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ABSOLUTE DOSE MEASUREMENTS ON A MEGACURIE CO-60 SOURCE BY MEANS OF CALORIMETRY

A Method for Calibration of Aqueous
Chemical and Biological Systems

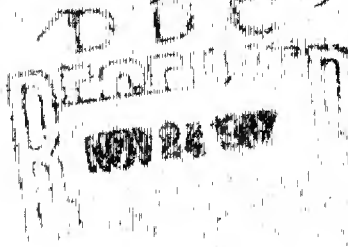
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Research Report

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A Method For Calibration Of Aqueous Chemical And Biological Systems

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ABSTRACT

This report describes the use of a 10-ounce Thermos flask, filled with water, for absolute standardization of a Megacurie Co-60 source. Once the absorbed dose rate is established in a given position relative to the source, other aqueous solutions can be irradiated with an identical, and known, dose. The radiochemical (or radiobiological) yield can thus be readily determined. A short discussion is given on the concepts of radiation calorimetry, and experimental findings are reported for several modes of operating this particular device. It is shown that a set of electrical calibrations combined with a set of irradiations, both carried out in a constant temperature environment, can give complete information on the properties of the calorimeter and the absorbed dose rate. Furthermore, it is shown that once these properties are established, the calorimeter can be operated in a simplified adiabatic mode for determining the absorbed dose rate in other locations or sources. The accuracy of the method is discussed, and application of the system for calibration of chemical dosimeters is suggested.

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INTRODUCTION

The present report describes an attempt to evaluate the characteristics of a water calorimeter for Co-60 dosimetry using a Thermos bottle as a sample container.

This particular system is chosen because it offers the possibility of calibrating in the same geometry, any diluted aqueous solution with similar absorption characteristics. In this way, the radiochemical yield* or biological response can be determined for any such solution, once the dose rate in the location is established by the absolute measurement.

The Thermos-bottle calorimeter has been used before by Klinger¹ who used a ten ounce Thermos bottle placed in a #10 can filled with ice and water. ** The starting temperature of the calorimetric body, i.e. water, was 0°C. The complete assembly was sent through a radiation field; after that the temperature was measured and the true temperature increase was extrapolated from knowledge of heat loss characteristics.

Brynjolfsson, et al.,^{2,3} used an ice-water system for absolute calibration of the Fricke dosimeter. However the basic concept of this system was quite different. The absorbed energy was monitored by the contraction in volume of an ice-water mixture, as part of the ice melted, and the change was detected by means of a calibrated capillary tube connected to the thermos bottle. In this experiment, the ice-water filled Thermos bottle was surrounded by a cylindrical jacket, also containing a mixture of ice and water, so that true isothermal conditions were realized.

* Radiochemical yield: G=number of molecular conversions per 100 eV absorbed energy

** A similar setup with water in the #10 can has been used in this Laboratory by Brynjolfsson and Jarrett (unpublished data)

For the present experiments it was desirable to irradiate fairly large samples, since G-value determinations require a highly accurate chemical analysis, preferably carried out with several determinations and by more than one analytical method. Therefore, it was decided to use a similar Thermos bottle set-up for the absolute calibration. The special design and the very high source strength of the cobalt facility (1 megacurie) prevented the use of the ice-water calorimeter described above. Further practical restrictions had to be observed. The set-up time had to be minimized because of the limited time the facility was available for use in this study. Therefore, the experiments used the standard conveyor carrier with no provisions for stirring during irradiation. Electrical power input calibrations of the system were made outside the cell.

Since the source plaques, and consequently the carrier doors, became quite hot during irradiation, it was decided to use a large Dewar as an additional temperature baffle.

Several different methods were employed for determining the energy input during irradiation. Experiments were carried out both for constant temperature environment and for near adiabatic modes of operation of the system. Electrical input calibrations were made similarly.

I. THEORY AND CONCEPTS OF RADIATION CALORIMETRY

Background:

The absorbed dose of radiation in a material is defined in terms of locally absorbed energy. The unit rad for absorbed dose was established by the International Commission on Radiological Units and Measurements in 1953.⁴ (1 rad = 100 erg/gram of the absorbing material). Calorimetry can be employed to measure directly the absorbed dose in a given sample by determining the heat formation, provided all dissipated energy appears as heat.

General Considerations:

In addition to the restrictions imposed on any conventional type of calorimetry, extra features have to be taken into account for radiation calorimetry. Petree and Ward⁵ give a very good discussion on this matter and point to the following general aspects:

"Size: Dose rate gradients are generally present in irradiated media. Consequently small calorimeters rather than large ones are generally required for measuring effectively the absorbed dose at a given point in the medium. This size limitation appears to be a serious difficulty in the calorimetric dosimetry of soft X-rays, where the half-value layer in aluminum is less than a few millimeters."

"Homogeneity: The dose rate at a point in a medium is affected by nearby voids or imbedded masses of different atomic composition. Consequently the calorimeter should be of a simple compact

design. Foreign matter, as for example, thermocouple wire, should be minimized."

"Chemical
Stability:

Radiation energy absorbed by a molecular substance generally has noticeable chemical effects in which energy is absorbed or released. When this occurs, the thermal effect, to which the calorimeter responds, does not correspond precisely to the quantity of energy initially deposited by the radiation. More than 2% of γ - Co^{60} energy, absorbed in polystyrene fails to appear as heat and is apparently stored in the altered chemical structure of some of the molecules. On the other hand, non-molecular substances, such as metals, store energy only by atomic dislocations, and this amounts to only about 0.001 percent of the radiation energy absorbed. Such materials should be used for calorimeters where possible."

Our calorimeter is very much different from the graphite sphere and aluminum cylinder devices designed by these authors. However, the above criteria are relevant for any type of calorimeter.

Dose-rate gradients pose no serious size limitation in our case. Measurements with chemical dosimeters carried out in a Maconite phantom⁶ indicate that in the geometry used, these variations do not exceed 2 - 3%. Furthermore, the determination of an absolute average dose rate which can be related to the change in a chemical system is the objective of the work, and for this reason

a large container is preferred. Nevertheless, because of the large volume of the calorimetric body, temperature gradient problems arise. This is discussed in detail later in this report.

The homogeneity of the Thermos bottle system is by no means excellent. Ideally the container should have been made up by a material having the same electron density as the water. This requirement, however, is not compatible with the requirement for chemical inertness and for good thermal insulation as is obtained with the Thermos bottle. Therefore, it is necessary to know the absorption characteristics of the glass and include this information in the dose calculations. Similar information with respect to cork stopper, and thermocouple wire, etc., has to be included. We have used published data and made additional experiments for checks on these characteristics. This is also described in a later section.

As for the chemical stability, it must be realized that a certain part of the energy deposited by the irradiation is turned into dissociation of the water (radiolysis). However, the energy absorbed in this manner in a closed system is negligible once a steady state condition exists between the primary radiolysis and the back reaction forming H_2O from H_2 and H_2O_2 .

The rate of the radiolysis may be followed by the formation of molecular hydrogen. In an open system, where the hydrogen can escape (e.g. by bubbling a gas through) this yield is of the order of 0.4 - 0.5 molecules for 100 eV absorbed energy. The corresponding decomposition of water requires only a few electron volts, thus it can be seen that the part of the total absorbed energy used in the chemical reaction cannot exceed a few percent. Hochanadel,⁷ Gordon and Hart⁸ have found that when H_2 is not removed, i.e. the system

is closed, a back reaction builds up, where water is reformed from the molecular products H_2 and H_2O_2 . At a steady state the net yield of H_2 production drops to approximately 3% of that for the open system. This means that only a small fraction of a percent of the energy is consumed by the chemical reaction.

According to Lazzo et al., this steady state occurs when less than 10^{18} molecules of H_2O_2 per liter are formed. Assuming $G_{H_2O_2} = 0.71^{10}$, this will correspond to a dose of:

$$\frac{10^{18} \cdot 100}{10^3 \cdot 0.71} \text{ eV/g} = 1.4 \times 10^{17} \text{ eV/g} = 2.2 \times 10^3 \text{ rads}$$

Even assuming that a somewhat larger dose is required due to a small air space above the solution, one is justified in concluding that steady state occurs so quickly that no correction is necessary for chemical effects at total doses in the megarad region.

It is not expected that any appreciable amounts of energy will be stored in the form of atomic dislocations in the irradiated glass. (See also p. 4).

Modes of Operation:

The ideal mode of operating a calorimeter is at adiabatic conditions, where the ambient temperature is adjusted continuously to be identical to the temperature of the detecting body. This means that heat losses by radiation, convection, and conduction, are minimized if not eliminated. Several calorimetric systems of this kind have been described in the literature, all including a power-compensating electronic circuit for heating the environmental material, i.e. a jacket.

In our case the heating rate of the water in the jacket exceeds that of the water in the bottle, partially because of heat transfer from the conveyor carrier and partially because of the slightly higher dose rate. This means that heat would have to be removed rather than supplied, calling for a circulatory water cooling system, which is not desirable for practical reasons. A description will be given later of how an attempt to approach adiabatic conditions was made by making heating-rate measurements at the time during irradiation when the jacket and sample temperature curves intersected. An alternative mode of operation is the constant temperature environment mode, in which the jacket is held at a constant temperature throughout the experiment. This condition was realized by having an ice-water mixture in the jacket.

Heat losses by convection or radiation are believed to be negligible in this experiment due to the characteristics of the Thermos bottle. Convection is kept at a minimum because of the vacuum mantle, and emission of light in form of radiation will be reflected to a large extent by the silver coated inner liner of the Thermos bottle and eventually be absorbed in the system.

Nevertheless, heat losses from conduction through the neck of the Thermos bottle are appreciable and, in fact, give rise to some of the problems described later.

In general,* heat losses are governed by Newton's law of cooling, which may be expressed by the following equation:

$$\frac{\Delta T}{\Delta t} = K (T_{\text{sample}} - T_{\text{jacket}}) \quad (I)$$

* Strictly speaking, discrimination should be made between the three different sources of heat losses. Convection and conduction losses can both be described by (I), but have independent K-values. Radiation losses are governed by $(T_{\text{sample}} - T_{\text{jacket}})$ in the fourth power, but play a negligible role in this setup. A complete discussion on these subjects can be found in Ref. 10.

where K is a constant, characteristic of the system, which can be determined experimentally with very good accuracy (Appendix 5). When K is determined experimentally, it will account for all three types of energy losses from the system.

By observing the variation in the sample temperature up to and for a time after irradiation, these temperature curves may be extrapolated to the middle of the irradiation. This extrapolation corrects for heat losses to the jacket, giving the temperature change for adiabatic conditions.

Thus the true temperature increase can be determined. Likewise the temperature increase per unit time in the system can be determined from the observed slope of the temperature curve, by correcting this curve for the known heat loss per corresponding unit of time.

Calculations:

The absorbed dose rate in the system may be calculated from the following formula:

$$D_r = \frac{\sum m_i \cdot c_i}{\sum m_i \cdot a_i \cdot b_i \cdot 2.389} \cdot \frac{\Delta T}{\Delta t} \quad \text{Mrads/h.} \quad \text{(II)}$$

where:

D_r = Dose rate, Mrads/h.

m_i = Mass of the "i" component, g.

c_i = Thermal capacity of the "i" component, cal/g^oC.

a_i = Correction factor for the different absorption characteristics per unit mass of water and the "i" component.

ΔT = Temperature increase in ^oC per hour of irradiation

Δt = (followed by the change in microvolts from the thermocouples).

The factor 2.389 is the number of calories equivalent to 1 gram-megard, or 10^8 ergs. The contribution from a_i and b_i is very small and can be neglected except for the glass which represents a relatively large fraction of the total mass.

System Calibration:

Although some of the constants included in the formula above are either known or relatively easy to determine with good accuracy, it is desirable to check on the system as a whole by introducing a precisely known amount of energy into the system and observing the temperature increase. To do this an electrical power supply is used to heat a resistance wire placed in the bottle (in some experiments another wire was heated simultaneously in the jacket to simulate adiabatic conditions). Voltage, current, and time are then measured accurately so that the energy deposited can be determined.

The calorimetric equation for the absorbed electrical energy is the following:

$$E_{elec} = \frac{\sum m_i \cdot c_i}{0.2389} \cdot \Delta T \text{ joules} \quad (III)$$

A common approach, which is especially applicable to homogeneous calorimeters, is to rely upon the calibration heating as the energy standard. This means that neither absolute calibration of the thermocouples nor knowledge of the thermal capacity of the system is necessary.

This is shown by substituting equation III into II; the dose formula is then reduced to:

$$D_r = \frac{E_{electric}}{10 \cdot \Delta t \cdot \sum m_i \cdot a_i \cdot b_i} \text{ Mrads/h}$$

and the absorbed dose rate is readily determined provided the respective masses and the correction constants a_i and b_i are known.

This method of system calibration and dose determination certainly can be used for a rough determination of the absorbed dose in the system. There are some reasons, however, why it is not adequate for accurate measurements with the Thermos bottle calorimeter. First, since the thermoelectric power of the thermocouple measuring junction is not a completely linear function of the temperature, the same temperature intervals should be used for each calibration and irradiation. This is often not possible. Because of the different modes of heat input and, thereby, different heat distributions in the two procedures, it is difficult to obtain similar temperature increases after system equilibrium is reached. This means that a calibration of the thermocouples (or use of reference tables) is necessary in order to compare the calibration and irradiation experiments.

