomogeneities in the thermocouples are ruled out on the basis of the design of the experiment as indicated previously. Gamma heating in the vicinity of the junction also could not account for the observed effects because:

1) Similar results were noted for both grounded and ungrounded thermocouples.
2) The time response of the observed effects is much slower than the thermal time response of the thermocouples employed.
3) The direction of the voltage changes observed is opposite

to the direction of changes which would be caused by gamma heating near the "hot" junction.

Gamma heating in leads and junction boxes is also not thought to be responsible, because only relatively small increases (opposite to the effects of interest) in the output voltage resulted upon external application of heat to leads and junction boxes. Since the above factors apparently are not responsible, the results obtained must be attributable to direct influence of radiation upon the emf produced by the junction of the two dissimilar materials.

Because direct radiation effects have apparently not been conclusively observed previously despite extensive use of thermocouples in reactors, it is important to reconcile previous pertinent work with the present findings. Factors which may account for the differences include:

1) Thermocouples used in high radiation fields (to monitor fuel element temperatures, for example), experience real temperature changes when power level is changed which are large enough to mask any direct radiation effects of the magnitude found in the present work.
2) Relatively low radiation levels obtain for most thermocouple installations (monitoring reactor coolants, for example) in which real temperature changes with power level are small enough that direct radiation effects might otherwise be detectable.
3) In applications such as pulsing, reactors where large radiation transients might be expected to reveal direct radiation effects on thermocouples, the large temperature changes involved may mask any direct effects, and also the duration of the radiation pulse is short relative to the time characteristics of the presently observed effects.
4) Virtually all previous studies of radiation effects on thermocouples reported in the literature[1,5] involved only post-irradiation calibrations rather than in-pile calibration during irradiation. Levy and Co-workers[6]

attempted in-pile calibration but experienced temperature control problems which apparently precluded conclusive observation of direct radiation effects.

5) The calibration temperature employed in the present work (100°C) was lower than in most reactor environments, which may result in enhancement of direct radiation effects.

6) In the present work, the sheathed thermocouples were oriented with their central axis normal to the direction of the incident radiation, whereas parallel orientation obtains in many reactor installations.

7) Transient effects on thermocouple output may have been observed previously but dismissed as gamma heating effects or other extraneous phenomena.

It is noteworthy that Madsen[7] reported rapid and unexplained changes in thermocouple output during irradiation in the BEPO facility, and Cunningham and Goldthwaite[8] recently reported significant decreases in thermocouple outputs soon after start of irradiation. Recent private conversations with other investigators also indicate that transient radiation effects may have been observed but not reported in published work.

The scope of this work is not yet extensive enough to identify the mechanism involved. It is presently thought, however, that the causative mechanism may be one or more of these factors:

- Liberation of free electrons by ionizing radiation in leads, sheath, and insulators. This is the mechanism cited by Dau and Davis[9] as contributing to radiation-induced changes in alumina insulation resistance and by Terry and Co-workers[10] as the cause of radiation-induced transient signals in electrical cables.
- Ionization-induced changes in electrical conductivity. For example, Lark-Horowitz[11] found that 230 kev electrons produced substantial instantaneous and reversible increases in the conductivity of n-type germanium.

- In terms of solid state theory, radiation may affect electron mobility, particle scattering mechanism, Fermi level, and lattice thermal conductivity as discussed by Krumhansl[12].
- Other solid state radiation effects such as production of interstitials, vacancies and "knock-on" events with the concomitant introduction of new energy levels, as predicted by Heikes[3].

CONCLUSIONS

The magnitudes of the radiation-induced transient changes in thermocouple voltage output found in this work correspond to errors in measured temperature of 15°F and more in some instances. Errors of this magnitude may be large enough to be of considerable significance in interpretation of in-pile thermocouple data. This is particularly true in the many applications (such as heat flux, conductivity, and coolant temperature rise measurements in-pile) where the difference between temperatures at two or more locations experiencing different radiation levels is the quantity of interest.

Direct radiation effects on calibration are of most concern in those applications where thermocouples are used in protective circuits or otherwise for control purposes. The findings of the present investigation indicate that direct radiation effects can result in apparent temperature indications lower than actual temperature, which could lead to unconservative operating conditions in some instances. As a minimum precautionary measure, in-pile calibrations should therefore be performed whenever possible before reliance is placed on data obtained from thermocouples exposed to appreciable radiation levels.

The scope of work performed thus far does not permit definitive statements regarding either the mechanism involved or the firm prediction of conditions under which important direct radiation effects would be expected. Additional experiments are needed to determine the influence of such parameters as thermocouple composition, geometry, and orientation, temperature, rate of change of power level, and of the relative influence of gamma radiation and neutrons. If further experiments are successful in isolating the influential parameters, the causative mechanism may be identified. This might suggest a way to design special thermocouples to minimize direct radiation effects. It is also speculated that eventually advantage might possibly be taken of such direct radiation effects to improve the efficiency of direct nuclear energy conversion devices employing thermoelectric elements.

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