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Final Report, Phase 2
DESIGN AND DEVELOPMENT OF
FAST-RESPONSE THERMOPILE-TYPE
NEUTRON-FLUX-SENSITIVE DEVICE

ATL Job 4041

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ABSTRACT

This program has been directed toward development of a thermopile-type neutron-flux-sensitive meter of small physical size and having a fast response time (25 milliseconds) and a high signal output. The meter configuration selected consists essentially of a metallic heat sink, a thermopile, and a thin layer of neutron-sensitive material in series. When the meter is exposed to a neutron environment, heat generated in the neutron-sensitive material flows across the thermopile unit to the heat sink. The signal output generated by the thermopile is proportional to the heat flow, which is in turn proportional to the neutron flux. Signal-output characteristics of meters of this configuration have been verified by experimental testing in the Battelle Research Reactor, the General Electric NTR Reactor, and the Materials Testing Reactor (Idaho Falls). A practical neutron meter, in which the sensing element measures approximately $1/8" \times 1/2" \times 1"$, with a time constant of less than 25 milliseconds and a millivolt-level signal output at a thermal-neutron flux of 10^{12} n/cm²-sec has been achieved, thus substantially fulfilling the program objectives.

INTRODUCTION

An instrument which could measure the local thermal-neutron-flux intensity in a reactor would be very useful. Not only would such a device be desirable for instrumentation purposes, but it would also serve well as an actuator for an upper-power-limit alarm. Ideally, such a device would be small in size and have a reasonably fast response time.

On the basis of an AEC contract award, Advanced Technology Laboratories (ATL), a Division of American Radiator & Standard Sanitary Corporation, initiated a development program for a thermopile-type neutron-flux meter with the objective of satisfying the following specifications:

- 1) The meter shall be capable of operating in a neutron flux ranging from 10^{10} to 10^{12} n/cm²-sec.
- 2) The response time shall be 25 milliseconds or less.
- 3) The useful life shall be 10,000 hours.
- 4) The meter shall be capable of operating in an environment characterized by temperatures up to 1000°F.
- 5) The meter output shall be sufficient to allow use of standard commercial indicating and recording devices.

DISCUSSION

To date, neutron-flux intensity has been measured primarily by fission chambers, although fission-heated thermocouples and thermopiles have recently proved to be satisfactory. In general, fission chambers are large, have fast time response, and require high-voltage supplies and high-gain amplifiers. Fission-heated thermocouples and thermopiles are generally characterized by comparatively small physical size, high signal output, and reasonably slow time response. While the fission chambers possess the necessary response characteristics, their physical size prevents their use in many areas where local flux measurements are desired. On the other extreme, the response times of fission-heated thermocouples and thermopiles are generally too slow to permit instantaneous detection of flux variations. However, both of these devices find widespread usage as average neutron-flux instruments.

A. Design Approach

The object of the effort reported here was to develop a neutron-meter configuration having the desirable features of both fission chambers and fission-heated thermocouples and thermopiles. The basic principle of the thermopile approach, with its high-signal-output capabilities, was therefore retained. The design approach used* (Figure 1) allows a considerable reduction of thermal mass, so that the fast response characteristics required can be obtained.

In theory, the neutron-flux meter functions are as follows. In a neutron field, the interaction between the neutron-sensitive material, such as B^{10} or U_2O_8 , and the neutrons generates heat. Since one face of the meter is perfectly insulated, the only path available for heat flow is across the low-conductivity material. A temperature drop therefore exists across this path in accordance with the heat-conductance equation

$$\frac{q}{A} = -K \frac{\Delta T}{L},$$

where q = heat rate,

A = cross-sectional area,

K = thermal conductivity,

ΔT = temperature drop,

L = path length.

* U. S. Patent No. 3,028,494, Advanced Technology Laboratories, a Division of American-Standard.

SCHEMATIC OF
THERMOPILE-TYPE HEAT METER* FOR
SENSING NEUTRON FLUX

FIGURE 1

* U.S. Patent No. 3,028,494, Advanced Technology Laboratories, a Division of American-Standard.