In addition to this, it is desirable to have this calibration available, when detailed information is desired concerning the sample and jacket temperatures in the course of the experiments.

Second, although masses of water, wires, and cork can be determined to sufficient accuracy by direct weighing, the situation is different with respect to the glass. A "mass" can be determined after conclusion of all experiments by breaking the Thermos bottle and weighing the inner liner only, but this mass is not necessarily the most accurate one, as the "effective" part of the bottle is not too well defined. There is a part of the neck which has heat transfer both to the inner and outer system, thus only partially contributing to the mass of the calorimetric body.*

* See also Appendix II.

The following improved method for the system calibration, which overcomes these difficulties, is therefore suggested. The thermocouples should be calibrated carefully against a normal thermometer and the change in thermoelectric power per °C be plotted as a function of °C on a large graph. (See also Appendix I). Masses of water, wires, and cork should be determined by weighing and the corresponding thermal capacities obtained from standard reference tables. By substituting all these values into II, a calibration heating should give an accurate value of the product $m_{\text{glass}} \times c_{\text{glass}}$. The value of c_{glass}^* may be obtained from a reference table, but this requires detailed knowledge about the composition of the glass and, to some extent, of the degree of crystallization which are not always available for a commercial product. However explicit values for c_{glass} and m_{glass} can be determined simultaneously by an additional experiment in the same setup; by adding a weighed amount of broken glass from an identical Thermos bottle to the water in the Thermos and determining the new value of $m_{\text{glass}} \times c_{\text{glass}}$. This gives us two equations which can be solved for m_{glass} and c_{glass} .

By now all necessary constants except a_{glass} and b_{glass} in equation II are accounted for. The value of a_{glass} can be obtained from the dose distribution curve referred to earlier.⁶ An accurate value for b_{glass} may be obtained from reference tables, if one has an exact knowledge of the composition of the glass and of the energy spectrum in the irradiation field. This approach, however, gives rise to various problems which are described in detail in Appendix II. It is therefore suggested that an experimental determination of b_{glass} be made by a method corresponding to the one described above for the determination of c_{glass} .

* See also Appendix II.

In the present case, there is no evidence that the defendant knew or had reason to know that the information was false or that the defendant intended to defraud the bank. The defendant's failure to disclose the information is not sufficient to establish a claim of fraud. The defendant's failure to disclose the information is not sufficient to establish a claim of fraud. The defendant's failure to disclose the information is not sufficient to establish a claim of fraud.

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II. EXPERIMENTAL DETAILS

Irradiation Source:

The megacurie Co-60 source at the U. S. Army Natick Laboratories consists of 98 source rods arranged in two 107 by 142 cm plaques separated by 28 cm. Each plaque has an average activity of about 35 curies per square cm. The source is stored below 20 feet of water and for use is raised to position above the water by an elevator system (see Figure 1). The absorbed dose rate between the plaques is of the order of 4 Mrads per hour.

Conveyor System:

The conveyor system is designed primarily for carrying food into position between the source plaques. It consists of chain-driven carriers along with the necessary stops and controls for either a continuous pass between the plaques or for bringing the carrier to exact position between them. Furthermore, one has the alternative of bringing the carrier into position first and then raising the source around it. This is the method used for the calorimetry. The carriers are made up of aluminum frames with $\frac{1}{4}$ " aluminum doors.

Calorimeter Assembly:

The calorimeter consists of a commercially available 0.3 liter Thermos bottle* positioned in a 4.5 liter cylindrical Dewar by means of Masonite spacers. The Thermos bottle was sealed with a plastic-covered cork, and the jacket was closed with a tight-fitting Styrofoam top (see Figure 2).

The thermocouples and the calibration heating wire, when in use, were inserted through either a hole in the cork or between the cork and the neck of the bottle. In most of the experiments the thermocouples were hanging

* No. 2015, King Seeley Thermos Company

freely down into the solution, but in some cases where information was sought for boundary locations they were spaced to the desired position at the wall. In some of the experiments, when a corrosive solution was to be irradiated, a glass tube sealed on the end was inserted through the cork to hold a thermocouple.

The thickness of the glass walls of the Thermos is 1.5 mm; those of the jacket walls are approximately 3 mm.

Temperature Measurements:

Thermistors as well as thermocouples were considered for temperature detection. Thermistors have a greater sensitivity but can be slightly more laborious to read. Since the temperature increases in these experiments were of the order of 10°C, the sensitivity of the thermocouples was sufficient, so they were chosen for ease of operation.

Honeywell .05 mm Copper-Constantan wire was used. Marlin* thermocouple connectors were employed to connect them to 1/8" wire running from the irradiation area to the control area. By shielding and guarding all the wires, a low noise level was obtained.

The circuit for the temperature measurements (Figure 3) included a Leeds & Northrup K-3 potentiometer, a Leeds & Northrup electronic galvanometer and a Joseph Kay & Company, Inc., triple point thermocouple reference.

The thermocouples used were selected on the basis of identical response from a large number of units calibrated against a mercury normal thermometer in a big temperature controlled water bath. The change in microvolts per degree centigrade as a function of temperature was found to agree better than 1% with tabulated values. Experimental calibrations are tabulated and graphed in

Appendix I.

* Marlin Electronics Corporation, Cleveland, Ohio

Power Input for System Calibration:

In order to get an even heat distribution in the water during the electrical calibration, a long resistance wire was curled into a spiral and placed in the Thermos as shown in Figure 2. The positioning was done very carefully in order to avoid short circuits. The resistance of the wires used varied from 15 - 75 ohms.

The power supply used was a 0.1% stable John Fluke power supply. The circuitry for the power input measurements and the temperature measurements is shown in Figure 3. A high wattage resistor was used in series with the heating wire to obtain the current readings. In order to determine the voltage, a potential divider was placed in parallel with the heating wire. All resistors were calibrated in a Wheatstone bridge before and after each experiment in order to assure that no appreciable heating effect occurred. The Leeds & Northrup K-3 potentiometer and the null detector were used in making current and voltage readings, as well as in the readings of the thermoelectric power. An electrical timer was started simultaneously with the current by means of a double-pole switch.

III. EXPERIMENTAL RESULTS

In this section three calibration experiments and three temperature determinations are described. The calibrations, of course, are aimed at simulating the irradiations. Therefore, each of the calibration experiments is related in some way to one or more of the irradiations. Although it may have been more correct to describe two experiments in pairs, this was not done because the calibrations served a more direct purpose, namely to obtain values for the effective mass and the thermal capacity of the part of the Thermos flask included in the calorimetric system. These values should be constant under all conditions (except for temperature dependence for glass), so the experimental values give a mean for evaluating the accuracy of the different procedures.

Calibration No. I

General:

The first calibration represents an attempt to check the integrated system. Efforts were made to obtain adiabatic conditions by heating the water in the jacket at the same rate as the water in the bottle. This operation was performed manually by following the temperature of the two bodies and adjusting the power input to the jacket at frequent intervals.

To check the size and persistence of temperature gradients, five thermocouples were placed in different positions in the Thermos and in the jacket. Temperature readings were taken from 2 hours before heat input to 20 hours after.

Observations:

(1) Rather large temperature gradients exist, which require 5 to 6 hours to subside. (See Figure 4).

(1) Whether it is possible to use a logarithmic extrapolation of the curves, once equilibrium is obtained, is practical or accurate since the change in the temperature difference between sample and jacket is not regular until about one half hour after termination of power input.

(2) The possibility of determining the heating rate from the slope of the temperature curve was found to be impractical because of electrical interaction between the thermocouples and the heating coils during the power input.

Results: (For data see Appendix III)

Since extrapolation was not possible, an average sample temperature immediately before power input and one hour after power input was found from the temperatures at the top and bottom of the Thermos bottle. A weighted average can be evaluated from the curves shown in Figure 6. For the intervening time, the average difference between the sample and jacket temperature was found, and by application of Newton's Law, the average rate of heat transfer from the sample to the jacket was evaluated.

The true (corrected) increase in temperature was found to be $9.09 \pm .01^\circ\text{C}$ for an electrical input of 1159.0 ± 1.0 joule.

From Equation III, the value of $m_{\text{glass}} \cdot c_{\text{glass}}(18.6^\circ)$ was calculated to be 15.7 ± 0.8 .

Calibration No. II:

General:

The second calibration had the same scope as the first one: The checking of the integrated system. The mode, however, was different since this calibration was carried out in a nearly constant temperature environment.

To cope with the large temperature gradients present in the sample during the former experiment, vigorous shaking of the Thermos bottle was introduced immediately after the termination of heat input.

The jacket temperature was uncontrolled, at room temperature. Logarithmic extrapolations to the middle of the heating period were made by following the temperature curve from about one hour before until three hours after heat input.

Observations:*

(1) Shaking after heat input practically eliminated the temperature gradients referred to above. From 15 minutes after shaking and thereafter, the largest temperature difference in the Thermos was found to be of the order of 0.06°C both in the calibration and in the irradiation experiments. (See dose-rate determination No. II).

Since the past history of the heat distribution in the Thermos becomes relatively unimportant after the shaking, this calibration becomes a promising method for obtaining not only a check of the integrated system, but a determination of the values of both the effective mass of the glass in the Thermos and the corresponding thermal capacity to be employed later in the dose-rate determinations.

(2) The drift in temperature of the jacket during the time in which readings were made amounted to 0.5°C .

Results: (For data, see Appendix III)

Logarithmic extrapolations of the temperature curves were made to the middle of the heating period by application of Newton's Law. The jacket

The temperature curves were similar to those of Calibration No. IIIA, Fig. 7.

temperature was assumed to be constant and equal to its average value during the experiment. The true increase in temperature was found to be $3.98 \pm 0.01^\circ\text{C}$ for an electrical input of 2656 ± 5 joules.*

From Equation III, the value of $m_{\text{glass}} \cdot c_{\text{glass}}(25^\circ)$ was calculated to be: 15.6 ± 0.8 .

Calibration No. III

General:

In the first two calibrations, values for $m_{\text{glass}} \cdot c_{\text{glass}}(T)$ were obtained, and it is seen that the two modes of operation gave good agreement in the results. However, explicit values for $c_{\text{glass}}(T)$ and m_{glass} are needed. In addition it is desirable to obtain these values with better accuracy than obtained in the former experiments since they participate with a relatively large weight in the dose equation (II). As described earlier, this information may be obtained through a two-step calibration, where, in the second calibration, a weighed amount of glass, m_{glass}^1 , identical in composition to that of the Thermos bottle, is added to the water. This gives two equations, which can be solved for m_{glass} and $c_{\text{glass}}(T)$:

$$m_{\text{glass}} \cdot c_{\text{glass}}(T) = K_1$$

$$(m_{\text{glass}}^1 + m_{\text{glass}}) \cdot c_{\text{glass}}(T) = K_2$$

A constant temperature environment mode of operation was chosen because of the ease in the procedure and extrapolation of the temperature curves. The accuracy was improved by using an ice-water mixture in the jacket, assuring

* The electrical energy was determined with less accuracy than usual due to occasional shorts in the heating coil.

a constant ambient temperature. The use of ice-water mixture in the jacket also makes it possible to perform irradiation experiments under comparable conditions.

The calorimeter was shaken vigorously after termination of the heat input.

Observations:

With an ice-water environment, large temperature differences between the jacket and the sample lead to a rapid rate of temperature change, decreasing the accuracy of the extrapolation. From some of the early experiments, as well as the present one, it was concluded that in order to obtain an accurate extrapolation, the heating (or irradiation) should be started with the sample temperature not more than a few degrees above 0°C.

Results:

Logarithmic extrapolations of the temperature curves were made to the middle of the heating period by application of Newton's Law. The true increase in temperature amounted to $9.332 \pm 0.012^\circ\text{C}$ (A) and $7.446 \pm 0.012^\circ\text{C}$ (B) for an electrical input of 12207 ± 4 joules and 8873 ± 4 joules respectively.

From Equation III, the values for $m_{\text{glass}} \cdot c_{\text{glass}}(T)$ and $(m_{\text{glass}} + m_{\text{glass}}^1) \cdot c_{\text{glass}}(T)$ can be calculated to be:

$$m_{\text{glass}} \cdot c_{\text{glass}}(8^\circ) = 15.37 \pm 0.30$$

$$(m_{\text{glass}} + m_{\text{glass}}^1) \cdot c_{\text{glass}}(6.8^\circ) = 40.60 \pm 0.30$$

Assuming that $c_{\text{glass}}(8^\circ) = c_{\text{glass}}(6.8^\circ)$, these two equations may be solved for $c_{\text{glass}}(8^\circ)$ and m_{glass} .

$$c_{\text{glass}}(8^\circ) = 0.208 \pm 0.003 \text{ cal/gm} - ^\circ\text{C}$$

$$m_{\text{glass}} = 74.0 \pm 1.5 \text{ gm}$$

Discussion of Calibration Data:

Insertion of $m_{\text{glass}} = 74.0 \pm 1.5$ gm into the product $m_{\text{glass}} \cdot c_{\text{glass}}(T)$ determined in Calibrations I and II gives the experimental values for $c_{\text{glass}}(T)$. The three calibrations are summarized in the following table:

T°C	$c_{\text{glass}}(T)$
8	0.208 ± 0.003
18.6	0.212 ± 0.005
25	0.211 ± 0.005

Referring to the formula for c_{glass} as function of the temperature given in Appendix II, it may be seen that the determinations are consistent within experimental accuracy in this respect. The deviations assigned to the values given above and in the appendix are estimated on the basis of deviations expectable or experienced of each parameter.

