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Calorimetric measurement of radiation dosage

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CALORIMETRIC MEASUREMENT OF RADIATION DOSAGE

by

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I. INTRODUCTION

Dosimetry, in the larger sense, is the determination of the energy
absorbed in a medium exposed to ionizing nuclear radiations. Accurate and
adequate dosimetry is essential in radiation effects studies. Since only
neutrons and gammas can penetrate more than a few centimeters of solid
matter, description of these radiations is usually the most important.
The gamma rays interact with the electrons of the material, producing
energetic electrons which produce ionization. Neutrons interact with the
nuclei, energizing the nuclei to cause intense ionization and also dis-
placing of other atoms of the material.

According to Collins and Calkins (3), there exist two philosophical
approaches to dosimetry: (a) description of the radiation field, and
(b) some type of description of the effect of the radiation field upon the
material being investigated. Most of the literature concerning radiation
effects describes the radiation field--in most cases inadequately because
of the experimental difficulties involved in measuring a radiation spectrum.

Actually, the mechanisms of radiation damage dictate different types
of dosimetry for different materials (3). For metals and alloys, damage
is caused chiefly by collision with fast neutrons; the best dosimetry is a
description of the neutron spectrum and time of exposure. Ceramic and in-
organic solids require the neutron spectrum, gamma energy deposition rate,
and time of exposure. Organic materials are damaged by the disruption of
their weak covalent bonds. The extent of the damage appears to be related
to the total energy absorption; measurement of this (including specification
of the individual contributions from neutrons and gammas) may adequately

describe the dose received by organic materials.

Calorimetry is a useful method employed to measure energy absorption. It has not enjoyed much popularity because other methods of radiation measurement involving ionization and activation are much simpler. However, calorimetry does make it possible to give a direct measurement of a dosage description useful for organic materials radiation effects studies--the rate of energy absorption.

In many cases it is desired to make these measurements within the core of a nuclear reactor. The remote operation of a calorimeter in an intense radiation field has many limitations and problems not encountered in ordinary calorimetry.

This thesis describes the design and testing of a simple calorimeter system which makes possible the separation of the contributions of neutron and gamma dosage within the core of a reactor. When properly calibrated, the temperature difference produced in the calorimeters is directly proportional to the rate of energy absorption.

The feasibility of the proposed design was first investigated through calculations and through study of other calorimetry projects. With the fast flux available in the Iowa State University UTR-10 Reactor, feasibility was indicated through the use of sensitive temperature measuring equipment. Three calorimeters have been constructed and tested, yielding satisfactory measurements of radiation dosage.

II. DESIGN CONSIDERATIONS

A. Calorimetry

The first recorded use of calorimetry in the nuclear field was by Curie and Laborde (10) in 1903 to measure the heat absorbed in a sample containing radium. Calorimetry has long been used for accurate determination of the rate of heat evolution from radioactive materials.

According to their function, calorimeters are typed as <u>adiabatic</u> or <u>isothermal</u>.

1. Adiabatic calorimeters

In adiabatic calorimetry, no heat is transferred from the calorimeter (inner part) to the jacket. This is best accomplished by maintaining both at the same temperature. The rate of heat generation is directly proportional to the observed rate of temperature rise.

The use of an adiabatic calorimeter is described by Lazo, Dewhurst, and Burton (8) for measuring the gamma ray energy absorbed in a sample of ferrous sulfate. Their calorimeter was calibrated by an electrical heating filament by observing the rate of temperature increase for various power inputs. Their apparatus included elaborate heating circuits to maintain all parts at the same temperature insuring adiabatic conditions.

Adiabatic calorimeters were used by Fischer of M. I. T. (4) and by Anderson and Waite of Harwell, England (1) for in-pile measurements. Because of the practical difficulties of using thermostats, these investigators found it necessary to have the calorimeters connected to vacuum pumps to promote adiabatic conditions. It was necessary to wait for the

calorimeter and the jacket to attain the same temperature before recording
the rate of temperature increase. If this condition did not occur, ex-
trapolation was used. Thermocouples were employed for temperature
measurements. Fischer used the specific heat of the absorbers in deter-
mining the proportionality constant; Anderson and Waite used electrical
calibration.

2. Isothermal calorimeters

Thermal equilibrium is necessary in isothermal calorimetry to measure
the rate of heat generation. The steady-state temperature difference is
directly proportional to the heating rate.

Measurements in the Oak Ridge Graphite Reactor using isothermal
calorimeters have been reported. Richardson, Allen, and Boyle (12)
utilized large (18 inches long) isothermal calorimeters containing samples
of H_2O, D_2O, carbon, aluminum, and bismuth. Binder, Bopp, and Towns (2)
describe a smaller calorimeter (3 inches long) which contained two samples
(nylon and graphite) at the same time. Both groups of investigators used
electrical calibration; thermocouples were used to measure wall temperatures.
Binder et al. found excellent agreement between the calorimetric measure-
ments and multi-group calculations.

The accuracy of isothermal calorimeters is improved by increasing the
temperature gradient between the calorimeter and the jacket. However,
evacuation of the calorimeter presents practical difficulties and is not
really necessary. For reproducibility, a constant vacuum must be main-
tained which is practically impossible at some distance from the pump.
In fact, an increase in the temperature gradient results in an increase
in the time required to achieve equilibrium.

3. Comparison

The adiabatic calorimeter possess the advantage of not requiring a long period of time to attain equilibrium. Measurements over a few minutes are usually sufficient. However, attaining strictly adiabatic conditions poses problems. A complex thermostat may be used to maintain proper temperatures. Heat transfer is minimized by the use of vacuum and highly polished surfaces. A jacket that is vacuum tight must be massive enough to withstand the pressure, even though any increase of mass in the calorimeter assembly increases heat production in the calorimeter itself. A long, heavy vacuum line would be required for operation within a reactor. No practical solution exists for eliminating heat conducted through connecting and support wires, hence this will be a source of error in the adiabatic calorimeter.

Isothermal calorimeters require a relatively long period of time to reach thermal equilibrium--perhaps thirty minutes to an hour. For purposes of reproducibility, it is desirable for the surroundings to be held at constant temperature. No vacuum is required, so that the mass of the calorimeter may be at a minimum. Extreme care to minimize conduction and radiant heat transfer is unnecessary.

The isothermal calorimeter was chosen for this investigation. The main reason is that only the measurement of temperature difference, not absolute temperatures of wall and jacket, is required. As shown in a following section, resistance thermometers in a Wheatstone bridge circuit can be profitably employed to measure this temperature difference directly. This direct measurement of temperature difference, plus the advantage of low mass, dictated the choice of isothermal calorimetry.

B. Estimated Dosage

Estimation of the rate of energy absorption in various samples is useful in checking the design of the calorimeters. A convenient location in the UTR-10 reactor for testing the calorimeters is at the inside end of the center stringer of the thermal column. Here, approximately six inches from the core tank, the thermal and fast neutron fluxes each are approximately $1.0 \times (10)^{10}$ neutrons/cm^2-sec.

The fission process furnishes energy in several different forms. The distribution of fission energy is useful for examining possible methods of energy transmission to the absorber sample. A typical distribution of fission energy is given in Table 1 (5, p. 71).

Table 1. Distribution of fission energy

Kinetic energy of fission fragments	162 Mev
Beta decay energy	5
Gamma decay energy	5
Neutrino energy	11
Energy of fission neutrons	6
Instantaneous gamma-ray energy	6
Total fission energy	195 Mev

The heavy fission fragments do not escape from the fuel region; their energy is converted to heat which is ultimately removed by the moderator-coolant. The range of beta particles is no more than a few centimeters through solid material, and they also cannot reach the absorber sample contained in the calorimeter. However, bremsstrahlung will affect the absorber, contributing dosage as gamma rays. The neutrinos are not detected

in the sample.