Since A , K , and L are fixed values, ΔT is proportional to q , which is in turn proportional to the neutron-flux intensity. By utilizing thermocouples on each face of the low-conductivity layer, an emf signal proportional to ΔT and representing the neutron-flux density can be obtained. A large metallic heat sink is used to absorb the neutron-generated heat. Since the thermocouples measure only the temperature difference caused by heat flow, and since the heat flow is a function of the neutron flux, the temperature of the components and of the surrounding area has essentially no effect on the output.

If the thickness, L , of the low-conductivity material is reduced, the thermal mass of the system is reduced and a faster time response is achieved; however, the ΔT and the emf signal output are thereby decreased. Since the L dimension will be dictated by the time-response requirement of 25 milliseconds, the only alternative way to provide the required signal output is to increase the number of thermocouple junctions (i. e., utilize a thermopile). Therefore, the design choice entails selecting the dimension L and the number of thermocouple junctions to satisfy the time-response and signal-output requirements.

B. Response Characteristics

In order to estimate the required thickness, L , of the low-conductivity material, the analysis presented below was conducted on the proposed design approach. Physically, the cross section of the design may be considered to be as shown in Figure 2.

CROSS SECTION OF METER DESIGN

FIGURE 2

The thermocouples contribute very little to the over-all time lag of the system. They can therefore be ignored in this approximation. The physical system for analysis is then as shown in Figure 3a; the equivalent electrical diagram is shown in Figure 3b.

In Figure 3b,

q/A = heat flux,

$C_B/2$ = heat capacity of neutron-sensitive material,

C_Q = heat capacity of low-conductivity material,

$R_B/2$ = thermal resistance (to heat flow) of neutron-sensitive material,

$R_Q/2$ = thermal resistance of low-conductivity material,

R_C = contact resistance,

C_S = heat capacity of heat sink (assumed infinite).

If only unit areas and heat fluxes are considered, q/A and A can be arbitrarily set to equal 1.

Writing the heat-balance equation between points 1 and 3:

Heat in = Heat conducted + Heat stored, or

$$\frac{q}{A} = 1 = \frac{t_1 - t_3}{\frac{R_Q}{2} + R_C + \frac{R_B}{2}} + \frac{C_B}{2} \frac{dt_1}{d\tau} \quad (1)$$

where t is temperature and τ is time, which reduces to

$$\frac{dt_1}{d\tau} + B t_1 - B t_3 = G \quad (2)$$

where $G = 2/C_B$,

$$B = 4/C_B (R_Q + 2R_C + R_B).$$

Writing a similar heat-balance equation between points 3 and 4:

$$\frac{t_1 - t_3}{\left(\frac{R_Q}{2} + R_C + \frac{R_B}{2}\right)} = \frac{t_3 - t_4}{\left(\frac{R_Q}{2} + R_C\right)} + C_Q \frac{dt_3}{d\tau} \quad (3)$$

Consider that t_4 = ambient temperature and is set arbitrarily at 0. Equation 3 then reduces to:

a. Physical Diagram

b. Equivalent Electrical Diagram

PORTION OF SYSTEM ANALYZED FOR
RESPONSE CHARACTERISTICS

FIGURE 3

$$\frac{dt_3}{d\tau} + D t_3 - E t_1 = 0 \quad (4)$$

$$\text{where } D = \frac{2(2R_Q + 4R_C + R_B)}{C_Q(R_Q + 2R_C + R_B)(R_Q + 2R_C)}$$

$$E = \frac{2}{C_Q(R_Q + 2R_C + R_B)}$$

Equations 2 and 4 are two simultaneous equations describing the transient heat-conduction phenomena in the neutron meter. Utilizing the LaPlace transformation technique:

$$\frac{dt_1}{d\tau} + B t_1 - B t_3 = G \rightarrow s T_1 + t_1(0) + B T_1 - B T_3 = \frac{G}{s} \quad (5)$$

$$\frac{dt_3}{d\tau} + D t_3 - E t_1 = 0 \rightarrow s T_3 + t_3(0) + D T_3 - E T_1 = 0 \quad (6)$$