It is concluded that a constant temperature environment mode of operation offers a satisfactory procedure for determination of m_{glass} and $c_{\text{glass}}(T)$ in a Thermos bottle calorimeter.

Dose Rate Determination I

General:

The first irradiation experiment to be described here was carried out in a nearly adiabatic mode, representative of several earlier attempts to determine the absorbed dose rate in a crude way.

The term "nearly adiabatic mode" needs a few explanatory comments. In the case of the calibration, the nearly adiabatic mode was realized by regulating the jacket temperature to follow the temperature increase (and level) of the

sample. The pattern could not be executed for the irradiation because the whole assembly was within the radiation field. Consequently the radiation absorption in the jacket plus the heat transport to the jacket from its surroundings resulted in a faster temperature increase in the jacket than in the sample. In this context "nearly adiabatic conditions" refers to controlling the temperature difference between the jacket and the sample by starting out with a somewhat lower jacket temperature so chosen to keep $(T_S - T_J)$ at a minimum following the irradiation.

In the preliminary experiments, from which data are not reported here, no particular care was given to controlling or recording accurately the jacket temperature. The temperatures in the samples were followed by a thermocouple, placed in a narrow glass tube, sealed on the end and inserted through the cork. Extrapolations were made to the middle of the irradiation by following the temperature for a short period before and after irradiation.

Because of results which fluctuated several percent, and because of the rather odd shapes of the temperature curves after irradiation, which could be only attributed to temperature equilization processes, it was decided to check on the size and persistence of temperature gradients by placing thermocouples in various positions in the Thermos bottle and in the jacket.

Temperature readings were taken for two hours before and twenty hours after irradiation.

Observations: (See Figure 8)

(1) As was experienced in Calibration I, large temperature gradients existed, which required five to six hours to subside.

(2) The temperature distribution and equalization curves permit the determination of the relative weight to be attached to each thermocouple reading in making an estimate of the average temperature. It can be seen, qualitatively, that the arithmetic average of the top and bottom temperature in the sample is a realistic choice.

(3) Logarithmic or linear extrapolation from the time-temperature curve, once equilibrium is achieved, is neither practical nor accurate, since the change in temperature difference between the sample and the jacket is not regular until about an hour after irradiation.

(4) No difficulty was encountered in making accurate temperature measurements during irradiation. It should be noticed, however, that there was an apparent displacement in the thermoelectric power readings from the thermocouples during irradiation. The displacement remained constant during irradiation and could easily be determined. The magnitude was around $15 \mu\text{V}$. It is probably due to different ionization in the air and in the system as a whole, including the thermocouple wires, jacket, and Thermos bottle.

Results: (See Appendix III for data)

An identical procedure to that used in Calibration I was employed in obtaining the results. The true temperature change was determined to be $12.52 \pm 0.08^\circ\text{C}$ for an irradiation time of 3000.0 ± 0.5 sec. Using the value of b_{glass} found in Dose-Rate Determination No. II, for a_{glass} the value obtained from dose distribution curves⁶ and the values m_{glass} and c_{glass} as obtained in the calibration experiments, the dose rate may be calculated from Equation II to be:

$$D_p = 4.18 \pm 0.04 \text{ Mrads/h}^*$$

* This as well as the following dose-rate determinations is corrected to be valid at June 16, 1964. Half life of Cobalt-60 taken to be 5.3 years.

General

The second experiment consists of two irradiations carried out in a constant temperature environment, maintained by having an ice-water mixture* in the jacket. This experiment is complementary to Calibration No. III in that the two heatings and the two irradiations are performed in the same mode and, combined, give experimental values for m_{glass} , c_{glass} , b_{glass} , and the dose rate D_r .

The correction for the different absorption characteristics in the glass and in the water b_{glass} is here determined simultaneously with the dose rate D_r in a two-step irradiation, where a weighed amount of glass, (21.2 gm) identical in composition to that of the Thermos flask, is added to the water in the second irradiation. This gives a set of equations, which can be solved for D_r and b_{glass} .

The procedure was identical to that of Calibration No. III.

Observations:

As in the case of the parallel calibration, it was found that after shaking, extrapolation of temperature curves was straightforward and is expected to give very reliable results. Nevertheless, it should be mentioned that when additional glass was added, extra time was required for all the glass at the bottom of the Thermos (as demonstrated in Figure 2) to come to temperature equilibrium with the water around it, even after shaking.

* The slightly smaller attenuation of the γ -rays in an ice-water mixture would be expected to cause a higher dose rate in the sample. However, because of the two-plaque source configuration in this facility, the change in dose rate, as estimated from dose-distribution curves, is less than 0.1%, and therefore not accounted for in the results.

Results: (See Appendix III for data)

Extrapolation of the time-temperature curves was made to the middle of the irradiation. For the two irradiations A and B, temperature increases of $6.079 \pm 0.005^\circ\text{C}$ (A) and $7.742 \pm 0.005^\circ\text{C}$ (B) were determined for an irradiation time of 1800.3 ± 0.1 sec and 1800.2 ± 0.1 sec respectively.

From Equation III, the following simplified equations can be derived:

$$D_{r(A)} (b_{\text{glass}} + K_1) = K_2$$

$$D_{r(B)} (b_{\text{glass}} + K_3) = K_4$$

where K_1 , K_2 , K_3 , and K_4 can be expressed in terms of the known parameters.

$D_{r(A)}$ and $D_{r(B)}$ are related by the decay equation:

$$\frac{-0.693}{5.3} \Delta t$$

$$D_{r(B)} = D_{r(A)} \cdot e$$

correcting for the time elapse between the two experiments.

Solving these two equations for D_r and b_{glass} gives:

$$\underline{D_r = 4.42 \pm 0.02 \text{ Mrads/h}}$$

$$\underline{b_{\text{glass}} = 0.98 \pm 0.02}$$

Dose Rate Determination III

General:

As previously stated, the ideal mode of operation is adiabatic, since no heat losses occur; therefore, correction of the observed temperature increase is unnecessary. It has also been pointed out that, owing to the large heat increase in the jacket during irradiation, this mode is impractical under the present conditions.

If, however, the jacket temperature is lower than that of the sample at the start of the irradiation and higher at the termination, intersection of the temperature curves must occur, and at this time adiabatic conditions will exist. By recording accurately both the temperature and time for a period starting before crossing takes place and stopping thereafter, the slope of the temperature curve can be obtained for adiabatic conditions of the sample. The slope is a direct measure of the absorbed dose rate.

This experiment describes a total of three irradiations, A, B, and C, where the intersections take place after different periods of irradiation (i.e. different $(T_{\text{jacket}} - T_{\text{sample}})$ at the start of each irradiation.)

Observations: (See Figure 10)

(1) The slope of the temperature curve (i.e. the heating rate) is not only dependent upon the dose rate, but also on the duration of the irradiation preceding adiabatic conditions. The reason for this indicated time dependence is the rather large temperature gradients existing in the system during irradiation. It was experienced, however, that after about one hour these gradients became relatively stationary in size, meaning that the rate of heat transfer

from the area of low specific heat, the glass, to the area of high specific heat, the water, is constant. Measuring the slope of the temperature curve during adiabatic conditions after such a "steady state" is achieved, should permit a reliable and accurate determination of the dose rate.

Figure 11 shows the heating rate of the sample as monitored by the thermocouple as a function of irradiation time. Each value $\frac{\Delta T}{\Delta t}$ has been corrected for heat losses to the jacket during the time Δt , by means of Newton's Law.

The curve is obtained from Irradiation C. The point X_C denotes the heating rate at the time when the jacket and sample temperature curves intersect. The points X_A and X_B are the corresponding sets of values found in the first two irradiations in this section. The graph indicates that for this particular system, stationary conditions are achieved after about one hour of irradiation, meaning that the starting temperatures of sample and jacket should be selected so that intersection of the temperature curves does not occur sooner than one hour after the start of irradiation. Positioning of the thermocouples in the jacket and sample are only important for determining the crossing point (for this they should be positioned near the neck of the Thermos). The fit of the points X_A and X_B to the curve indicates that once this curve is determined, it can be applied to correct heating rates measured under pre-stationary conditions.

A very small systematic error may be expected from this method compared to the constant temperature environment method, since the cork and neck of the bottle may not contribute as much to the effective mass as when shaking is performed after the irradiation. This means that determinations made in this way may be slightly low.

Results:

A. The rate of temperature rise at intersection (500 seconds after start of irradiation) was:

$$\frac{\Delta T}{\Delta t} = 0.003205 \pm 0.000020 \text{ } ^\circ\text{C/sec}$$

corresponding to a dose rate of:

$$D_r = 4.03 \text{ Mrads/h}$$

Stationary conditions as defined above were not obtained. If correction is applied from Figure 11, the following dose rate is found:

$$D_r = 4.18 \pm 0.04 \text{ Mrads/h}$$

B. The rate of temperature rise at intersection (750 seconds after start of irradiation) was:

$$\frac{\Delta T}{\Delta t} = 0.003205 \pm 0.000015 \text{ } ^\circ\text{C/sec}$$

corresponding to a dose rate of:

$$D_r = 4.10 \text{ Mrads/h}$$

Stationary conditions as defined above were not obtained. If corrections are applied from Figure 11, the following dose rate is found:

$$D_r = 4.21 \pm 0.03 \text{ Mrads/h}$$

C. The rate of temperature rise at intersection (3300 seconds after start of irradiation) was:

$$\frac{\Delta T}{\Delta t} = 0.003333 \pm 0.000010 \text{ } ^\circ\text{C/sec}$$

corresponding to a dose rate of:

$$D_r = 4.20 \pm 0.02 \text{ Mrads/h}$$

This determination is obtained under stationary conditions.

Summary of Dose Rate Determinations:

The results of the dose rate determinations may be summarized as follows:

Determination No.	D_r Mrads/h (\pm estimated error)
I	4.18 ± 0.04
IX	4.22 ± 0.02
IIIA	4.18 ± 0.04
IIIB	4.21 ± 0.03
IIIC	4.20 ± 0.02

If these results are given weight according to their estimated error, the average can be calculated to be 4.205 Mrads/h. The dose rate in the irradiation position is then determined to be: (As of June 16, 1964)

4.205 ± 0.018 Mrads/h

IV. DISCUSSION

Deviation and Errors:

In an attempt to assess the errors introduced in the dose-rate determination, and the weight of each source of error on the result, all parameters have been accounted in the following Table:

Parameter	A: % Error On $D_r(I)$	B: % Error On $D_r(II)$	C: % Estimated Experimental Error	D: % Result- ing Error On D_r
m_{water}	0.14	3	0.03	0.09
m_{glass}	0.17	-	2	-
m_{cork}	0.003	0.003	25	0.086
m_{wire}	0.0001	< 0.0001	100	< 0.001
c_{water}	0.83	3	0.01	0.03
c_{glass}	0.0042	-	1	-
c_{cork}	0.0002	0.003	10	0.033
c_{wire}	0.0001	< 0.0001	100	< 0.001
a_{glass}	0.21	0.21	0.3	0.063
a_{cork}	0.006	0.006	5	0.028
a_{wire}	0.001	0.01	1	0.001
b_{glass}	0.28	-	2	-
b_{wire}	0.002	< 0.0001	10	< 0.0001
b_{cork}	0.006	< 0.0001	2	< 0.0001
$\mu V/^\circ C$	1	4.2	0.03	0.120
E_{elec}	-	3.2	0.03	0.096
μV	1	4.5	0.08	0.36
Estimate of Total Error:				0.4%

Column A gives the error in the dose-rate determination, when an error of 1% is introduced in any one of the parameters listed, thus indicating the relative importance of the errors. This picture, however, is too simplified to be used for estimating the accuracy of the integrated system. Many of the parameters are "interwoven," e.g. the experimental determinations of m_{glass} and c_{glass} are dependent on the accuracy of the power-input measurement, the determination of $\mu\text{V}/^\circ\text{C}$, and the reading of μV , etc. This will increase their impact as sources of errors in the dose-rate determination. On the other hand, because of the parallelism in the calibrations and the irradiations, errors in other parameters such as m_{wire} , c_{wire} , b_{cork} , and b_{wire} tend to negate each other.

To illustrate this, the error on the dose-rate determination introduced by a 1% deviation in any of the independent parameters is tabulated in Column E. Of the parameters playing an important role, only m_{glass} and b_{glass} cannot be determined directly. However, these were determined in terms of the other parameters along with c_{glass} and are therefore not included in Columns B and D.

In Column C and Column D the standard error in each of the parameters and the effect on the results is estimated for a typical experiment carried out in an adiabatic or a constant temperature environment mode. These estimates are of course only approximations with a questionable reliability. The best assurance of accuracy are reproducible results and experimental methods of checking dubious parameters.

The estimated errors for essential parameters such as $\mu\text{V}/^\circ\text{C}$ and μV may be considered fairly optimistic, when related to the equipment employed

in the experiments and even when realizing that these parameters are derived from a large number of readings. However, the estimate is supported by the calibration experiments.