The dosage received from the fission neutrons is an important contribution. Activation of the sample material is possible by neutron absorption processes; excitation of the atoms is likely by collision with the energetic neutrons. If the absorbers used have low cross sections for the absorption of neutrons, as is the case for hydrogen, carbon, and lead, the energy absorbed through activation will be small. The transfer of kinetic energy by fast neutron flux, however, will be important. The general expression for the number of processes in a nuclear reaction may be adapted to predict the dose rate, d_n (watts/gm), received by one gram of absorber material,

$$d_n = k \int_0^\infty N\, \sigma(E)\, \phi(E)\, E\, dE \qquad\qquad 1$$

where k is the average fraction of kinetic energy transferred per neutron collision $(K = 2A/(A+1)^2)$.

N is the number of atoms of absorber present per gram $(N = A*/A)$.

$\sigma(E)$ is the cross section for scattering (barns).

$\phi(E)$ is the neutron flux (neutrons/cm^2-sec).

E is the energy of the incident neutrons (Mev).

For an approximate evaluation of this integral, two simplifying assumptions are made: (a) the neutron flux is assumed to consist of one group of 0.5 Mev neutrons at $1.0 \times (10)^{10}$ neutrons/cm^2-sec; (b) an assumed average cross section for fast neutrons is estimated from the charts in Hughes (6). The estimated dose rates are given in Table 2. The values for the masses of the samples are those actually used in the experiments.

As seen in Table 2, the hydrogen will be the principal absorber of fast neutrons. Since the mass of a hydrogen atom nearly equals that of a

Table 2. Estimated fast neutron dose rates in absorber materials

Element	A	Fast neutron cross section (barns)	Estimated dose rate (10^{-3} watts/gm)	Mass of sample (gm)	Estimated total dose (10^{-3} watts)
H	1	3	0.72	3.2	2.3
C	12	1.5	0.0085	18.2	0.15
Al	27	3	0.0037	----[a]	----[a]
Pb	207	6	0.0001	266.0	0.03

[a]Aluminum not tested as a sample.

neutron, the maximum kinetic energy is transferred from neutron collision. The fast neutron heating of the heavier atoms (carbon, aluminum, and lead) is practically negligible compared to that of hydrogen.

As seen from the fission spectrum, approximately twice as much energy is released in the form of gammas than in the form of neutrons. The relatively high gamma flux in the reactor, plus the penetrating power of gamma radiation indicate that the gammas will constitute an important energy contribution.

The absorption of gamma rays in the absorber takes place by three principal mechanisms: (a) photoelectric absorption, (b) Compton scattering by the electrons in the atoms, and (c) production of electron-positron pairs. The absorption coefficient μ can be expressed in terms of the coefficients for photoelectric, Compton, and pair production, τ, σ, and κ, by the following expression (7):

$$\mu = \tau + \sigma + \kappa$$

2

The Compton scattering process predominates for gamma energies in the intermediate range. If it is assumed that all of the gamma flux in the reactor may be characterized as having energy in that range, the mass absorption coefficient μ/ρ may simply be expressed:

$$\frac{\mu}{\rho} = \frac{\sigma}{\rho} \qquad\qquad 3$$

The Compton scattering coefficient σ is a cross section for the inter-action of the incident photon with the electrons of the absorber. If $_e\sigma$ is the cross section for interaction with one electron, then σ depends upon the number of electrons in the absorber:

$$\sigma = \frac{\rho A^* Z}{A} \, _e\sigma \qquad\qquad 4$$

The dose rate r_γ (mev/sec-gm) from gamma rays in the absorber is given by

$$r_\gamma = I \frac{\mu}{\rho} \qquad\qquad 5$$

where I is the gamma intensity (mev/cm^2-sec) and μ/ρ is the mass absorption coefficient (cm^2/gm) for the absorber material. Since it is assumed that all interactions may be characterized as Compton scattering, the dose rate from gammas may be written,

$$r_\gamma = I \frac{A^* Z}{A} \, _e\sigma \qquad\qquad 6$$

In order to compare the gamma dosage received by two different materials, the above expression may be used to obtain the ratios of the gamma dosage:

$$\frac{r_{\gamma_1}}{r_{\gamma_2}} = \frac{(Z/A)_1}{(Z/A)_2} \qquad\qquad 7$$

It is noted that I, A*, and $_e\sigma$ are constants.

In the proposed calorimeters, aluminum finds use as a structural material. Aluminum, in addition to the heating by fast neutrons and gammas, undergoes the (n,γ) reaction with thermal neutrons forming Al-28 which has a 2.4 minute half life. The decay of the Al-28 releases a beta ray and a gamma ray. The beta rays will mostly be absorbed in the aluminum and the gamma rays may be partially absorbed. Therefore, the dosage in an empty calorimeter should be subtracted from the total observed dosage to determine the sample dosage.

In the case of the samples to be used, it is seen that the dosage received is due only to the effects of fast neutrons and gammas. From the preceding discussion, it may be stated that the heating of the lead sample is due to gamma activity only; the sample containing hydrogen and carbon (paraffin) will receive dosage from both gammas and fast neutrons. Thus, the lead absorber sample suffices for gamma dosimetry. Then, by computing the gamma dosage in the paraffin and subtracting this from the combined dosage, the fast neutron dosage is obtained.

III. DESCRIPTION OF APPARATUS

Three calorimeter assemblies, identical in every detail except for the absorber samples used, were constructed. A cross-section of a calorimeter is shown in Figure 1. The first calorimeter contained no absorber, the second contained paraffin, and the third contained lead.

The calorimeter (i.e., the inner can) was constructed from a spun aluminum cartridge 0.012 inches thick, 1.20 inches long, and 1.20 inches in diameter. The end closure of the calorimeter was the same material, pressed on for a tight friction fit. In order to assure consistency among calorimeters, the masses of these assemblies were made as close as possible. In this case the combined mass was 6.065 ± 0.020 grams.

The jacket was made from a commercial aluminum 6-ounce-capacity can 0.006 inches thick, 4.70 inches long and 2.12 inches in diameter. The end closure of the jacket was also a tight friction fit. The masses of the jackets and their end closure also were as close as possible; this combined mass was held to 14.060 ± 0.020 grams. All aluminum surfaces were cleaned carefully with acetone prior to assembly in order to minimize the accumulation of aluminum oxide which increases radiant heat transfer.

Four resistance temperature gages were used in each calorimeter. All of the gages used were RdF Stikons, manufactured by the RdF Corporation, Hudson, New Hampshire. The gages were RdF type BN-200, containing CP Nickel resistance wire and having a bakelite carrier material. The nominal resistance of the gages was 200 ± 2.0 ohms at 70 °F. At room temperature, the calibration factor supplied by the manufacturer is 0.00310 ohms/ohm-°F.