Considering zero initial conditions and solving for T_3 ,

$$T_3 = \frac{GE}{s(s-x)(s-y)} \quad (7)$$

$$\text{where } x, y = \frac{-(B+D) \pm \sqrt{(B+D)^2 - 4B(D-E)}}{2}$$

Using the Inverse LaPlace Transformation:

$$t_3 = \frac{GE}{xy} \left[1 + \frac{x e^{yt} - y e^{xt}}{(y-x)} \right] \quad (8)$$

By a similar consideration, the solution for t_1 is:

$$t_1 = G \left[\frac{D}{xy} - \frac{D+x}{x(y-x)} e^{xt} + \frac{D+y}{y(y-x)} e^{yt} \right] \quad (9)$$

Since the same quantity of heat is transferred through each surface,

$$\left(\frac{q}{A} \right)_{1-2} = \frac{t_1 - t_2}{R_C + \frac{R_B}{2}} = \left(\frac{q}{A} \right)_{2-3} = \frac{t_2 - t_3}{\frac{R_Q}{2}} \quad (10)$$

and solving for t_2 :

$$t_2 = \frac{1}{R_C + \frac{R_B}{2} + \frac{R_Q}{2}} \left[\frac{R_Q}{2} t_1 + \left(R_C + \frac{R_B}{2} \right) t_3 \right] \quad (11)$$

where t_3 , t_1 are represented by equations 8 and 9.

Substituting values of t_1 and t_3 :

$$t_2 = \frac{1}{\left(R_C + \frac{R_B}{2} + \frac{R_Q}{2} \right)} \left[\frac{G}{xy} \left\{ \frac{DR_Q}{2} + E \left(R_C + \frac{R_B}{2} \right) \right\} + \frac{Ge^{yt}}{y(y-x)} \left\{ \frac{R_Q}{2} (D+y) + \left(R_C + \frac{R_B}{2} \right) E \right\} - \frac{Ge^{xt}}{x(y-x)} \left\{ \frac{R_Q}{2} (D+x) + \left(R_C + \frac{R_B}{2} \right) E \right\} \right] \quad (12)$$

This expression for the temperature difference ($t_2 - t_4$) across the thermopile unit is rather unwieldy. Fortunately, on closer analysis of the physical dimensions and material properties of interest, the second term of equation 12 is quite small and can be neglected when compared to the third term. Also, the coefficient of the first term and that of the third term are sufficiently similar that an approximate solution for t_2 (which is equal to $t_2 - t_4$, since t_4 is arbitrarily set at zero) is of the form

$$t_2 = A \left[1 - e^{-xt} \right] \quad (13)$$

where x is essentially the predominant time constant of the system.

$$x = \frac{-(B+D) + \sqrt{(B+D)^2 - 4B(D-E)}}{2} \quad (14)$$

$$\text{where } B = \frac{4}{C_B(R_Q + 2R_C + R_B)}$$

$$D = \frac{2(2R_Q + 4R_C + R_B)}{C_Q(R_Q + 2R_C + R_B)(R_Q + 2R_C)}$$

$$E = \frac{2}{C_Q(R_Q + 2R_C + R_B)}$$

Substituting the physical constants into equation 14 gives an approximate estimate of the value of proposed neutron-meter response time. For example, use of a 0.007-inch-thick piece of glass, a 0.001-inch-thick layer of boron, and no contact resistance results in an estimated time constant of 33 milliseconds, which is reasonably close to the desired 25 milliseconds. The variation of the B^{10} thickness has little or no effect on the resultant time constant, since the predominant time lag was constituted by the glass structure. By comparison, the inclusion of a contact resistance of the order of $R_C = 1/2000 \text{ Btu/hr-ft}^2\text{-}^\circ\text{F} = 0.0005$ increased the calculated value of the time constant to approximately 75 milliseconds. This indicates the vital necessity for minimizing contact resistance.