Equation II can be written as follows:

$$E_{\text{elec}} = \frac{\sum m_i \cdot c_i}{0.2389} \cdot \frac{\Delta \mu V}{\Delta \mu V / ^\circ C} \quad \text{Joules}$$

A 1% error in any of the parameters E_{elec} , $\Delta \mu V$, or $\Delta \mu V / ^\circ C$ would cause a 20% error on the determination of m_{glass} , i.e. the experimental value would be 15 gm off.

The value for m_{glass} obtained in the calibration experiments and the value found by breaking the Thermos flask and weighing the inner liner are consistent. (See Appendix II). Although the mass obtained by the weighing may not be an accurate measure of the effective mass, it seems permissible from the results and the way in which they are obtained to assume that the two values should agree within 2%. If so, it may be concluded that the combined errors in E_{elec} , $\Delta \mu V$, and $\Delta \mu V / ^\circ C$ do not exceed 0.1%, this being consistent with the respective standard errors listed in Column C.

The estimate of the total error, 0.4%, is in agreement with the standard deviation obtained for the five dose-rate determinations.

The possibility of improving the experimental accuracy by introducing temperature controls in the jacket, so that true adiabatic conditions could be achieved throughout the experiment, seems doubtful because of the lack of uniformity in the absorption characteristics. By increasing the accuracy of the temperature readings, it may however be possible to obtain determinations with accuracies of $\pm 0.1\%$. Further improvements must incorporate better determinations of other parameters such as m_{cork} and m_{glass} .

General Conclusions:

This report has described how the Thermos bottle calorimeter can be applied to make accurate dose-rate determinations. The major part of the work concerned the problems originating in the inhomogeneity of the calorimetric body, comprised of such different components as the "inner" part of the Thermos bottle, the water, the cork, and the wires.

This inhomogeneity affects the results in two significant ways:

First of all it causes rather large temperature gradients in the system, which complicate the measuring of the temperature increases. However, it has been shown that this problem could be dealt with in several ways. When operating the calorimeter in a constant temperature environment, one could either await temperature equilization and correct for the heat losses during this period, or force an early equilibrium by shaking the Thermos bottle immediately after irradiation. Equally feasible was the determination of the rate of temperature increase at a time during irradiation, when both adiabatic conditions and stationary temperature gradients were present. Adiabatic conditions exist at the time when the temperature curves for the jacket and the sample intersect during the irradiation. It was found that both modes of operation rendered equally accurate results. Preference of one mode to the other may thus be dictated by practical conditions.

The second way in which the lack of homogeneity is manifested is in the absorption characteristics of the system. Accurate knowledge must be available concerning mass, thermal capacity, and radiation absorption capability for each component. In this respect the glass causes problems. The effective mass of the glass is not well defined physically and the specific heat and absorption capability vary significantly from one type of glass to another.

It was shown that this information can be obtained experimentally and simultaneously with the dose-rate determination by doing two sets of calibrations and irradiations; in the second set, a weighed amount of glass, identical in composition to that of the Thermos bottle is added to the water. The findings were consistent with those which could be estimated by other means.

This set of experiments was carried out in a constant temperature environment, since it was felt this mode offered a greater thermal similarity in the calibrations and the irradiations. It was demonstrated, however, that if the parameters determined in this way are used for a dose-rate determination, carried out under adiabatic conditions, equally accurate and precise results can be obtained in a simple and straightforward procedure.

In our experiments the constant temperature environment was realized by placing the Thermos bottle in a large Dewar filled with a mixture of ice and water. For applications with underwater sources, however, the large constant temperature volume of water generally used, makes an ideal constant temperature jacket, if adequate agitation is provided.

Comparison With Other Methods:

To place the Thermos bottle calorimeter in the right perspective, it should be emphasized that for an absolute calibration of the strength of a source, or for studying the field surrounding it, other types of calorimeters, especially of the homogeneous type have proven excellent and may be superior for these applications. However, for determining the absorbed dose rate in an aqueous solution in a relatively large and uniform field, the Thermos bottle calorimeter serves dual functions by acting as a convenient sample holder and an accurate measuring instrument.

For measuring radiochemical or radiobiological yields, its advantages should be obvious. There is no need for calculations to transform the absorbed dose from one material to another, nor to correct for different geometries or for surface-to-volume effect in the sample container, as is the case where a graphite or metal calorimeter is used. As long as the chemical or biological system in view does not contain large amounts of heavy ions, which could change the energy spectrum in the bottle appreciably, only minor corrections for different absorption characteristics may have to be applied for obtaining the absorbed dose in the sample relative to that received by the pure water.

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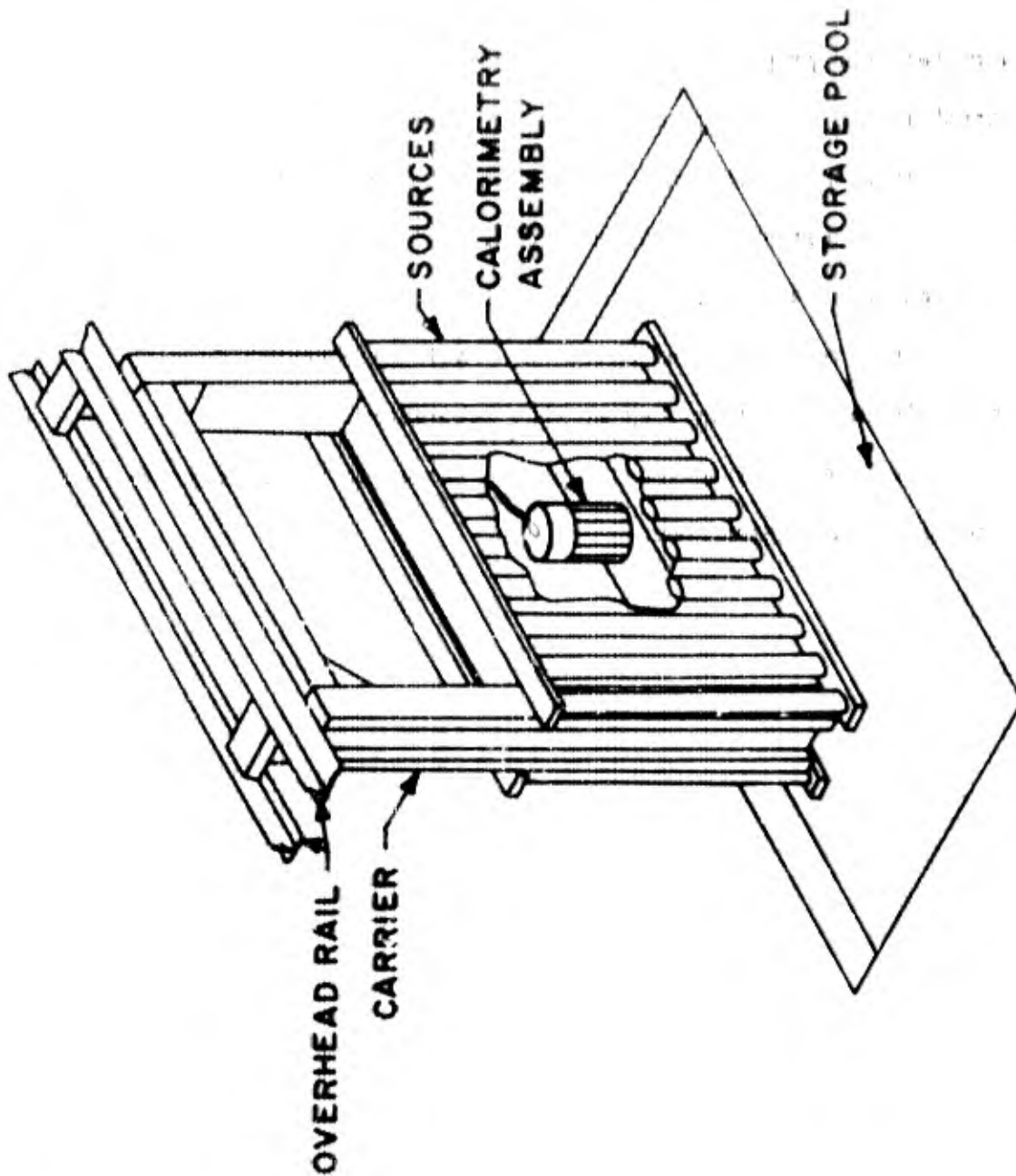