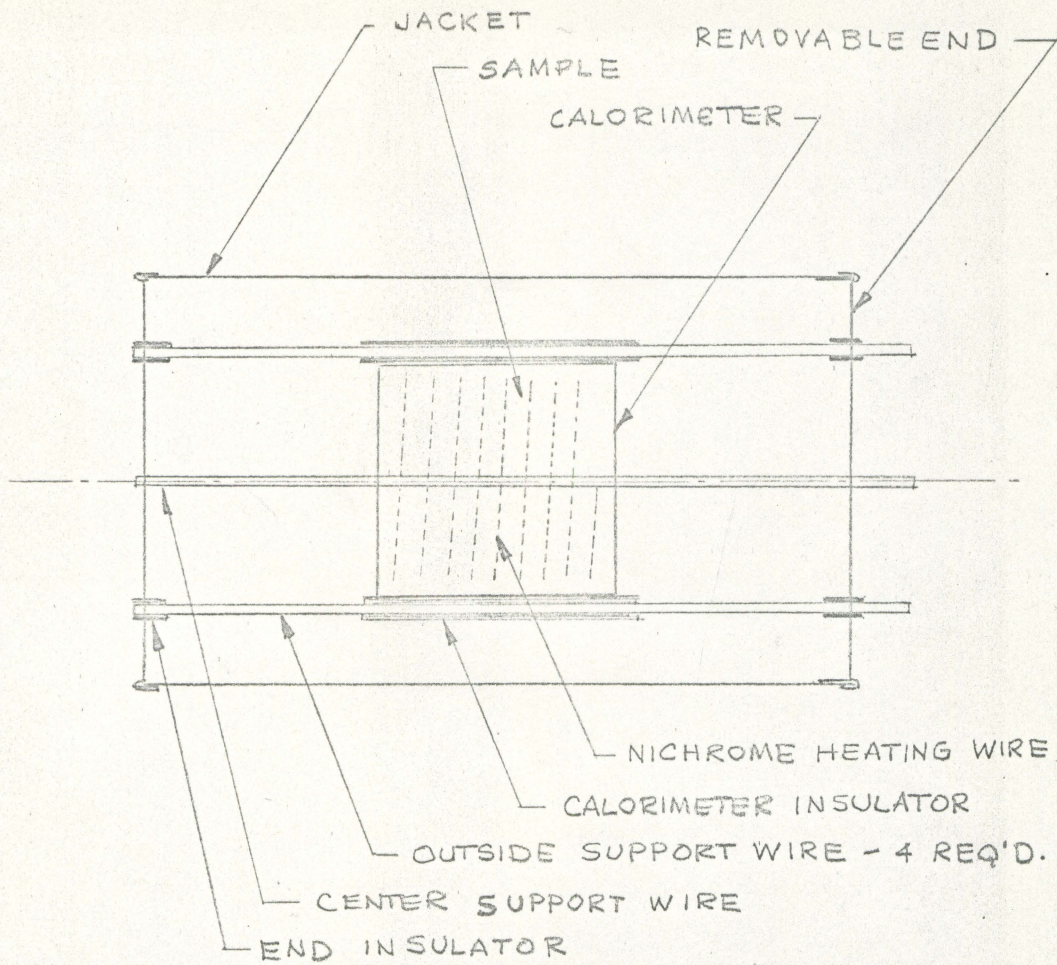Two gages were mounted on opposite sides of the outside of the

Figure 1. Cross section of calorimeter assembly (full scale)

calorimeter, and two gages were similarly mounted on the outside of the jacket. Mounting was done with RdF high-temperature (ceramic) cement, using contoured clamps cushioned with neoprene, and maintaining approximately 150 psi on the gages for curing. The gages were cured, in accordance with the manufacturer's recommendations, for one hour at 140 °F, two hours at 175 °F, and two hours at 260 °F.

The calorimeter was supported by five 0.064-inch diameter EC grade aluminum wires which also serve as electrical connectors. Copper wires would have been more satisfactory for this purpose, except that the copper has undesirable nuclear characteristics. The aluminum support wires were isolated from the calorimeter and from the jacket ends with polyethylene insulation. All electrical connections were soldered with a special aluminum eutectic.

A heating coil was contained in each calorimeter and connected to the outside of the jacket through the support wires. This heating filament was made from 24 inches of 0.0031-inch diameter enameled nichrome wire. The resistance of this filament was approximately 120 ohms. In the lead and paraffin absorbers, the wire was uniformly distributed, attempting to approximate a uniform heat source. For the empty calorimeter, the filament was coiled next to the inside surface.

For the actual construction, the support wires, the jacket end closure, and the calorimeter with absorber and gages in place were subassembled as pictured in Figure 2. After this subassembly was fitted into the jacket, electrical connections were completed, and the lead wires were connected (Figure 3).

A styrofoam holder to replace the inner central stringer of the re-
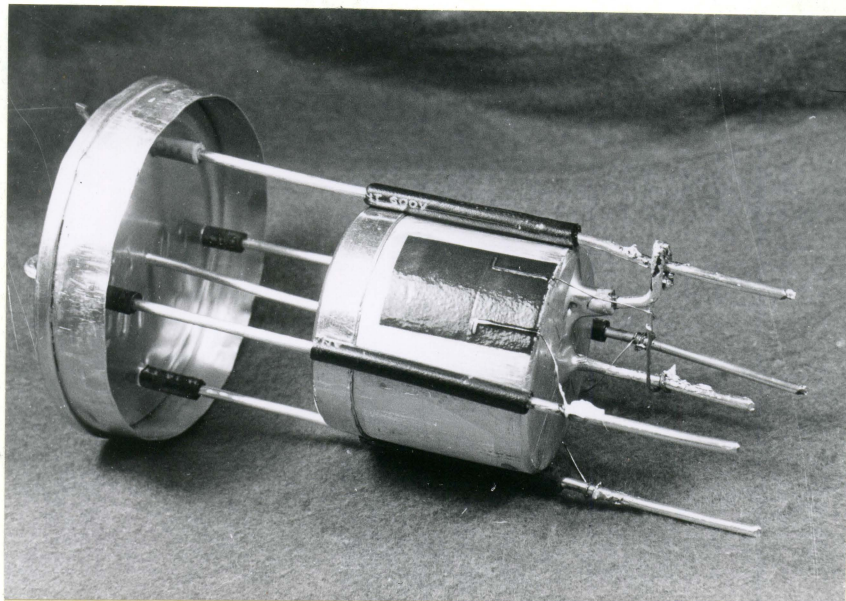
Figure 2.   Subassembly consisting of calorimeter,
support wires, temperature gages, and
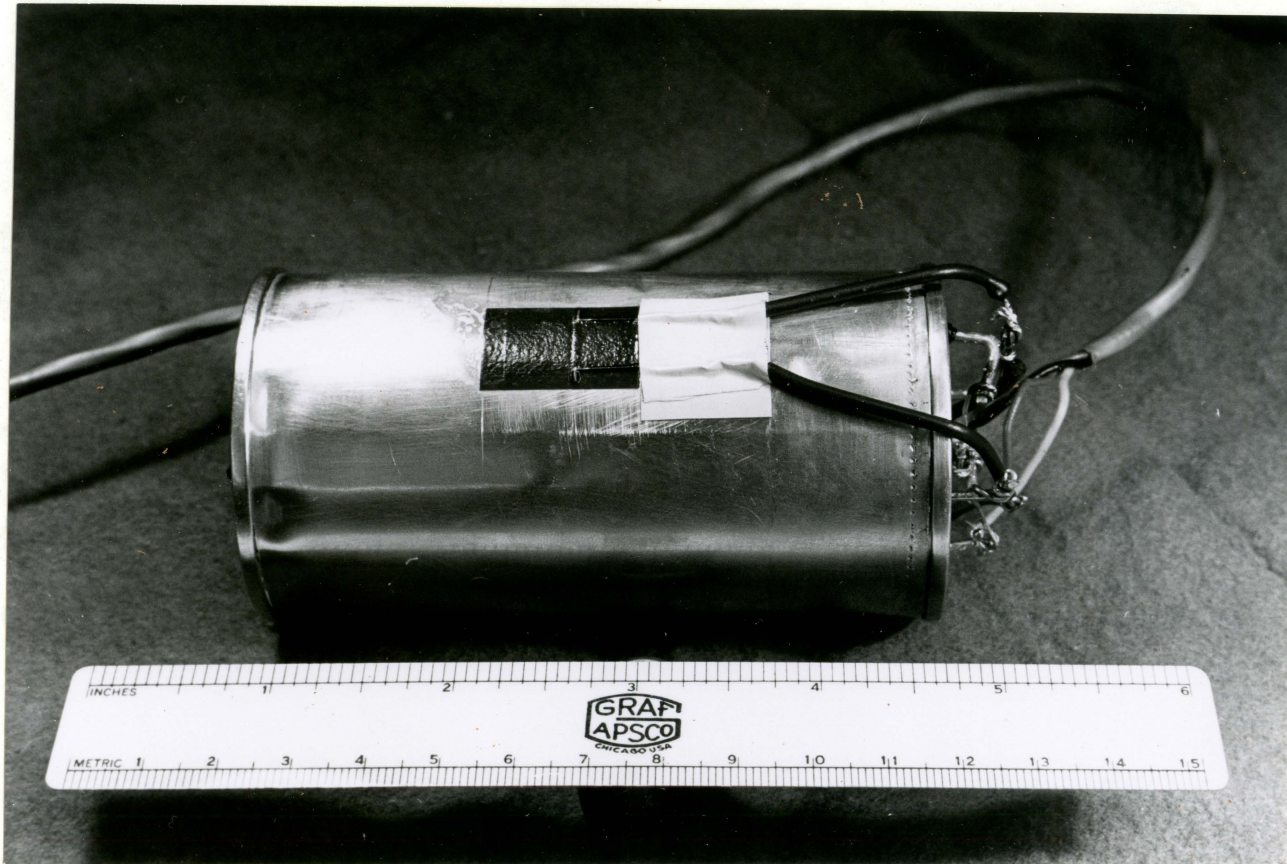end closure (approximately full size)