C. Signal-Output Characteristics

The signal-output characteristics of the neutron-flux meter are estimated in the following fashion. The number of atoms per cubic centimeter of B^{10} is given by:

$$N = \frac{\rho N_o}{A}, \quad (15)$$

where ρ = density = 2.3 gm/cm^3 ,

N_o = Avogadro's Number = $6.02 \times 10^{23} \text{ atoms/gm-atom}$,

A = atomic number = 10 gm/gm-atom ;

therefore,

$$N_{B^{10}} = 0.138 \times 10^{24} \text{ atoms/cm}^3.$$

For neutron capture in a 0.001-inch layer of B^{10} ,

$$\text{Reduction of Neutron Flux} = 1 - e^{-N\sigma X}, \quad (16)$$

where $N = 0.138 \times 10^{24} \text{ atoms/cm}^3$,

$\sigma = B^{10}$ cross section = 3990 barns,

X = attenuation length $\cong 0.001 \text{ in.} \cong 0.00254 \text{ cm}$.

Therefore,

Reduction of Neutron Flux = 75%.

Heat release for a maximum flux of 10^{12} n/cm²-sec is calculated by:

$$\text{Number of Collisions} = (0.75) (10^{12}/\text{cm}^2\text{-sec});$$

$$\text{Heat-Release Rate} = (0.75 \times 10^{12} \text{ collisions/cm}^2\text{-sec}) (2.5 \text{ Mev/collision})$$

$$(1.6 \times 10^{-6} \text{ erg/Mev}) (10^{-7} \text{ joule/erg}) (9.5 \times 10^{-4} \text{ Btu/joule})$$

$$(3600 \text{ sec/hr}) (930 \text{ cm}^2/\text{ft}^2) = 950 \text{ Btu/hr-ft}^2.$$

This heat rate flowing across the thermopile unit will cause a temperature difference of the following value to exist across the hot and cold junctions:

$$\Delta T = \frac{q/A}{K} \Delta X = \frac{(950 \text{ Btu/hr-ft}^2)(0.000666 \text{ ft})}{(0.5 \text{ Btu/hr-ft-}^\circ\text{F})} = 1.26^\circ\text{F},$$

where q/A = heat rate (950 Btu/hr-ft²),

ΔX = distance between hot and cold junction (0.008 in. or 0.000666 ft),

K = thermal conductivity of glass (0.5 Btu/hr-ft-°F).

Consequently, for a silver-constantan thermocouple in which the emf output is approximately 0.022 mv/°F, the signal output is about 2.2 mv, if 80 active junctions are used.

The desired useful life of the meter is 10,000 hours. The calculated burnup of B^{10} during this time is estimated as follows (ref. equation 15). For a 0.001-inch (0.00254-cm) thickness, the number of particles of B^{10} is

$$N_{B^{10}} = 0.138 \times 10^{24} \text{ atoms/cm}^3 \times 0.00254 \text{ cm} = 3.5 \times 10^{20} \text{ atoms/cm}^2.$$

Assuming an average collision rate of 10^{11} /cm²-sec,

$$\begin{aligned} \text{Total burnup in 10,000 hours} &= (10^{11}/\text{cm}^2\text{-sec})(10,000 \text{ hr})(3600 \text{ sec/hr}) \\ &= 3.6 \times 10^{18}/\text{cm}^2. \end{aligned}$$

Percent burnup is then $\frac{3.6 \times 10^{18}}{3.5 \times 10^{20}} \cong 1\%$, indicating that the output signal would be reasonably constant over the total meter life.

D. Procedure for Meter Fabrication

Several different methods and materials for obtaining a good thermal bond between the thermopile section and the heat sink of the meter configuration shown in Figure 1 were reviewed and their possible utilization investigated. These included:

- 1) Flame spraying a metallic heat sink directly onto the thermopile unit.
- 2) Using metal-to-glass sealing techniques similar to those employed in sealing electronic tubes.
- 3) Using beryllium oxide for the heat sink.
- 4) Using an amalgam-type filler material for the heat sink.

Critical analysis led to the conclusion that the direct glass-to-metal seal had the greatest potential for solving the problem. Several techniques for using this method in the fabrication of a meter were therefore tried. Satisfactory results were finally achieved using oxidized 416 stainless steel for the heat sink and Corning No. 0211 glass. The various steps in the fabrication process are shown in Figure 4, and complete details of the procedure are developed as follows.