FIG. 1 SOURCE ARRANGEMENT

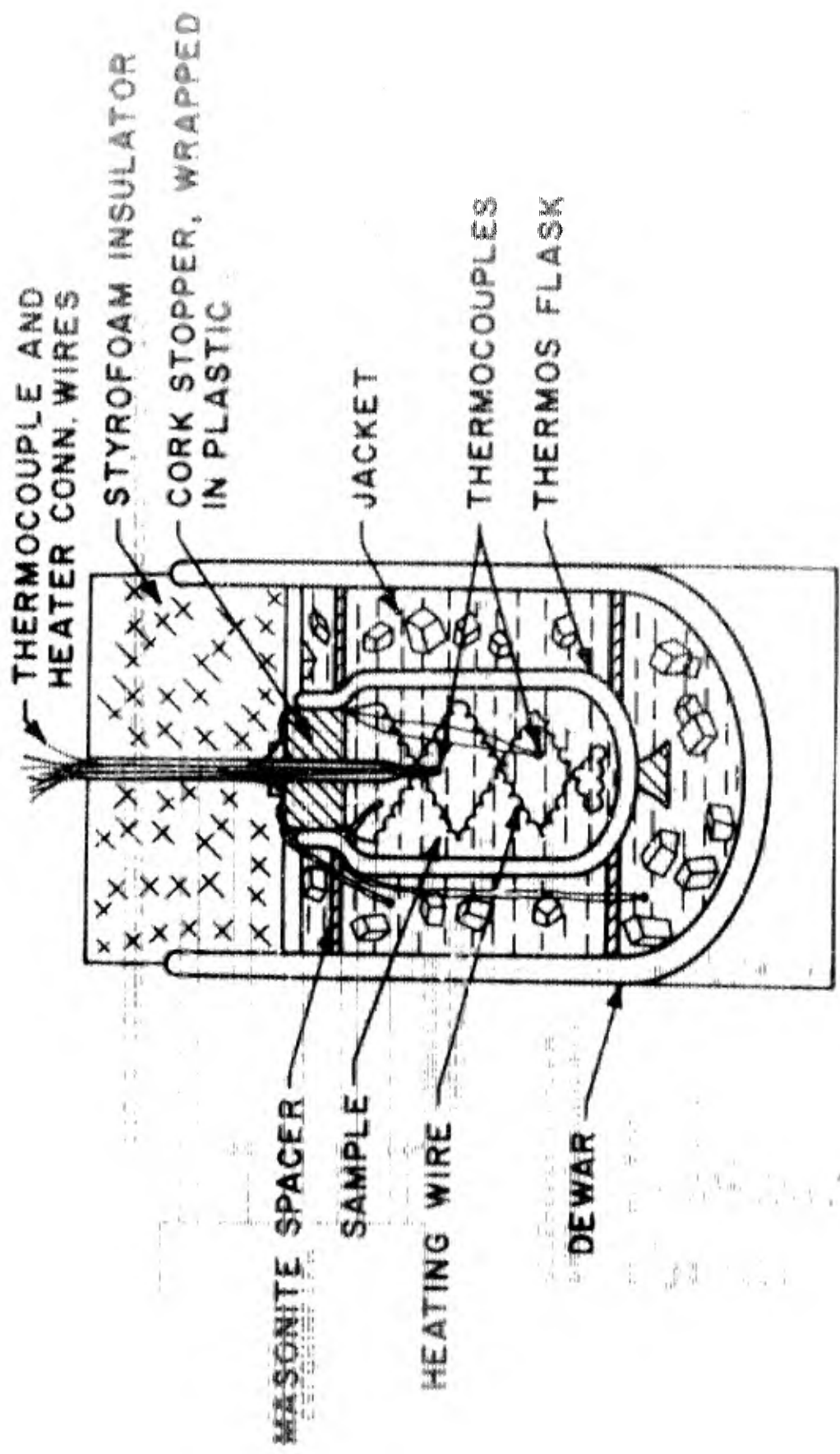


FIG. 2 CALIBRATION ASSEMBLY

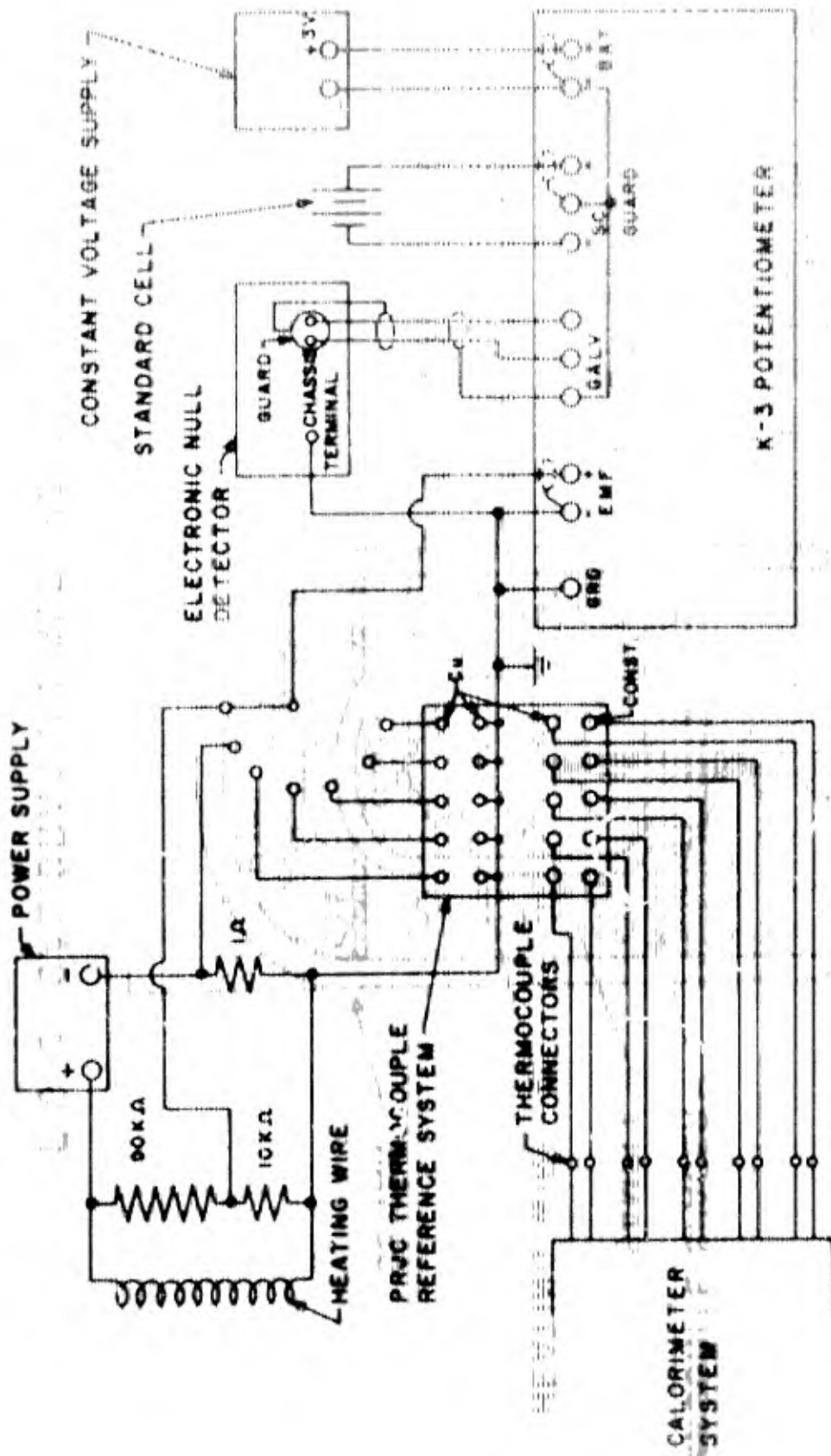


FIG. 3 TEMPERATURE AND POWER MEASUREMENT CIRCUITS

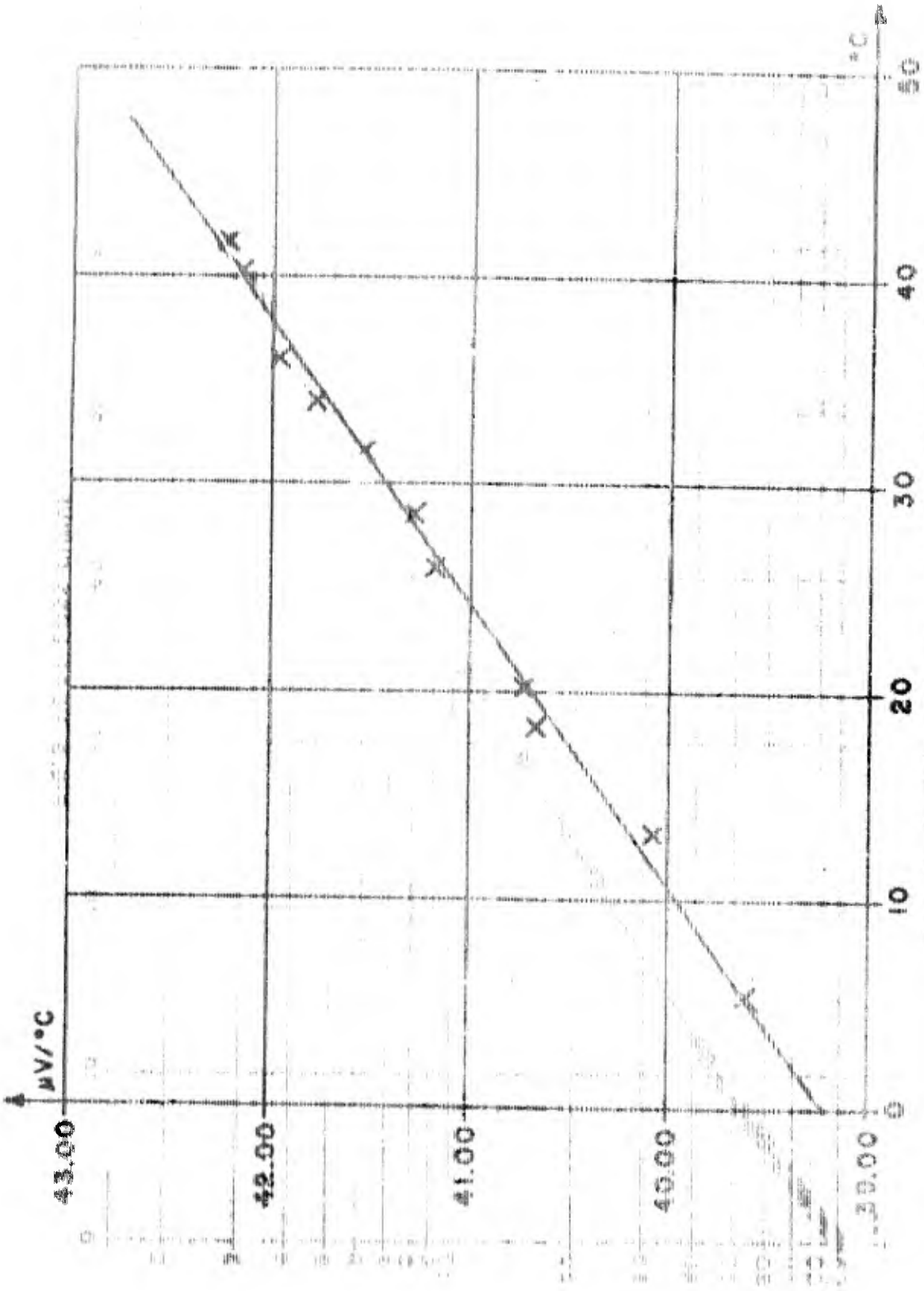


FIG. 4 THERMOCOUPLE CALIBRATION CURVE

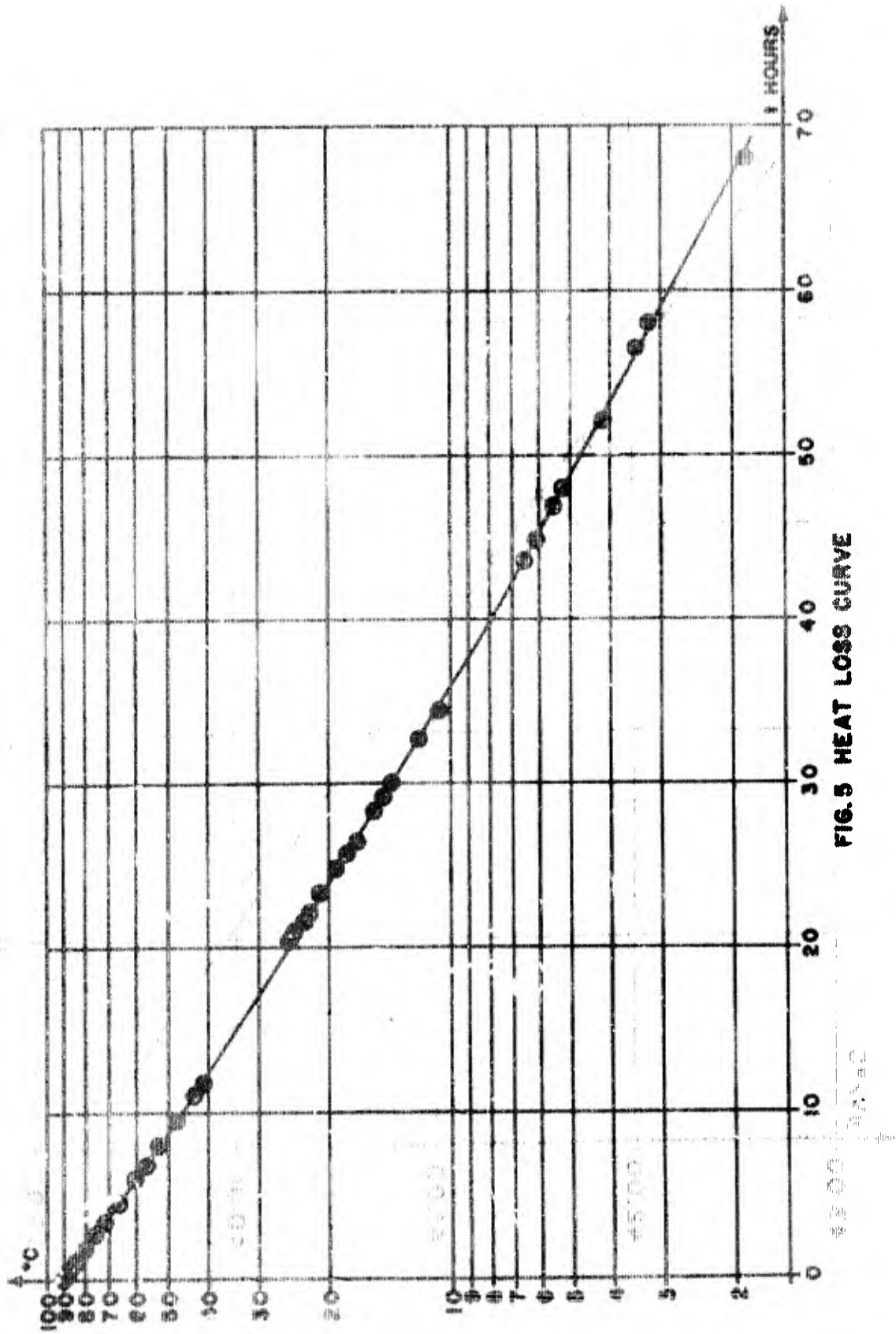


FIG. 5 HEAT LOSS CURVE

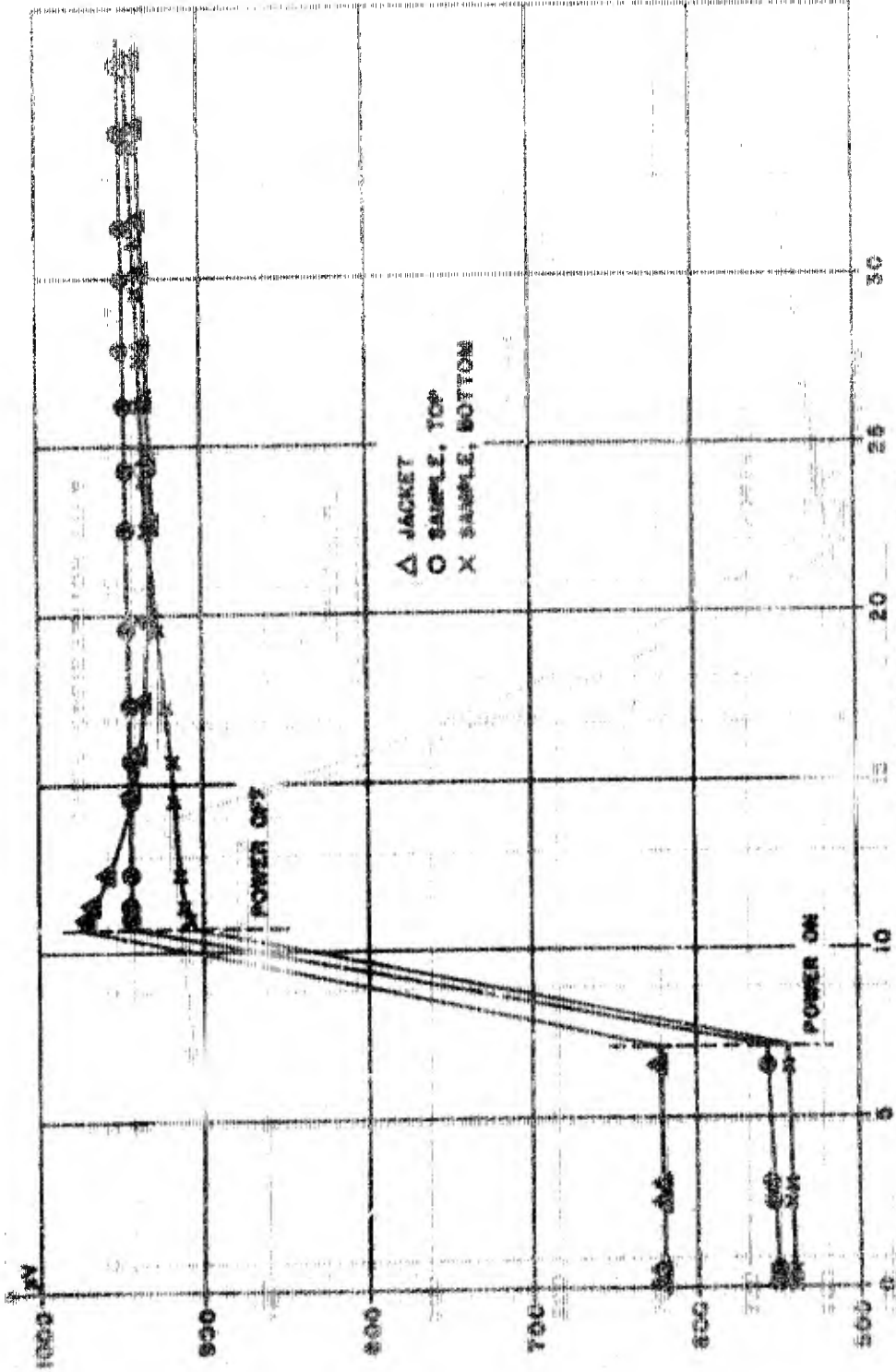


FIG. 8 CALIBRATION NO. 1

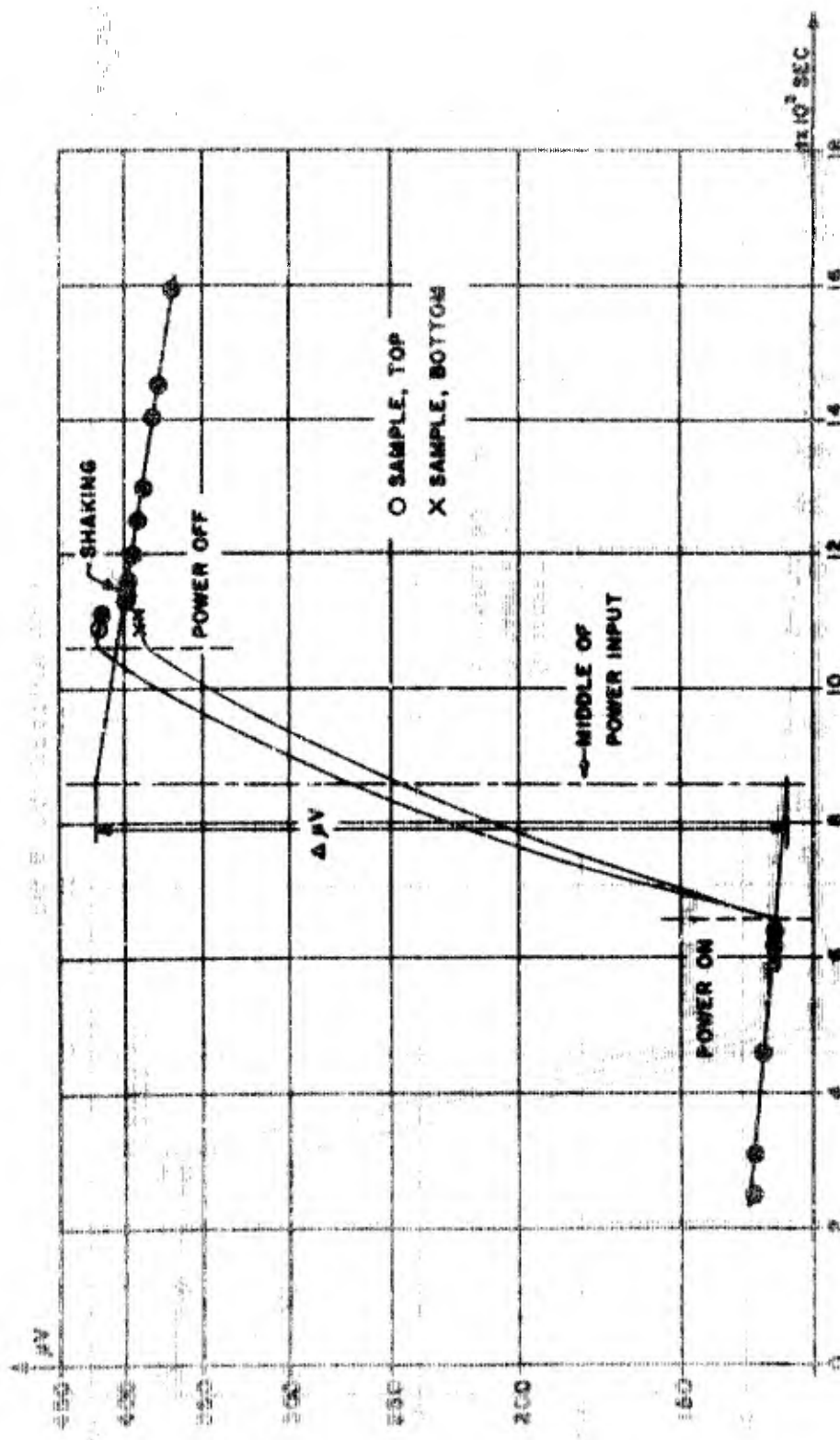


FIG. 7 CALIBRATION III A

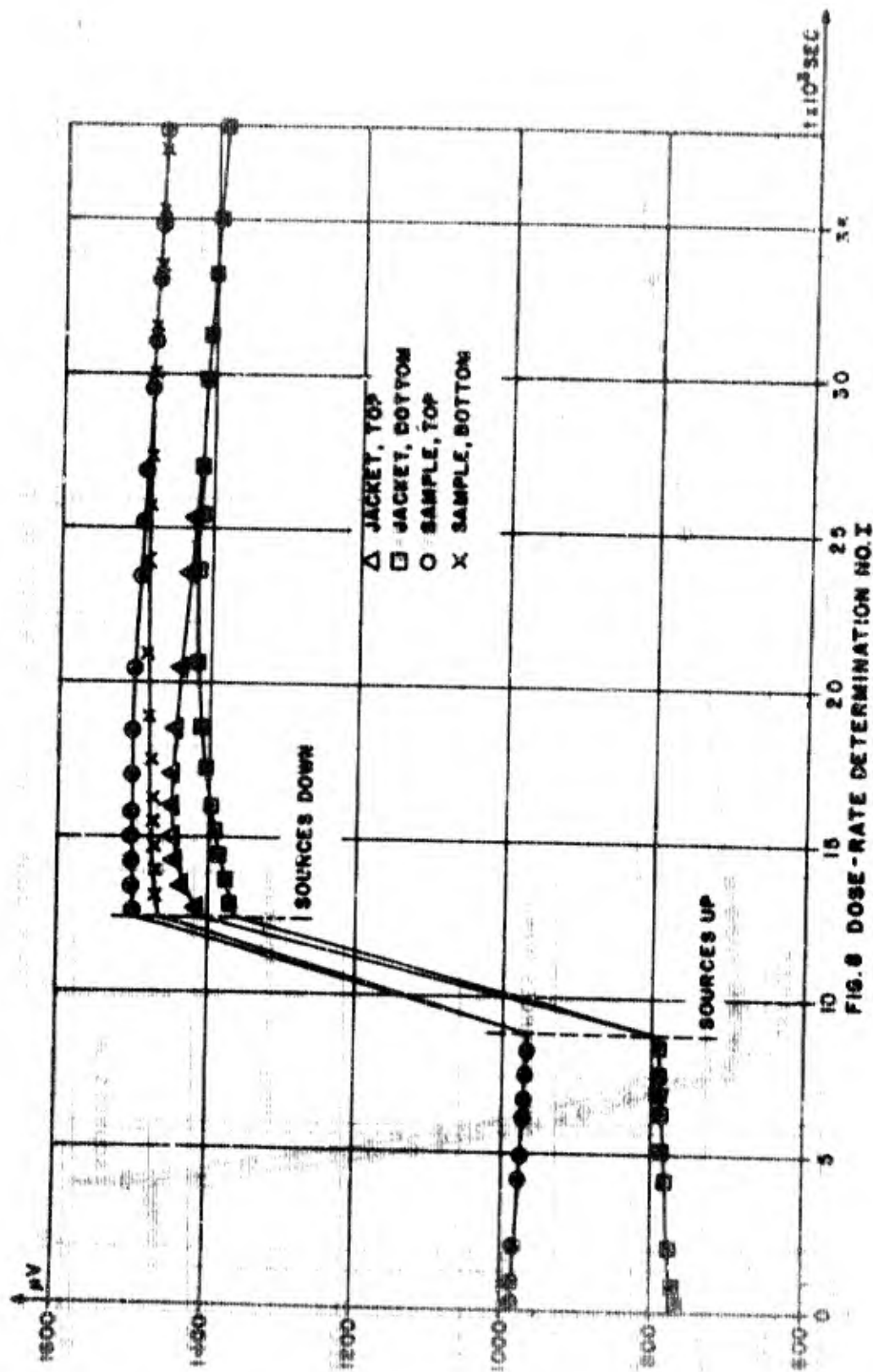


FIG. 6 DOSE-RATE DETERMINATION NO. I

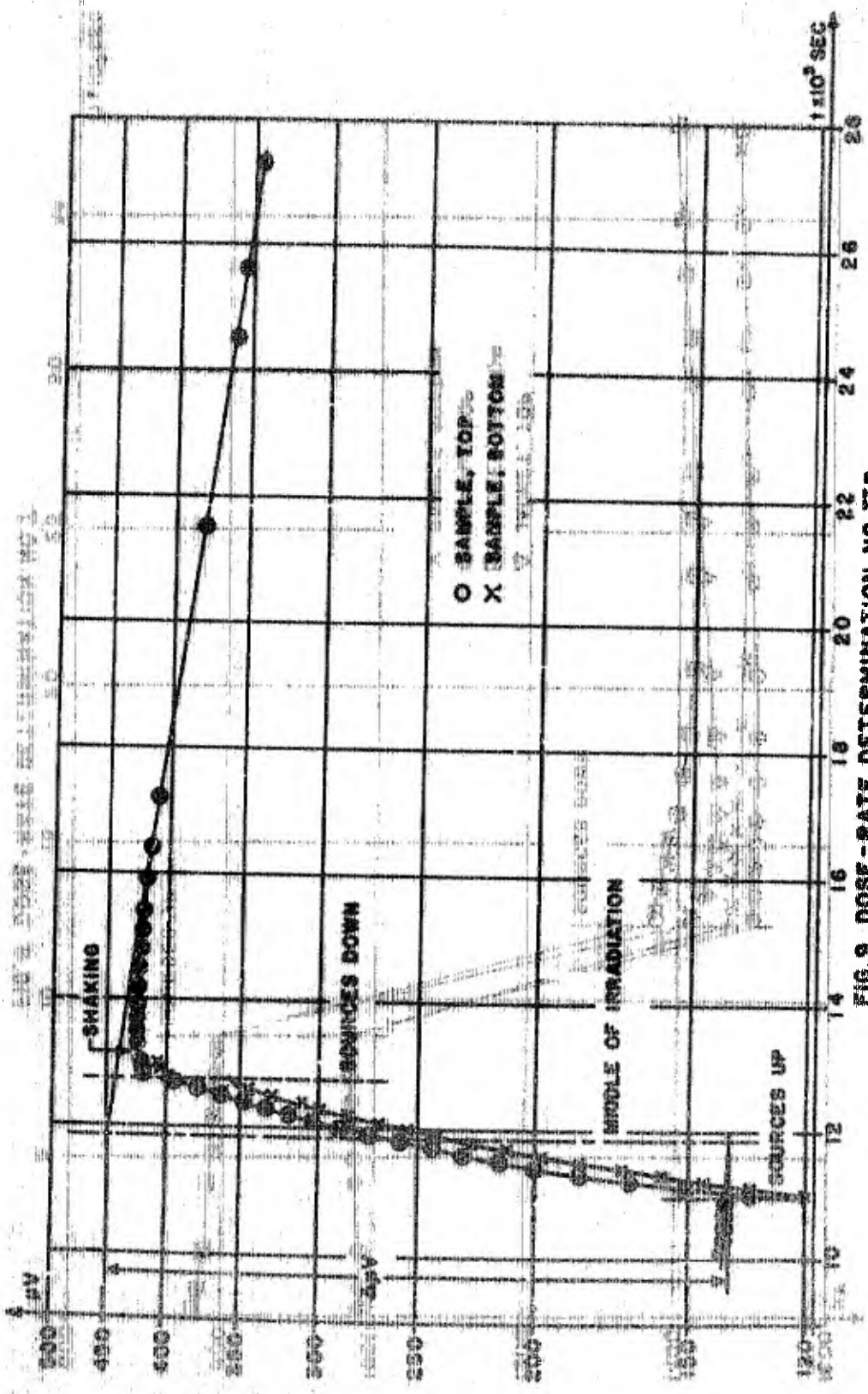


FIG. 9 DOSE-RATE DETERMINATION NO. IIB

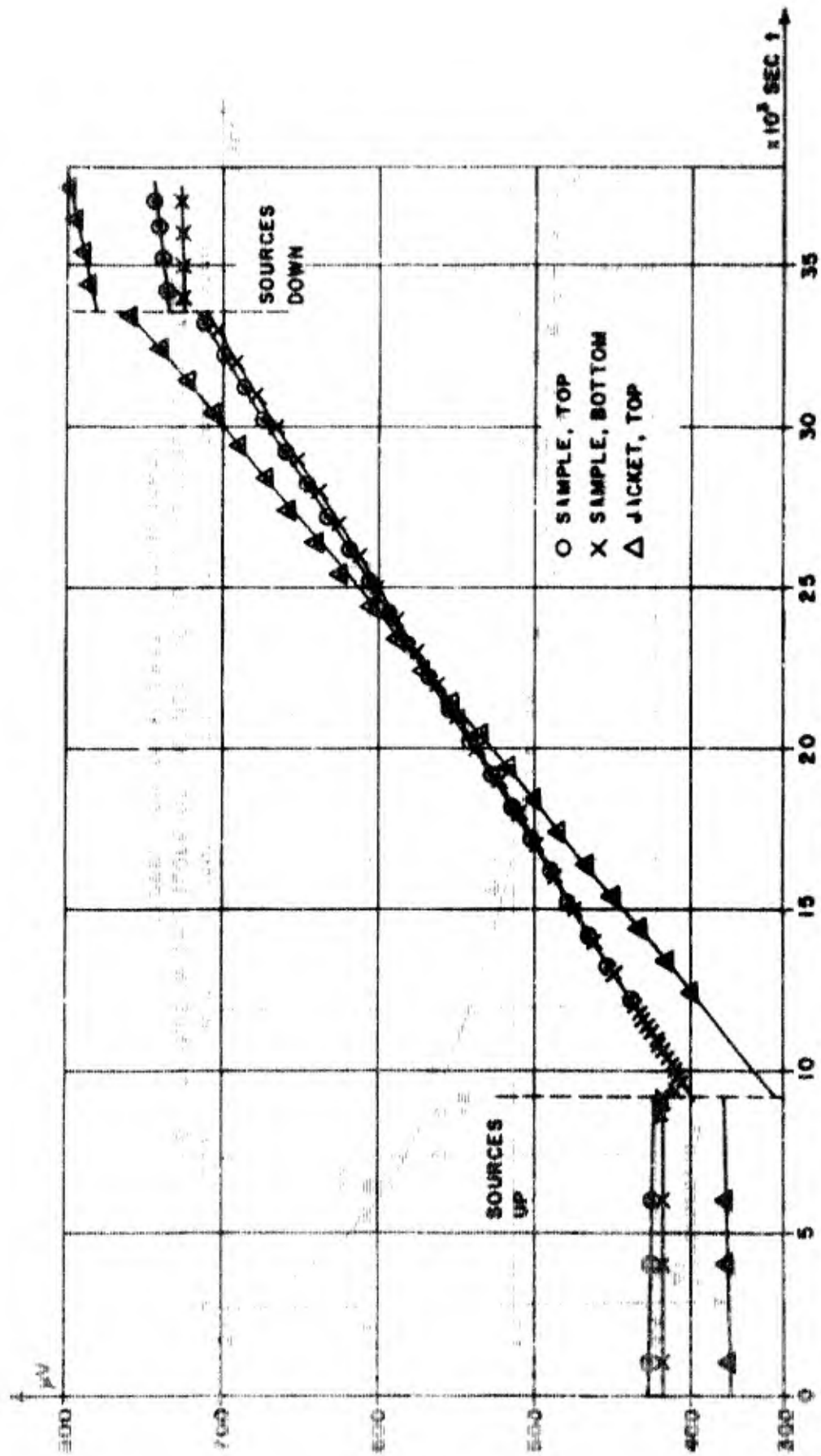


FIG. 10 DCSE-RATE DETERMINATION NO. III C

THE HYDROGEN-BLEND INVESTIGATION NO. III

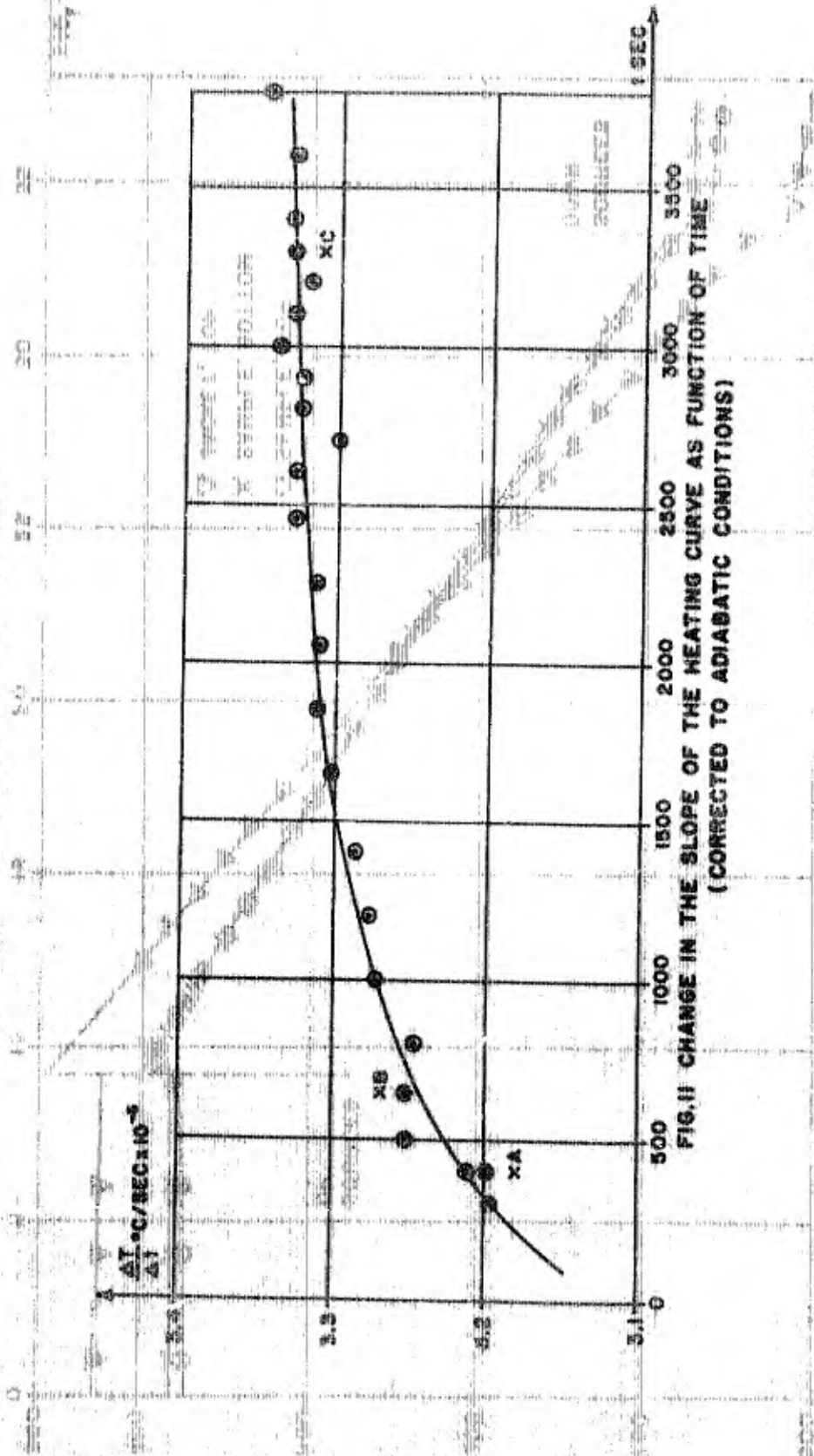


FIG. II CHANGE IN THE SLOPE OF THE HEATING CURVE AS FUNCTION OF TIME (CORRECTED TO ADIABATIC CONDITIONS)

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The thermocouples employed in these experiments were made from the 0.05 mm. diameter insulated Copper-Constantan wire. The junctions were prepared by screwing, twisting, and soldering of the wire pairs. Calibration took place in a temperature controlled water bath using a Mercury normal thermometer as a reference. Since all thermocouples were made from the same batch of wire, no difficulties were encountered in obtaining thermocouples with identical responses.

The change in thermoelectric power per degree centigrade, $\mu V/^{\circ}C$ as a function of temperature, was obtained by making thermocouple readings at different temperatures over the region of interest.

If μV_i and T_i are the i^{th} reading and μV_j and T_j are the j^{th} reading, then at the temperature:

$$\frac{T_i + T_j}{2}$$

we will have:

$$\frac{\mu V_j - \mu V_i}{T_j - T_i} = \frac{\mu V_j}{T_j} - \frac{\mu V_i}{T_i}$$

The response of a group of thermocouples used in these experiments is given in the following table and in Figure 4.

Temperature T	$\mu V/^{\circ}C$
19.14	40.60
19.31	40.08
19.54	40.67
20.20	41.23
25.01	42.17
26.32	41.27
30.01	42.43
35.01	43.13
40.01	43.13
41.75	43.20

APPENDIX III

Properties of the Glass

Effective Mass:

The effective glass mass, i.e. the mass of the part of the Thermos bottle included in the calorimetric body, is not clearly defined, and will vary somewhat with the experimental arrangement. Just how much of the top part of the bottle, i.e. the neck and the edge, should be included, is a function of the following parameters:

- (a) Relative thickness of inner and outer walls.
- (b) Relative diameters of inner and outer walls at the neck.
- (c) Water level inside and outside the Thermos bottle.

In addition, mass and positioning of the cork stopper, along with the natural convection of the atmosphere in contact with the bottle will have a small, yet significant impact on the effective mass.

For the Thermos bottle used here, the inner and outer walls were the same thickness. The ratio of the diameters was 0.7, and the water level was kept similar inside and outside the bottle in all experiments. From this it may be concluded, that all of the inner neck, and approximately 40% of the top edge of the bottle are included in the effective mass.

A quantitative assessment of the contribution of the cork and of the natural convection is difficult. It will tend to reduce the effective mass slightly. In our estimate, this correction will not exceed 1 - 2%.

After completion of all experiments, the outer wall of the Thermos was removed and the following weights were obtained:

An almost identical list of weights was obtained with two other Thermos bottles, indicating that Thermos bottles of the same make are interchangeable for all practical purposes.

Outer Liner With Neck And Edge On Top	76.4 gm
Outer Liner With Neck and Half Edge On Top	75.3 gm
Inner Liner With Neck	73.9 gm
Inner Liner With 2/3 Neck	73.0 gm
Inner Liner With No Neck	69.9 gm

From the figures given above, it is predicted that the effective mass should be somewhere between 73 and 75 grams. The experimental determination of the "effective mass" of the inner liner seems to be very consistent with what should be expected from the figures reported above.

Specific Gravity:

The specific gravity of the glass in the Thermos was determined to be 2.42 gm/cm³.

Composition:

By activation analysis, the metals Na, Ca, and traces of K and Zn were found. No Pb or other heavy metals were present.