Figure 3. Completed calorimeter assembly, with electrical connections
(approximately full size)
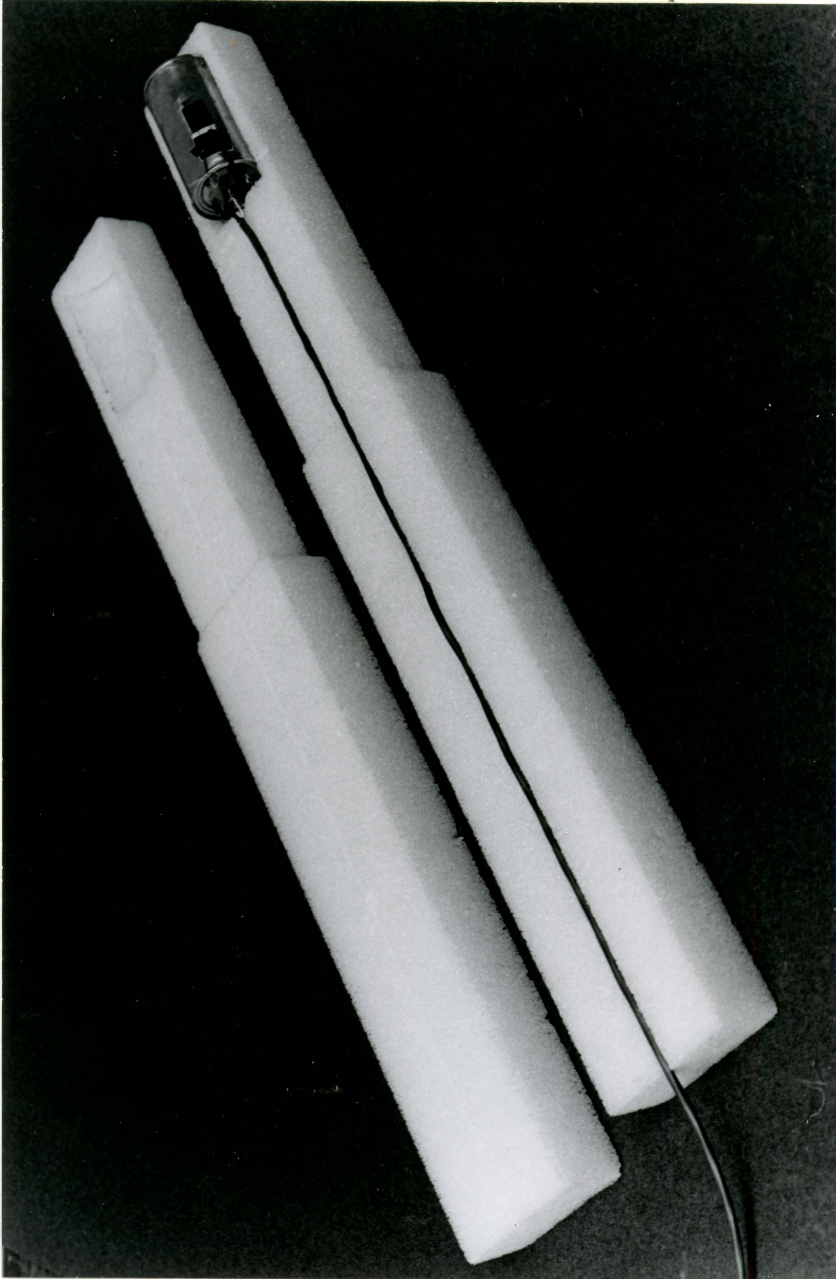
Figure 4. Styrofoam holder used in reactor thermal column, opened into its two halves

actor thermal column served the dual purpose of positioning the calorimeters and insulating them from their surroundings. This holder was made from two identical pieces of styrofoam and is shown in Figure 4, open, with the calorimeter assembly and lead wires in place. This holder also was used during the electrical calibration runs.

The Wheatstone bridge used in conjunction with the resistance temperature gages was a Baldwin-Lima-Hamilton SR-4 Indicator, Type N. Although this instrument reads unit strain in microinches per inch, the readings may be easily converted to degrees Fahrenheit. A two-arm bridge circuit was used, with two gages in series in each of the arms. Parallel connection of the gages is undesirable, because self-heating is considerably increased.

An attempt was made to keep all conditions similar in the construction and testing of the calorimeters, so that the difference in each sample caused by the effects of radiation heating is accurately observed.

IV. DESIGN ANALYSIS

Analysis of the operating characteristics of the calorimeters is useful for predicting the requirements for testing conditions. Heating of a calorimeter is to be from one of two sources: (a) the electrical power applied to the filament for calibration, or (b) the energy absorbed from neutrons and gammas in the reactor environment.

An estimation using heat transfer to determine the temperature difference expected between the calorimeter and the jacket is first made to study the requirements for the temperature measuring apparatus. Then, the Wheatstone bridge circuit used with the resistance thermometers is analyzed, showing how this temperature difference may be measured. Finally, the feasibility of resistance thermometers for use in radiation fields is discussed.

A. Heat Transmission

Heat transmission calculations are used to predict an approximate temperature difference between the calorimeter and jacket. For all calculations, it is assumed that the total dose received by the sample is 0.050 watts. The mechanisms of conduction, radiation, and natural convection are checked separately. The temperature difference for each case is found, assuming that each is the sole method of heat transmission. McAdams (9) has been used as a reference.

1. Conduction

For steady-state conduction, it is convenient to employ the potential

form for the rate of heat flow, q (Btu/hr):

$$q = \frac{\Delta t}{R}$$ 8

where Δt is the temperature difference (degrees F) between the jacket

and calorimeter, and R is the total equivalent thermal resistance (hr-$^\circ$F/Btu)

between the surface of the calorimeter and the surface of the jacket.

Using one-dimensional geometry, R is estimated to be 162 hr-$^\circ$F/Btu. For

a heat flow of 0.050 watts exclusively by conduction, the temperature

difference is then estimated to be 24 $^\circ$F.

2. Radiant heat transmission

The radiant heat transmission between the calorimeter and the jacket

can be estimated with Hottel's equation (9, p. 72).

$$q_{net} = s\,A_1\,f_{12}\,(T_1^4 - T_2^4)$$ 9

where s is the Stefan-Boltzmann constant, A_1 is the effective area of the

inside cylinder, and f_{12} is a function of geometry and surface condition.

An estimation of the factor f_{12} is made treating the two surfaces as

parallel planes, and the surface condition as "polished aluminum". For

a heat flow of 0.050 watts by radiation, substitution of numerical values

yields a temperature difference on the order of 10^3 $^\circ$F. This mode of heat

transmission is probably negligible.

3. Convection

The prediction of heat transfer by convection is based on Newton's

law of cooling (9, p. 5),

$$q_c = h_c\,A_1\,\Delta t$$ 10

where h_c is the coefficient of heat transfer (Btu/hr-ft^2-^{0}F). A_1 is the

surface area of the calorimeter (ft^2), and Δt is the temperature difference

(^{0}F). If it is assumed that the temperature difference is small (a few

degrees), an approximate procedure for horizontal tubes given McAdams

(9, p. 177) may be employed to estimate h_c,

$$h_c = 0.27 \left(\frac{\Delta t}{D_1} \right)^{1/4} \qquad\qquad 11$$

where D_1 is the diameter of the calorimeter. If 0.050 watts are trans-

ferred by convection, Equations 10 and 11 predict a Δt of 1.4 ^{0}F, showing

that convection may be the principal mode of heat transfer in the calorimeters.

Even though the calculations are approximate, the necessity of measuring

small temperature differences in the calorimeters is indicated by this re-

sult.