1. Thermopile Preparation

The body of the thermopile unit is prepared by cutting a rectangular section of Corning No. 0211 drawn 0.008-inch-thick micro-sheet glass, as shown in Step 1, Figure 4. This glass plate is then wrapped with 75 to 80 turns of Constantan ribbon measuring 0.001 inch thick by 0.005 inch wide (see Step 2, Figure 4). Fast-drying clear cement is used to secure the wires at each end and prevent unwinding. Ribbon was employed, rather than wire, to provide more substantial leads.

Preparation for and the silver plating of the lower half of the Constantan ribbon to form the thermocouple junctions is rather critical. The portion of the wrapped unit that is not to be silver plated is coated with a commercial stop-off lacquer and placed in a fixture that makes electrical contact with each turn of the Constantan wire. This assembly is placed in an acid solution for 10 seconds. The purpose of the acid dip is to clean, etch, and activate all surfaces to be plated. The acid solution is composed

STEP 1 - CENTER GLASS SHEET
(CORNING NO. 0211)

STEP 2 - WINDING 0.001" $\times$ 0.005" RIBBON

STEP 3 - SILVER PLATING

STEP 4 - OUTER GLASS SHEETS
(CORNING NO. 0211)

STEP 5 - FUSING

FUSE UNDER
PRESSURE AT $\sim 1500^{\circ}\text{F}$

STEP 6 - APPLYING B¹⁰

CONSTRUCTION OF NEUTRON-FLUX METER*

FIGURE 4

* U. S. Patent 3,028,494, Advanced Technology Laboratories, a Division of American-Standard.

of 2 parts of reagent-grade sulphuric acid, 1 part of reagent-grade nitric acid, and 1 part water. The assembly is removed from the acid dip, rinsed in distilled water, placed in weak potassium cyanide solution, rinsed again in distilled water, and placed in the silver-strike tank. The purpose of the cyanide dip is to quench the acid action and carryover.

The silver-strike bath has the following composition per gallon: potassium silver cyanide, 1.0 oz avdp; potassium cyanide, 11 oz avdp. The assembly is placed in the silver-strike tank for 3 minutes at 5 ma and $1\frac{1}{2}$ vdc. The assemblies are then transferred to the silver-plating bath for 2 hours at 1 ma and $1\frac{1}{2}$ vdc.

The silver-plating solution has the following composition per gallon: potassium silver cyanide, 9.0 oz avdp; potassium cyanide, 14.0 oz avdp; potassium carbonate, 3.0 oz avdp; Wilbro Brightever Make-up "A" (available from Wildberg Bros., San Francisco), 230 ml. The units are then rinsed in cold distilled water for about 1 minute, followed by rinsing in hot running water for several minutes. This procedure results in a very adherent, smooth silver plate approximately 0.0005 inch thick. Since approximately this amount of material is removed by the acid-etch step, the resultant wire is of uniform dimensions over both the plated and unplated portions.

2. Bonding of Thermopile to Heat Sink

The wrapped thermopile unit is bonded to the heat sink in four basic steps:

a. The metallic heat sink, of 416 stainless steel, is prepared by polishing one surface and then pre-oxidizing. The oxidizing is accomplished by placing the metal in a wet-hydrogen furnace at about 1700°F for $\frac{1}{2}$ hour and allowing to air cool in the atmosphere. The result is a thin, tenacious film (primarily of chromium oxide) on the surface.

b. The thermopile unit is prepared for bonding by stripping the stop-off lacquer from the unit and cleaning first in acetone and then in alcohol. Since the acetone also removes the cement used to secure the wires on each end, the cement is reapplied. This step is shown in Figure 4, Step 3.

c. Two pieces of Corning No. 0211 glass, 0.002 inch thick, are cut to the dimensions shown in Figure 4, Step 4.

d. These prepared pieces are assembled as shown in Figure 4, Step 5, and put in a fixture prior to fusing of the various pieces. The fixture serves three purposes: 1) positioning the pieces properly, 2) applying sufficient weight to compress the pieces together during the heating and bonding, 3) providing a large thermal mass around the thermopile unit so that the glass will cool slowly through the annealing range to the strain point. The loaded fixture is then placed in a hydrogen-atmosphere furnace and fired to about 1500°F. The firing requires about 15 minutes to complete, because the fixture must be at the desired temperature for from 3 to 5 minutes to insure good fusion of the laminations. The fixture is then placed in a cool portion of the furnace until it reaches about 500°F, at which point it is withdrawn and allowed to finish cooling in the room atmosphere.