Thermal properties were therefore expected to be those of a commercial light alkaline glass.

Temperature Dependence On The Thermal Capacity Of Glass:

The thermal capacity of glass is principally a function of its composition, degree of crystallization and temperature.

The temperature dependence on the thermal capacity of glass is reported by Sharp and Ginther¹² in the form of the following equation:

$$c_{\text{glass}}(T) = \frac{a \cdot T + c_0}{0.00116 T + 1}$$

where:

T = Temperature, °C

c_0 = Thermal Capacity at 0°C

$\alpha \approx 0.5 \cdot 10^{-6}$ for alkaline glass

In the temperature range employed for these experiments, the temperature variation amounts to approximately 0.1% per degree C.

Radiation Absorption Characteristics:

While the mass absorption coefficients for the components are well known (see e.g. Reference 13), the relative energy absorbed by the glass and the water is somewhat difficult to estimate because of the uncertainty in the energy spectrum.

For several simple geometries, such as a point source surrounded by water, the spectrum has been calculated.¹⁴ However, for the complex source geometry which we have, only an educated guess as to the γ -ray spectrum can be made.

From 0.1 MeV and up, glass absorbs about 0.89 times as well as water (provided that the particular glass does not contain lead or other elements with absorption edges in this region). Below 0.1 MeV, glass begins to absorb several times better than water.

It has been estimated that over the whole range, glass probably absorbs somewhere between 0.92 and 1.02 times as well as water.

It was concluded from the above, that an experimental determination was desirable. As described under Dose-Rate Determination No. II, it was found that this particular glass absorbs 0.98 ± 0.02 as well as water.

APPENDIX III

Calorimeter Properties

Calibration I

Mode of Operation: Near Adiabatic

Average Temperature: 18.6°C

Calorimeter Properties:

	Water	Glass	Cork	Wire**
Mass (gm)	287.4	m_{glass}^{+2*}	2	1.2
Thermal Capacity	0.999	$c_{\text{glass}}(18.6^\circ)$	0.4	0.1

Heat Input Data:

Time: 3600 ± 1 sec

Current: 283.31 ± 0.20 mA

Voltage: 11.366 ± 0.010 volts

Energy: 1159.2 ± 1.6 joules

Energy Rate: 3.220 ± 0.005 joules/sec.