4. Over-all heat transmission

It is readily apparent that the heat transfer characteristics of the

calorimeters should be determined experimentally. Error in the estimations

above probably exists due to lack of sufficient information and to over-

simplification. In fact, it is to be expected that the characteristics will

vary from one calorimeter to another since small differences may exist in

surface condition and in details of geometry.

In the calibration of the calorimeters by adding known heating rates

electrically, it is possible to determine an over-all coefficient taking

into account all geometry and all modes of heat transfer. The over-all

coefficient times the measured temperature difference gives the rate of

energy absorption by the calorimeter.

B. Temperature Measurements

Remote measurement of a small temperature difference is required for
an in-pile calorimeter. All of the investigators previously cited used
thermocouples for this purpose. Some uncertainty exists with the use of
thermocouples in a radiation field, even though they are in very common
usage. Thermocouples require the measurement of very small voltages and
the use of a reference point which make operation inconvenient.

A resistance thermometer represents another, though less commonly
used, approach to the remote measurement of temperature. Nickel is the
most popular material for this application, since its resistance varies
greatly with temperature and since its response is nearly linear at or-
dinary temperatures. A convenient form of resistance thermometer, known
as an "RdF Stikon", incorporates a grid of very small diameter nickel
wire sandwiched between two thin sheets of insulating material. This
assembly is cemented to the surface of which the temperature is being
measured. In every respect, the resistance thermometer gage is similar to
an SR-4 strain gage, except that temperature is measured instead of mechan-
ical strain.

For an isothermal calorimeter it is only necessary to measure the
temperature difference between the calorimeter and the jacket. A Wheat-
stone bridge circuit, which is used in experimental stress analysis with
SR-4 strain gages for measurement of bending strains with temperature com-
pensation, adapts well to the measurement of temperature differences with
the resistance thermometers. Such a circuit is shown in Figure 5. In
operation, the calibrated ratio of the resistances R_a and R_b is adjusted
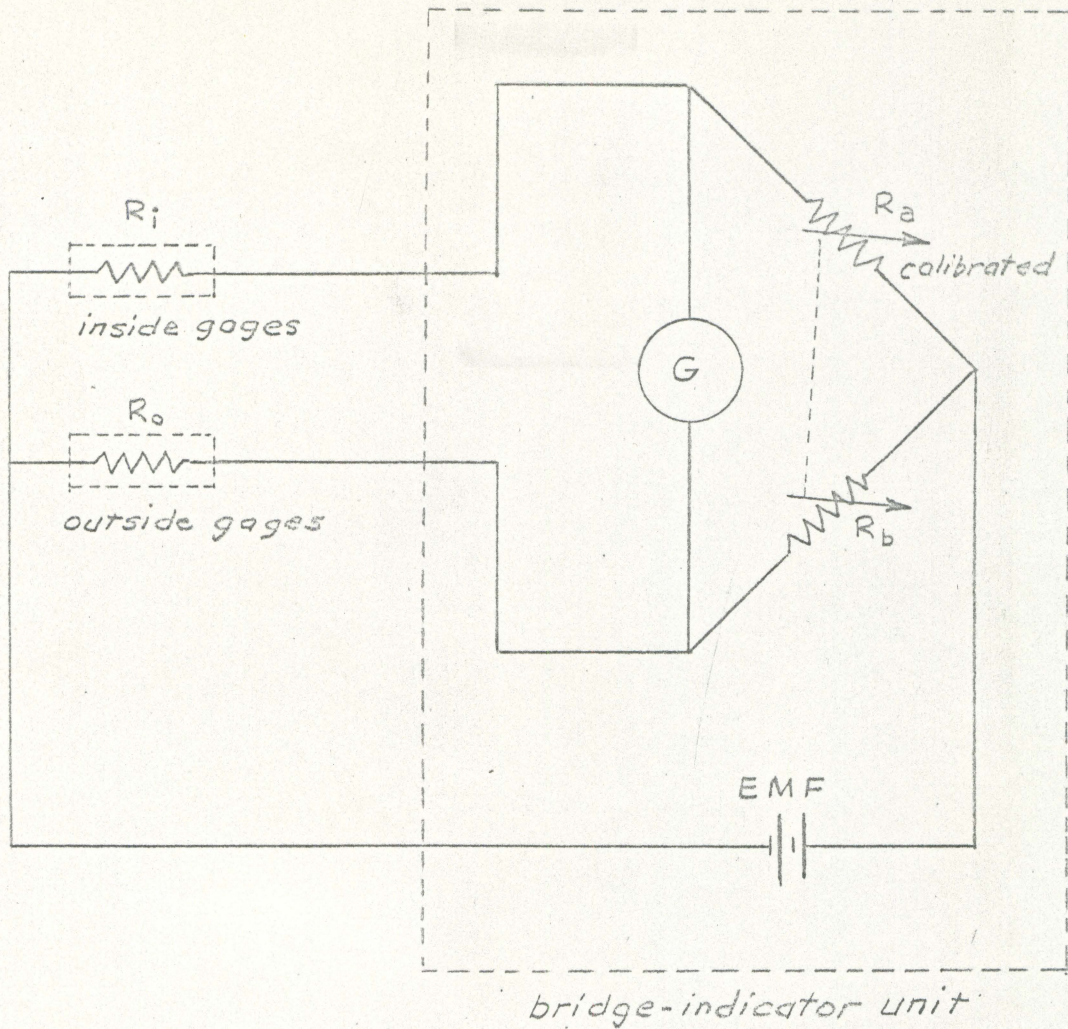to null the galvonometer G. Resistances R_i and R_o represent the resistance

Figure 5. Wheatstone bridge circuit for remote measurement of temperature difference

thermometer gages on the calorimeter and jacket respectively. For a
balanced bridge, the ratios of the resistances may be written,

$$\frac{R_i}{R_o} = \frac{R_a}{R_b} \qquad\qquad 12$$

If the gages both experience the same change in temperature,

$$\frac{\Delta R_i}{R_i} = \frac{\Delta R_o}{R_o} \qquad\qquad 13$$

the balance of the bridge is not changed, since $R_i \approx R_o$.

$$\frac{R_i + \Delta R_i}{R_o + \Delta R_o} = \frac{R_i}{R_o} = \frac{R_a}{R_b} \qquad\qquad 14$$

If the inside gage undergoes a temperature increase denoted as
$\Delta R_o + \Delta R$, which is greater than the increase of the outside gage ΔR_o,
then a ΔR_a of the calibrating resistance is needed to balance the bridge:

$$\frac{R_i + \Delta R_o + \Delta R}{R_o + \Delta R_o} = \frac{R_a + \Delta R_a}{R_b} \qquad\qquad 15$$

Since $R_i \approx R_o$, $R_a \approx R_b$, and the ΔR terms are small, Equation 15 becomes

$$1 + \frac{\Delta R}{R_o} = 1 + \frac{\Delta R_a}{R_b}$$

$$\frac{\Delta R}{R_o} = \frac{\Delta R_a}{R_b} \qquad\qquad 16$$

showing that ΔR_a is proportional to the difference of the change in R_i
and R_o. Thus, the Wheatstone bridge makes possible the direct measure-
ment of a temperature difference.

The suitability of SR-4 strain gages for use in radiation fields has
been investigated by Smith and Rendler of the Naval Research Laboratory
(11), (13). Their findings show that bakelite gages bonded with ceramic
cement operated satisfactorily with an integrated fast flux up to 6 (10)[17]

neutrons/cm^2, where the fast flux was 5 (10)^{11} neutrons/cm^2-sec.[1] They found that the metal element of the strain gage was unaffected, but the critical area was the breaking down of the insulation between the element and ground. Paper backed gages were satisfactory only in very low fluxes.

The conductors of the strain gages tested by Smith and Rendler were Constantan and Nichrome V--both are alloys of nickel. Hence, the all-nickel conductors of the resistance temperature gages should show a similar immunity when proper backing and cement are used.

The temperature coefficient of the nickel used in the gages for these experiments is given by the manufacturer as 0.0031 ohms/ohm-$^\circ$F, which provides excellent sensitivity. This, radiation resistance, and the instrumentation to measure temperature difference directly make the use of these gages very attractive.