e. The final step in fabrication of the transducer is application of the B^{10} as shown in Figure 4, Step 6. Finely ground B^{10} powder is mixed with turpentine and oil to form a slurry, which is then brushed onto the transducer face. The transducer is placed in a 700°F oven for 10 minutes, baking off the turpentine and oil. The result is an even film of B^{10} 0.001 to 0.002 inch thick on the surface.

The completed neutron-meter is as shown in Figures 5a and 5b.

E. Time-Response Evaluation

The response characteristics of the neutron meter were evaluated through use of thermal methods by mounting the meter on the tip of a large aluminum heat sink and immersing the tip suddenly in hot water. The resulting signal output was registered on a Sanborn Recorder. Obviously, this approach does not quite impose a step temperature input. However, in the period of interest, the initial phase, a step input is approximated. The fixture used is shown in Figure 6; typical results are shown in Figure 7, which indicates a time response of about 15 milliseconds.

F. Thermopile Signal-Output Characteristics

Thermal-simulation methods were used to check for the existence of a good thermopile and to investigate signal-output characteristics of the neutron meter. The test fixture for this study (Figure 8) consisted of a large aluminum rod with an electrofilm-type heater potted to one end and the other end attached to a large heat sink. The neutron meter is inserted midway in the rod. Heat generated by the electrofilm must flow

a. Neutron-Meter Assembly

b. Completed Neutron Meter

COMPLETED NEUTRON METER

FIGURE 5

TIME-RESPONSE TEST FIXTURE

FIGURE 6

TYPICAL RESULTS FOR
TIME-RESPONSE TEST OF
NEUTRON METER

FIGURE 7

FIXTURE FOR SIGNAL-OUTPUT TESTS

FIGURE 8

through the rod to the heat sink and, in so doing, must flow through the neutron meter. A signal output proportional to the heat flow is thereby produced. The heat flow rate is determined by measuring the power input to the electrofilm heater. Figure 9 illustrates signal output versus heat input of a typical neutron meter. The slight curvature is due to heat loss by radiation and convection from the test unit.

G. Experimental Testing

During the course of development, three series of tests were performed at three different reactor facilities. Each test series and the results are described below.

1. Battelle Research Reactor

The initial tests were conducted at the Battelle Research Reactor (Columbus, Ohio). This reactor was selected because of its availability and the general ease with which the meters under test could be inserted into the reactor. The test location was an unused fuel-pin location within the reactor core. Maximum neutron flux at this location was estimated to be 5×10^{12} n/cm²-sec (unperturbed) at a reactor power level of 2 megawatts. Flux variations were obtained by changing the reactor power level.

The meter configuration used during these tests differed slightly from the description in Section D. The major differences were that an aluminum heat sink was used and the thermopile unit was bonded to the heat sink by liquid-silver cement. Also, 1-mil diameter Constantan wire was used instead of the 0.001" x 0.005" Constantan ribbon.

The purpose of this test series was to observe the general signal-output characteristics of the neutron meters. Of particular interest were the magnitudes of the signal and the behavior of the signal output with flux changes and time. In general, some of the results were as expected and others were difficult to explain.

The fixture used for these tests is shown in Figure 10, and results are illustrated by Figures 11 through 16. In Figure 11, some typical signal outputs as a function of reactor power (and flux levels) are shown. Meters B5-II, B4, and B1 indicated the desired signal magnitudes, while B3 was considerably lower. Figures 12 through 16 illustrate signal output as a function of time and of reactor power. Figure 12, the test results of meter B1, illustrates the expected type of performance; the signal level is linear with reactor power and is reasonably constant with time at any fixed reactor