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[1]No neutron spectrum is given, nor is any specification of the gamma field. This is a typical example of inadequate information.

V. EXPERIMENTAL PROCEDURE

A. Calibration

After the calorimeter assemblies were completed the gages were checked with a resistance bridge to insure that they had not been damaged. Also, the resistance of the gages to ground was measured to verify that they had been properly mounted and wired. It was not possible to check the temperature calibration factors supplied by the manufacturer because of the lack of suitable temperature-measuring equipment for comparison. Actually, the temperature calibration factors themselves are not important, since the conversion to degrees Fahrenheit is only for a convenient frame of reference.

For a calibration run, the calorimeter assembly was sealed in the styrofoam enclosure which fits the reactor thermal column. The assembly was tested in an environment of approximately 90 °F, surrounded by additional blocks of styrofoam to protect from convection currents.

About an hour was first required for the temperature gages to warm up. Since current from the SR-4 indicator causes some self-heating, the gages drifted until they reached equilibrium. This initial reading corresponded to the zero reading—since no current was applied to the heating filament. After stabilizing at the initial reading, the filament was heated using dry-cell batteries. The voltage ranged from 1.5 to 3.0 volts, with a rheostat used for voltage adjustments. The power supplied to the calorimeters was measured with a voltmeter—ammeter circuit.

Since time was not of the essence, approximately an hour was allowed at each setting to give more than enough time for the attainment of equilibrium.

It was necessary to calibrate each calorimeter separately. Extreme care in handling was necessary so as not to alter the characteristics between the time of calibration and the time of reactor operation.

To convert observed strain indicator readings to degrees Fahrenheit for convenient reference, the definition of the gage factor for SR-4 strain gages was used:

$$\text{Gage Factor} = \frac{\text{Unit change in resistance}}{\text{Unit strain}} \qquad 17$$

The manufacturer supplies a calibration relating the unit change in resistance to the temperature enabling completion of the calculation.

B. Reactor Operation

The calorimeter assembly and the styrofoam holder unit were placed in the thermal column of the UTR-10 the day before the scheduled run. This was to assure complete equilibrium with the thermal conditions of the reactor and the fission-product gamma activity present.

Prior to reactor start-up, the moderator-coolant (light water) was maintained at 90 °F and circulated through the core. The presence of the warm moderator-coolant shields some of the residual gamma activity, and heats the reactor structure to approximate operating conditions. The removal of the control and safety rods was not initiated until the indicated temperature difference for the calorimeter became constant, showing that the calorimeter was in equilibrium with the new conditions.

This equilibrium value was taken as the zero. It took into account the residual gamma activity present and the temperature of the operating reactor. Differences in the readings from the run, then, accounted for the neutron flux and the gamma activity accompanying fission.

The increase of reactor power from zero to the 10-kw full power level required about fifteen minutes. During this time, some energy absorption was noted. During the period at full power operation, temperature difference readings were taken at two-minute intervals and plotted against time. The plot showed an approach to equilibrium conditions, the reactor run was ended.

The three calorimeters were tested in three separate runs. In order that the data obtained may be consistently comparable, the 90 °F moderator-coolant temperature was controlled as closely as possible, the power was automatically controlled at 10 kw, and the same shim rod positions were used to minimize flux tilting.

The measured temperature difference observed in the reactor run was correlated with the rate of energy absorption using the curves obtained in the calibration. Then, based on the measurements from the three calorimeters, the final results for the energy absorbed from fast neutrons and gammas have been obtained.

VI. RESULTS

The results from calibrating and testing of the nuclear calorimeters
have been converted from strain indicator readings into degrees Fahrenheit
merely to provide a convenient frame of reference. The experimental program
is conveniently divided into two phases, the electrical calibration, and
the in-pile determination of radiation dosage. From the results of the
experimental work, the dosimetry received from fast neutrons and gammas
may be separated.

A. Calorimeter Calibration

The steady-state temperature difference as a function of the electrical
power supplied to the heating filament is given in Table 3 for the three

Table 3. Electrical calibration of calorimeters

Calorimeter	Absorber material	Electric power on filament (10^{-3} watts)	Equilibrium bridge reading (μin/in)	Computed temperature difference ($^{\circ}$F)
1	None	0	0	0
		15.7	825	0.53
		38.0	1820	1.12
		61.8	3270	2.11
2	Paraffin	0	0	0
		14.0	720	0.46
		39.0	2080	1.34
		62.7	3630	2.34
3	Lead	0	0	0
		16.5	825	0.53
		40.0	2020	1.30
		64.0	3070	1.98

calorimeters. Approximately one hour was allowed for each setting of voltage and current to insure equilibrium, although only about one-half hour was required for a 0.015 watt increase.

For use in reducing the data from the in-pile testing, the calibration curves in Figure 6 are useful. It is apparent from these calibration curves that the over-all coefficient of heat transfer for the calorimeters is constant for the range of temperature differences encountered. However, a slightly different value of this coefficient is observed for each calorimeter assembly.

B. Measurements in Reactor

In order to determine the required length of time for a dosage measurement in the reactor, data relating the time and temperature difference are plotted. Such a plot for the three reactor runs is shown in Figure 7. The temperature difference which is approached asymptotically is taken as the steady-state value for the purpose of determining the dose rate. In Table 4, the dose rates taken from the calibration curves corresponding to the observed temperature differences are tabulated.

Table 4. Observed total dosage rates in UTR-10 at 10 kw

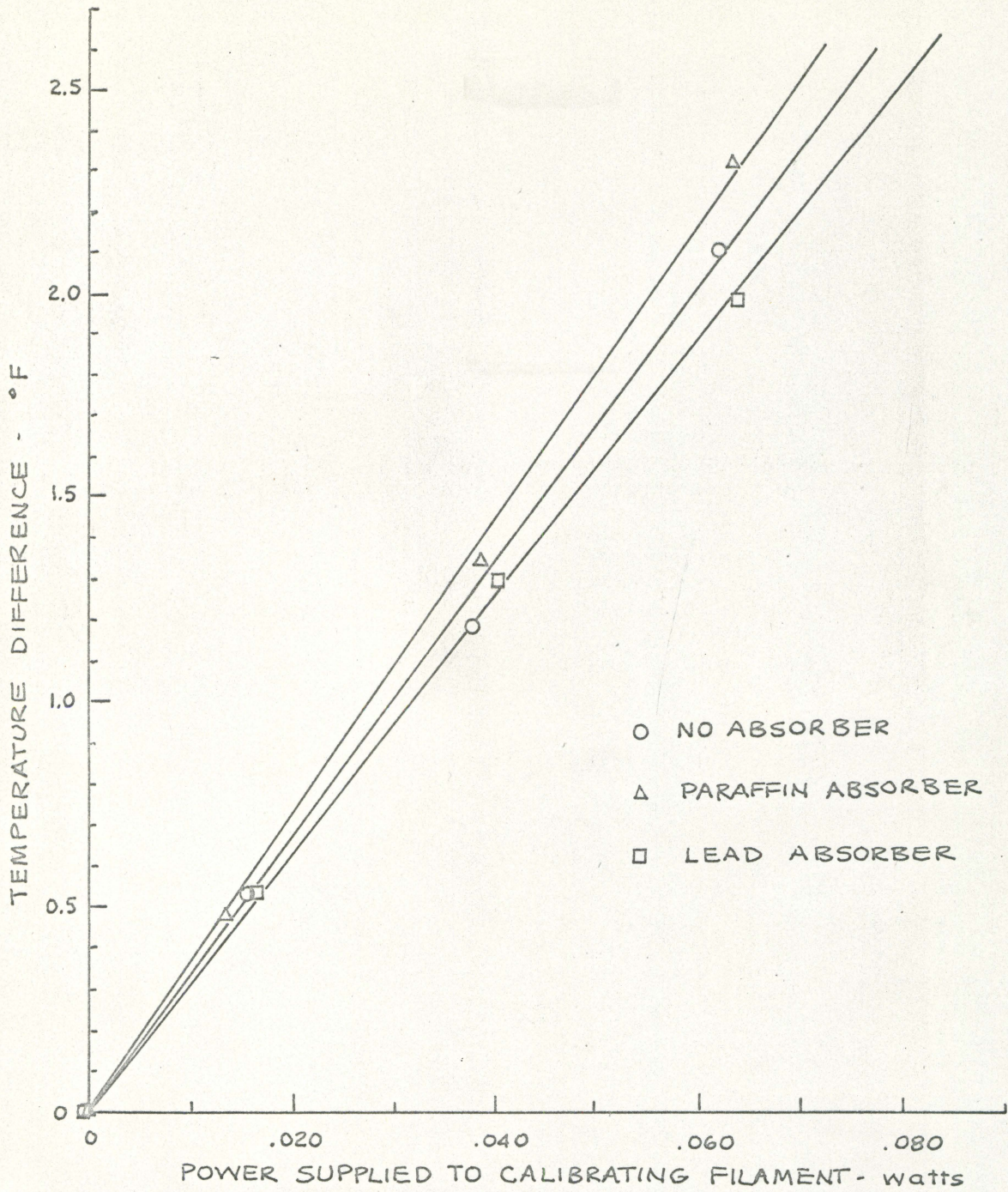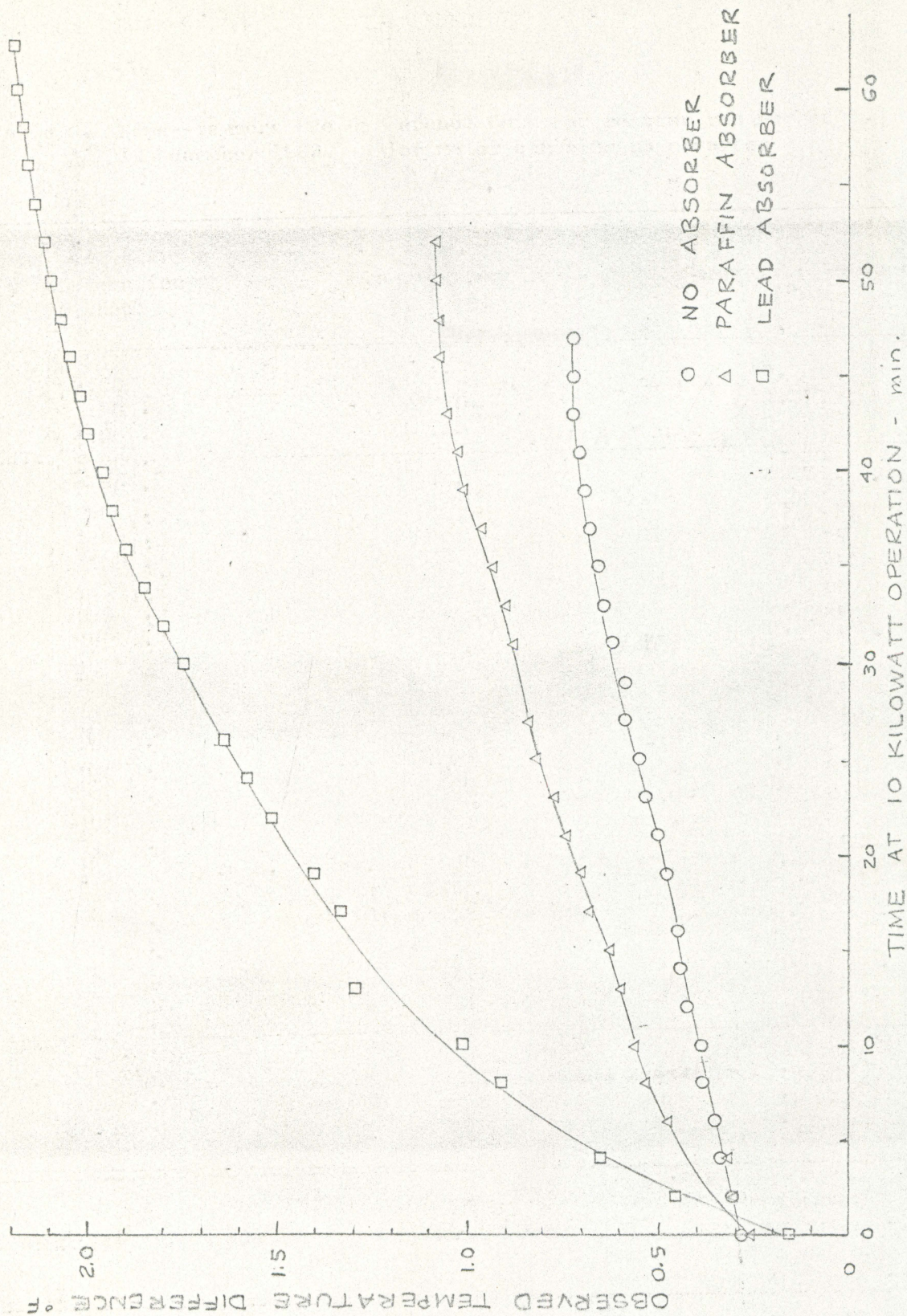| Calorimeter	Absorber material	Mass of sample (grams)	Observed temperature difference ($^\circ F$)	Total observed dose rate (watts)	Total dosage in sample (watts)
1	None	0	0.732	0.021	----
2	Paraffin	21.4	1.090	0.030	0.009
3	Lead	266.0	2.193	0.069	0.048

Figure 6. Calorimeter calibration curves

Figure 7. Approach to thermal equilibrium of calorimeters in reactor

The dose rate in the sample only is obtained by subtracting the total observed dose rate in the empty calorimeter from the total observed rate tabulated for paraffin or lead.

C. Computed Dosages

In order to correlate the data, it is assumed that the same neutron and gamma flux is presented to the calorimeters at each separate run. This is reasonable, since the neutron flux is a measure of the instantaneous power of the reactor, and the gamma activity which is measured is that accompanying the fission process and the fission products of short half life.

As seen from Table 2, the energy transferred by fast neutrons to carbon, aluminum, or lead may be expected to be only about one per cent (or less) of that transferred to hydrogen. Thus, it is valid to assume that the heating in the lead sample is due almost entirely to the absorption of gamma rays. Then the dosage rate observed for lead can be taken as a gamma dosage rate.

Table 5 is based on the experimentally observed combined dosages for neutrons and gammas in the lead and paraffin absorber samples. The calculations are made in the following order: (a) all gamma dose rates, (b) fast neutron dose rates in paraffin, hydrogen, and carbon, and (c) the combined dose rates in hydrogen and carbon.

First, the rate of energy absorption from gamma rays in the elements hydrogen and carbon may be found from the previously derived Equation 7,

$$\frac{r_1}{r_2} = \frac{(Z/A)_1}{(Z/A)_2} \qquad 7$$

where r_1 is known for lead. The total dose rate from gammas in the paraffin is then determined by adding the total gamma dose rate for 3.2 grams of hydrogen to the total gamma dose rate for 18.2 grams of carbon. This total dose rate thus determined for paraffin is divided by the mass, giving the tabulated dose rate in watts per gram.

After the gamma dose rates have been computed, the determinations for the fast neutron dose rates may be made. The fast neutron dose rate for paraffin is the difference between the observed combined dose rate and the calculated gamma dose rate. From Table 2, the fraction of the total neutron dose rate in paraffin due to absorption in hydrogen is shown to be (2.30)/(2.45). Multiplying this fraction by the total neutron dose rate in paraffin and dividing by the weight of hydrogen present gives the tabulated fast neutron dose rate in hydrogen. A similar procedure is followed for carbon.

Finally, the dose rates from the combined effects of gammas and neutrons for hydrogen and carbon are given as the sum of the individual gamma and neutron dose rates. This completes the construction of Table 5.