TYPICAL SIGNAL-OUTPUT VERSUS HEAT-INPUT CHARACTERISTICS
OF NEUTRON-FLUX-METER THERMOPILE

FIGURE 9

NEUTRON-FLUX-METER TEST FIXTURE
BATTELLE RESEARCH REACTOR TESTS

FIGURE 10

TYPICAL NEUTRON-METER SIGNAL-OUTPUT CHARACTERISTICS
 BATTELLE RESEARCH REACTOR TESTS

FIGURE 11

NEUTRON METER B1
 SIGNAL OUTPUT VERSUS TIME AND REACTOR POWER
 BATTELLE RESEARCH REACTOR TESTS

FIGURE 12

NEUTRON METER B2
SIGNAL OUTPUT VERSUS TIME AND REACTOR POWER
BATTELLE RESEARCH REACTOR TESTS

FIGURE 13

NEUTRON METER B3
 SIGNAL OUTPUT VERSUS TIME AND REACTOR POWER
 BATTELLE RESEARCH REACTOR TESTS

FIGURE 14

NEUTRON METER B4
 SIGNAL OUTPUT VERSUS TIME AND REACTOR POWER
 BATTELLE RESEARCH REACTOR TESTS

FIGURE 15

NEUTRON METERS B5-I AND B5-II
SIGNAL OUTPUT VERSUS TIME AND REACTOR POWER
BATTELLE RESEARCH REACTOR TESTS

FIGURE 16

power setting. Figure 13, 14, and 15 illustrate the signal versus time and reactor power for meters B2, B3, and B4. In these meters, the signal output tended to decrease with time and constant reactor power, and, furthermore, had a negative output on reactor shutdown. It was suspected that the liquid-silver bond between the thermopile and the heat sink was the primary source of these peculiar results. Poor bonding is due to the drying characteristics of liquid-silver cement, which contracts during drying, leaving a certain amount of void space. This void space acts as a large contact resistance to heat flow from the thermopile to the heat sink. If only a small amount of voids occurs then a reasonably good bond between the thermopile and the heat sink is obtained, resulting in signal characteristics such as typified by meter B1, Figure 12. If large amounts of voids occur, then a relatively poor bond is obtained and signal characteristics such as those in Figures 13, 14, and 15 result.

Meters B5-I and B5-II were tested to evaluate this theory. Meter B5-I was fabricated in the usual fashion except that an aluminum-paste cement similar in nature to the liquid-silver cement and somewhat superior was used to bond the thermopile to the heat sink, while meter B5-II was fabricated using a silicone grease. Since the test was to be of a relatively short duration, the silicone grease would be more than adequate. Figure 16 illustrates the test results. The signal-output characteristics of meter B5-II were linear with reactor power and constant with time, whereas those of meter B5-I exhibited the previous decay with time.

Since this test proved the deficiency of liquid-metallic cements, the next phase was devoted to seeking better methods of bonding the thermopile to the heat sink. The various techniques considered were:

- a) Flame-spraying a metallic heat sink directly onto the thermopile unit.
- b) Using metal-to-glass sealing techniques.
- c) Using beryllium oxide heat sinks.
- d) Using amalgam-type filler material.

After due consideration, method b, direct metal-to-glass sealing, was selected as offering the most practical solution to the problem. The technique used is as described in Section D.

2. General Electric NTR Facility

A test series was conducted at the General Electric NTR facility (Vallecitos, California) using neutron meters fabricated as described in Section D, with the metal-to-glass bond technique. Unfortunately, none of the meters tested had signal outputs greater than 150 microvolts. It was subsequently determined that these low signal outputs were caused by a) a poor boron (B^{10}) layer on the meters, and b) a lower than expected maximum neutron-flux environment (2.4×10^{11} n/cm²-sec) at the test location. No conclusions could be drawn from these tests.

3. MTR Facility

Since the maximum neutron flux available at the General Electric NTR facility proved to be below expectations, it was decided to conduct further tests at the MTR facility (Idaho Falls, Idaho). The MTR facility not only provided significantly higher neutron-flux environments but also allowed for long-term testing, which was not possible in the two previous reactor facilities.