Table 5. Experimental dosage rates in absorbers

Absorber	Mass (gm)	Z/A	Observed dose rate from neutrons and gammas combined (10^{-3} watts/gm)	Dose rate from gammas only (10^{-3} watts/gm)	Dose rate from fast neutrons only (10^{-3} watts/gm)	Calculated dose rate from neutrons and gammas combined (10^{-3} watts/gm)
Lead	266.0	.396	0.180	0.180	-----	-----
Paraffin	21.4	----	0.420	0.261	0.159	-----
Hydrogen	3.2	1.000	-----	0.455	1.00	1.45
Carbon	18.2	.500	-----	0.228	0.011	0.239

VII. DISCUSSION OF RESULTS

It is interesting to compare the experimentally determined rates of
dosage with those predicted theoretically. The dose rate estimated with
simplifying assumptions for hydrogen (0.00072 watts) is somewhat less than
the experimental value (0.00100 watts). This probably does not mean that
the experimental value is in error; more probably, the computed value is
in error. In order for the dose rate prediction to be correctly made,
a complete specification of the flux distribution at that point in the
reactor is necessary. The term "fast flux" is not an adequate specification.
The discrepancy here points out very vividly the importance of properly
describing the radiation--and how an inadequate specification can lead to
large errors.

Comparison of the experimental dosage rate in hydrogen with the work
of other investigators leads to the same difficulty. Richardson et al.
(12) and Binder et al. (2) separately tested isothermal calorimeters in the
Oak Ridge Graphite Reactor at 3500 kw. Richardson reports a dose rate in
hydrogen of 0.007465 cal/gm-sec which is equal to 0.0312 watts/gm. Binder
publishes $1.5 (10)^{17}$ ev/gm-sec which equals 0.0240 watts/gm. It should be
noted that the tests were made in different locations in the same reactor,
but that the results show fairly good agreement. In addition, Binder
measured the flux spectrum, computed the neutron dose rate, and showed
excellent agreement between theory and calorimetry.

No comparison is possible for the observed gamma dosage rates, since
information is lacking concerning the gamma spectrum as a function of in-
stantaneous reactor power, core history, and other factors. However, it

is reasonable that the gamma energy release during the fission process adds the same contribution at any given reactor power. Since the gammas from fission, and the gammas from secondary processes furnish much more available energy than do the fission neutrons, it is reasonable that the dosage received from gammas should be several times that received from neutrons.

A limited amount of experience has also been gained concerning the time required for making a dosage measurement with the isothermal calorimeter. It may be generalized that calorimeters which measure greater dose rates require longer to achieve equilibrium. For some work, the time required to make a measurement may be disadvantageous. It is possible that the speed of response could be increased by making the calorimeters smaller, but at the expense of accuracy.

The measurement of extremely small dosages accurately may not be possible with a calorimeter of this design, since the temperature difference would probably be too small. On the other hand, it appears possible to use the calorimeters without modification to measure much larger doses than those encountered.

The use of resistance thermometers and the use of the Wheatstone bridge circuit for direct measurement of temperature difference appears to be satisfactory in all respects. This system has many advantages over the more widely used alternative--thermocouples. Any uncertainty associated with the temperature conversion factor of the resistance thermometers is not a problem, since the calorimeters are calibrated and operated with the same gages.

The electrical calibration scheme is a possible source of error.

The nichrome heating wire may only approximate an uniform heat source; the wires may conduct away more heat during calibration than during operation. Therefore, the calibrated values may either be too high or too low. Electrical calibration, however, seems to be the best method.

VIII. CONCLUSIONS

The following conclusions are made on the basis of this work:

1. The isothermal calorimeter is a satisfactory device for dosimetry of intense nuclear radiation in terms of the rate of energy deposition in an absorber material.

2. The isothermal calorimeter has the disadvantages of a relatively long time required for making measurements, and of comparatively low sensitivity to small quantities of radiation.

3. By the judicious choice of absorber materials, isothermal calorimeters can be utilized to separate types of radiation.

4. Resistance thermometers offer excellent possibilities for temperature measurement in the reactor environment.

IX. SUGGESTIONS FOR SUBSEQUENT INVESTIGATION

During the course of this project, several possible areas for further investigation have become apparent. It is believed that the calorimetric method is a valuable tool as an adjunct to study of radiation effects on organic materials.

In the construction of another calorimeter, it would be advantageous to attempt further reduction of mass. Perhaps thinner aluminum could be used for the calorimeter—it may only be necessary to vapor deposit aluminum directly on a sample. The mass of the support wires could be eliminated by using nylon threads to support the calorimeter in the jacket, and using fine copper wire for electrical connections.

Another area of investigation is to study the effects of size and geometry of the calorimeters. Possibly there exists an optimum sample size. This information would be very helpful in the design of more efficient calorimeters.

Alternate methods of calibration should be attempted to substantiate further the results. One method would involve testing in a mixed radiation field of known energy and distribution, checking experimental results with those obtained by calculations. In another method, an adiabatic calorimeter could be tested against an isothermal calorimeter. Other possibilities involve the use of chemical dosimeters, activation analysis, threshold detectors, or ionization chambers either singly or in combination with other methods.

A novel project would be the construction of a calorimeter that permits simultaneous operation in both the adiabatic and isothermal mode.

A calorimeter containing two absorbers, one for gammas and the other for neutrons, would be much more convenient than the separate calorimeters used here. For investigation of radiation effects, provision could be made to hold the sample as well as the two detector absorbers. Measurement of the gamma and neutron dosage simultaneously with the sample exposure would simplify correlating data and would require less time to make the measurements.

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XI. ACKNOWLEDGEMENTS

XII. APPENDIX

Table 6. Time--temperature difference data for reactor run number 1,
          12 December 1963. Calorimeter contains no absorber

Time from start of 10 kw operation (min:sec)	Δε Observed reading (μin/in)	Δt Computed difference (°F)
0:00	435	0.281
2:00	480	0.310
4:00	515	0.332
6:00	550	0.355
8:00	580	0.374
10:00	600	0.387
12:00	655	0.423
14:00	675	0.435
16:00	685	0.442
19:00	750	0.484
21:00	790	0.510
23:00	820	0.530
25:00	855	0.552
27:00	920	0.594
29:00	951	0.591
31:00	970	0.626
33:00	1000	0.646
35:00	1030	0.664
37:00	1060	0.684
39:00	1070	0.691
41:00	1100	0.710
43:00	1130	0.728
45:00	1135	0.732
47:00	1135	0.732
Scram		

Table 7. Time--temperature difference data for reactor run number 2,
17 December 1963. Calorimeter has paraffin absorber

Time from start of 10 kw operation (min:sec)	$\Delta \epsilon$ Observed reading (μin/in)	Δt Computed difference (°F)
0:00	420	.271
2:00	480	.310
4:00	505	.326
6:00	770	.497
8:00	832	.538
10:00	860	.556
13:00	910	.588
15:00	980	.633
17:00	1060	.685
19:00	1104	.713
21:00	1160	.750
23:00	1230	.794
25:00	1280	.826
27:00	1310	.846
31:00	1350	.872
33:00	1400	.905
35:00	1460	.944
37:00	1518	.980
39:00	1575	1.016
41:00	1600	1.032
43:00	1640	1.058
46:00	1670	1.078
48:00	1680	1.083
50:00	1685	1.088
51:00	1690	1.090
Scram		

Table 8. Time--temperature difference data for reactor run number 3,
19 December 1963. Calorimeter has lead absorber

Time from start of 10 kw operation (min:sec)	$\Delta \epsilon$ Observed reading (μ in/in)	Δt Computed difference (°F)
0:00	255	0.164
2:00	730	0.471
4:00	1017	0.656
8:00	1425	0.919
10:00	1585	1.022
14:00	2015	1.300
17:00	2060	1.329
19:00	2185	1.409
22:00	2355	1.519
24:00	2455	1.583
26:00	2545	1.642
28:00	2630	1.696
30:00	2715	1.751
32:00	2795	1.803
34:00	2875	1.854
36:00	2945	1.900
38:00	3000	1.935
40:00	3040	1.961
42:00	3087	1.991
44:00	3135	2.022
46:00	3175	2.048
48:00	3215	2.074
50:00	3255	2.099
52:00	3287	2.120
54:00	3320	2.141
56:00	3355	2.164
58:00	3380	2.180
60:00	3395	2.190
62:00	3400	2.193
Scram		