The test location selected was a vertical-graphite (VG-48) hole where unperturbed neutron fluxes of approximately 4×10^{11} to 6×10^{12} n/cm²-sec could be obtained. Two neutron meters were tested during each MTR operating cycle for a period of about two weeks. Figure 17 illustrates a typical test arrangement. Each neutron meter is mounted in an individual capsule and suspended on a stainless-steel support cable. A lead weight is used to prevent "floating" due to the updraft cooling air in the VG hole. The electrical leads are taped to the stainless-steel cable and are connected to a Brown recording potentiometer. The stainless-steel cable is marked so that the distance from the top of the reactor to the meters may be easily ascertained. The unperturbed flux levels at the meter locations are then determined from the flux plot for VG-48. Figure 18 shows a typical test assembly before insertion into the VG hole.

A total of eight neutron meters were used during this test series. Unfortunately, the lead wires on four of these were severed in operational mishaps of one sort or another at the MTR facility. Test results on the remaining four units are shown in Figures 19, 20, and 21.

EXPERIMENTAL ARRANGEMENT
MTR NEUTRON-METER TESTS

FIGURE 17

TYPICAL NEUTRON-METER TEST ASSEMBLY
MTR TESTS

FIGURE 18

The signal output-time history of the meters is shown in Figures 19 and 20. Minor fluctuations in signal-output characteristics have been omitted for clarity, outputs shown being the average for the flow levels indicated. The signal output of meter No. 1 was terminated after 204 hours of operation by a broken lead wire, which resulted from the fluttering of the test capsules caused by the high velocity of the updraft cooling air. Larger weights could not be used because of the difficulty of insertion into the VG access holes. However, all the meters tested indicated reasonably constant output over the entire test period. These signal-output characteristics are as expected, and it is concluded that the bonding problem between the thermopile and the heat sink has been satisfactorily resolved.

Figure 21 shows the signal-output versus neutron-flux-level characteristics of the meters tested in the MTR. All the meters exhibit reasonably linear output. However, as is readily apparent, the output levels of the meters are significantly different, which may be attributed to two causes.

a) Dimensional Differences Due to Method of Fabrication

In the fusing process during fabrication, the distances between the hot and cold junctions of the thermopile may vary. Since signal output is directly related to this distance, a corresponding variation in the signal output will exist.

b) Heat-Loss Characteristic of Test Capsule

The signal output of the meters is a function of the heat flow across the thermopile unit. Ideal maximum signal outputs are obtained if all the heat generated in the boron layer flows across the thermopile to the heat sink. In the MTR tests, the high cooling-air velocity in the VG holes is suspected to have caused some air leakage into the test capsules, resulting in convective cooling from the surface of the boron layer, thus reducing the conductive heat flow across the thermopile and as a consequence lowering the signal output.

SIGNAL OUTPUT VERSUS TIME OF OPERATION
METERS 1 AND 2
MTR TESTS

FIGURE 19

SIGNAL OUTPUT VERSUS TIME OF OPERATION
METERS 3 AND 4
MTR TESTS

FIGURE 20

SIGNAL OUTPUT-NEUTRON FLUX CHARACTERISTICS
IRRADIATION TEST OF NEUTRON-FLUX METERS IN MTR
JULY-AUGUST 1962

FIGURE 21

CONCLUSIONS

The results of the MTR tests indicate that most of the program objectives have been achieved. The fast-response thermopile-type neutron-flux-meter concept has proved to be a practical flux-measuring device. Signal-output characteristics are reasonably linear with flux density and have shown good long-term stability. However, each meter will require individual calibration, since fabrication techniques have not yet been perfected to the extent that absolutely uniform results can be obtained. The only design objective not yet fully met is the desired high-temperature limit of 1000°F. Use of the silver/Constantan type plated thermocouple junctions in this application limits the upper temperature level to approximately 500 to 600°F. The use of more oxidation-resistant combinations such as platinum/ platinum-rhodium will yield higher temperature limits but will reduce signal output if the same number of junctions is maintained.

RECOMMENDATION

The feasibility of the thermopile-type neutron-flux meter has been reasonably established. Therefore, future work should be directed toward application of this device as a neutron-measurement instrument in reactor installations to 1) obtain further test history, 2) investigate practical use problems, and 3) determine packaging requirements. At the completion of the above application phase, further consideration should be given to fabrication techniques to insure more uniform output characteristics.

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