Temperature Measurements: (See Figure 6)

$\Delta \mu\text{V}/^\circ\text{C}$ at 18.6°C = $40.60 \pm 0.01 \mu\text{V}$

Reading Before Heat Input: $554.0 \pm 0.2 \mu\text{V}$

Reading Two Hours Later: $931.5 \pm 0.2 \mu\text{V}$

$\Delta \mu\text{V}$: $377.5 \pm 0.3 \mu\text{V}$

Measured Temperature Increase $\Delta T_m = 9.298 \pm 0.015^\circ\text{C}$

* 2 gm is the weight of the glass tube inserted through the cork.

** Mass and heat capacities of the thermocouples are neglected both in the calibrations and in the dose-rate determinations. The contribution is not only very small, but it will also tend to cancel out when excluded in both cases.

Correction ΔT_c for heat transfer from jacket to sample during calibration, as estimated from Figure 6:

$$\Delta T_c = K(T_J - T_S)_{\text{Average}} \cdot \Delta t$$

where:

$$\Delta t = 2 \text{ Hours}$$

$$K = 0.05 \text{ (See Figure 5)}$$

$$(T_J - T_S)_{\text{Average}} = 2.10^\circ\text{C} \text{ (Estimated from Figure 6)}$$

$$\Delta T_c = 2.10 \cdot 0.05 \cdot 2$$

$$= 0.210 \pm 0.004^\circ\text{C}$$

The true temperature increase ΔT_t is then found to be:

$$\Delta T_t = \Delta T_m - \Delta T_c = 9.09 \pm 0.02^\circ\text{C}$$

Results:

The product $(m_{\text{glass}} + 2) \cdot c_{\text{glass}}(T)$ can now be calculated from Equation III in the text:

$$E_{\text{elec}} = \frac{(m_{\text{water}} \cdot c_{\text{water}}(T)) + (m_{\text{glass}} \cdot c_{\text{glass}}(T)) + (m_{\text{cork}} \cdot c_{\text{cork}}) + (m_{\text{wire}} \cdot c_{\text{wire}})}{0.2389} \cdot \Delta T$$

By rearranging the equation and inserting the values given above, one gets:

$$\underline{(m_{\text{glass}} + 2) \cdot c_{\text{glass}}(18.6^\circ) = 15.7 \pm 0.8}$$

Calibration II

Mode of Operation: Constant Temperature Environment

Average Temperature: 25°C

Calorimeter Properties:

	Water	Glass	Cork	Wire
Mass (gm)	288.3	$m_{\text{glass}} + 2$	2	0.5
Thermal Capacity	0.998	$c_{\text{glass}}(25^\circ)$	0.4	0.1

Heat Input Data:

Time: 1860 ± 1 sec.

Current: 387.0 ± 0.3 mA

Voltage: 6.864 ± 0.006 volts

Energy: 4941 ± 6 joules

Energy Rate: 2.656 ± 0.005 joules/sec.

Temperature Measurements:

$$\Delta \mu\text{V}/^\circ\text{C at } 25^\circ\text{C} = 41.05 \pm 0.01 \mu\text{V}$$

Increase in temperature as extrapolated from curve:

$$\Delta \mu\text{V} = 159.2 \pm 0.5 \mu\text{V}$$

$$\Delta T_m = 3.98 \pm 0.02^\circ\text{C}$$

Results:

Like above, the product $(m_{\text{glass}} + 2) \cdot c_{\text{glass}}(25^\circ)$ can now be calculated from Equation III in the text:

$$(m_{\text{glass}} + 2) \cdot c_{\text{glass}}(25^\circ) = 15.6 \pm 0.8$$

Calibration III, A and B

Mode Of Operation: Constant Temperature Environment

Average Temperatures: 8.0°C(A); 6.8°C(B)

Calorimeter Properties:

	Water		Glass		Cork		Wire	
	A	B	A	B	A	B	A	B
Mass (gm)	295.72	242.50	m_{glass}	$m_{\text{glass}} + 121.2^*$	2	2	0.5	0.5
Thermal Capacity	1.0021	1.0022	$c_{\text{glass}}(8.0^\circ)$	$c_{\text{glass}}(6.8^\circ)$	0.4	0.4	0.1	0.1

Heat Input Data:

	A	B
Time:	4000.0 \pm 0.5 sec.	4000.0 \pm 0.5 sec.
Current:	208.96 \pm 0.05 mA	233.30 \pm 0.05 mA
Voltage:	14.605 \pm 0.003 volts	9.508 \pm 0.003 volts
Energy:	12207 \pm 4 joules	8873 \pm 4 joules
Energy Rate:	3.052 \pm 0.001 joules/sec.	2.218 \pm 0.001 joules/sec.

Temperature Measurements: (The temperature curve for IIIA is shown on Figure 7)

$$\Delta\mu\text{V}/^\circ\text{C at } 8^\circ\text{C} \quad (\text{A}): 39.81 \pm 0.01 \mu\text{V}$$

$$\Delta\mu\text{V}/^\circ\text{C at } 6.8^\circ\text{C} \quad (\text{B}): 39.73 \pm 0.01 \mu\text{V}$$

* Mass of broken glass added to the Thermos bottle.

Increase in temperature as extrapolated from the curves:

$$\begin{aligned}\Delta \mu V &= 371.5 \frac{\text{A}}{\text{R}} \pm 0.2 \text{ uV} & 295.3 \frac{\text{R}}{\text{A}} \pm 0.2 \text{ uV} \\ \Delta T_m &= 9.332 \pm 0.012^\circ\text{C} & 7.446 \pm 0.012^\circ\text{C}\end{aligned}$$

Results:

From Equation III in the text, it can be calculated that:

$$m_{\text{glass}} \cdot c_{\text{glass}(8^\circ)} = 15.37 \pm 0.40$$

and

$$(m_{\text{glass}} + 121.2) \cdot c_{\text{glass}(6.8^\circ)} = 40.60 \pm 0.40$$

Assuming the same thermal capacity of the glass at 8°C and at 6.8°C , these two equations can be solved for $c_{\text{glass}(8^\circ)}$ and m_{glass} giving:

$$\underline{c_{\text{glass}(8^\circ)} = 0.208 \pm 0.003 \text{ cal/gm}}$$

$$\underline{m_{\text{glass}} = 74.0 \pm 1.5 \text{ gm}}$$

Dose Rate Determination I

Date: March 27, 1964

Mode Of Operation: Near Adiabatic

Average Temperature: 30.4°C

Calorimeter Properties:

	Water	Glass	Cork
Mass (gm)	266.1	74.0	2.0
Thermal Capacity	0.998	0.214	2.0
a_1	1	1.01	≈ 1
b_1	1	b_{glass}	≈ 1

a_{glass} is determined from dose distribution curve, as obtained by means of chemical dosimeters.⁶

Mass and thermal capacity of the thermocouples are neglected both in the calibrations and in the dose-rate determinations. The contribution is not only very small, but it will also tend to cancel out, when excluded in both cases.

Irradiation Data:

Time: 3600.0 ± 0.5 sec.

Transient Dose: 0.023 ± 0.002 Mrad*

Temperature Measurement: (See Figure 8)

$\Delta\mu\text{V}/^\circ\text{C}$ at 30.4°C = 41.46 ± 0.01 μV

Reading Before Heat Input: 966 ± 1 μV

Reading Two Hours Later: 1478 ± 1 μV

$\Delta\mu\text{V}$: 512 ± 2 μV

Measured Temperature Increase ΔT_m : 12.35 ± 0.10 °C

* Transient dose is the dose delivered to the sample during the operation of raising and lowering the sources, before and after triggering the timer.

Correction ΔT_c for heat transfer from jacket to sample during irradiation is estimated from Figure 8.

$$\Delta T_c = K (T_J - T_S)_{\text{Average}} \cdot \Delta t$$

where:

$$\Delta t = 2 \text{ Hours}$$

$$K = 0.05 \text{ (See Figure 5)}$$

$$(T_J - T_S)_{\text{Average}} = 1.70^\circ\text{C (estimated from Fig. 8)}$$

$$\Delta T_c = -1.70 \cdot 0.05 \cdot 2$$

$$= -0.170 \pm 0.020^\circ\text{C}$$

The true temperature increase ΔT_t is then found to be:

$$\Delta T_t = \Delta T_m - \Delta T_c = 12.52 \pm 0.10^\circ\text{C}$$

Results:

The dose is calculated by the following equation (II in text):

$$D_r = \frac{(m_{\text{water}} \cdot c_{\text{water}}(T)) \cdot (m_{\text{glass}} \cdot c_{\text{glass}}(T)) + (m_{\text{cork}} \cdot c_{\text{cork}}) \cdot \Delta T \text{ Mrads/h}}{(m_{\text{water}} + (m_{\text{glass}} \cdot a_{\text{glass}} \cdot b_{\text{glass}}) + m_{\text{cork}}) \cdot 2.389} \cdot \frac{1}{\Delta t}$$

By inserting the values given above and the value of b_{glass} obtained in the following experiment, the dose rate is calculated to be:

$$D_r = 4.31 \pm 0.04 \text{ Megarads/h}$$

Dose rate corrected to June 16, 1964:

$$D_r = 4.18 \pm 0.04 \text{ Megarads/h}$$

(D_r is corrected for the transient dose in this as well as in the following results.)

Dose Rate Determination II

Dates: June 16, 1964 (A) and July 13, 1964 (B)

Mode Of Operation: Constant Temperature Environment

Average Temperatures: 3.1°C (A); 5.6°C (B)

Calorimeter Properties:

	Water		Glass		Cork	
	A	B	A	B	A	B
Mass (gm)	286.6	250.0	74.0	195.2	2	2
Thermal Capacity	1.0050	1.0034	0.207	0.208	0.4	0.4
a_i	1	1	1.01	1.01	≈ 1	≈ 1
b_i	1	1	b_{glass}	b_{glass}	≈ 1	≈ 1

Irradiation Data:

Time: 1800.3 ± 0.5 sec (A) 1800.2 ± 0.5 sec (B)

Transient Dose: 0.023 ± 0.002 Mrad (A and B)

Temperature Measurements: (The temperature curve for IIB is shown on Figure 9)

$$\Delta \mu\text{V}/^\circ\text{C at } 3.1^\circ\text{C (A)} = 39.45 \pm 0.01 \mu\text{V}$$

$$\Delta \mu\text{V}/^\circ\text{C at } 5.6^\circ\text{C (B)} = 39.63 \pm 0.01 \mu\text{V}$$

Increase in temperature as extrapolated from the curves:

A	B
$\Delta \mu\text{V} = 239.8 \pm 0.2 \mu\text{V}$	$\Delta \mu\text{V} = 306.8 \pm 0.2 \mu\text{V}$
$\Delta T_m = 6.079 \pm 0.005^\circ\text{C}$	$\Delta T_m = 7.742 \pm 0.005^\circ\text{C}$

Results:

Inserting the parameters into Equation II in text, the following equation is derived:

$$D_{r(A)} = \frac{(286.6 \cdot 1.0050) + (74.0 \cdot 0.207) + (2 \cdot 0.4)}{(286.6 + (74.0 \cdot b_{\text{glass}} \cdot 1.01) + 2) \cdot 2.389} \cdot \frac{\Delta T}{\Delta t} \text{ Mrads/h}$$

$$D_{r(B)} = \frac{(250.0 \cdot 1.0034) + (195.2 \cdot 0.208) + (2 \cdot 0.4)}{(250.0 + ((121.2 + 74) \cdot 1.01 \cdot b_{\text{glass}}) + 2) \cdot 2.389} \cdot \frac{\Delta T}{\Delta t} \text{ Mrads/h}$$

where $D_{r(A)}$ and $D_{r(B)}$ are related by the decay equation:

$$D_{r(A)} = D_{r(B)} \cdot e^{\frac{-0.693}{5.3} \cdot \Delta t}$$

correcting for the time elapse between the two experiments.

Solving the equations for b_{glass} and D_r (at June 16, 1964) gives:

$$\underline{b_{\text{glass}} = 0.98 \pm 0.02}$$

$$\underline{D_r = 4.22 \pm 0.02 \text{ Mrads/h}}$$

Dose Rate Determination III

Dates: June 16, 1964 (A); June 22, 1964 (B); and June 22, 1964 (C)

Mode Of Operation: Adiabatic

Average Temperatures: 26.6°C(A); 14.0°C(B); and 41.4°C(C)

Calorimeter Properties:

	Water			Glass			Cork		
	A	B	C	A	B	C	A	B	C
Mass (gm)	290.0	294.7	290.0	74.0	74.0	74.0	2	2	2
Thermal Capacity	0.998	1.000	0.998	0.212	0.210	0.216	0.4	0.4	0.4
a ₁	1	1	1	1.01	1.01	1.01	≈1	≈1	≈1
b ₁	1	1	1	0.98	0.98	0.98	≈1	≈1	≈1

Irradiation Data:

Time: 2040.0 ± 0.5 Sec (A)

2400.0 ± 0.5 Sec (B)

4400.0 ± 0.5 Sec (C)

Transient Dose: 0.023 ± 0.002 Mrads (A, B, and C)

Temperature Measurements: (The temperature curve for IIIC is shown in Fig. 10)

$$\Delta \mu\text{V}/^\circ\text{C at } 26.6^\circ\text{C(A)} = 41.18 \pm 0.01 \mu\text{V}$$

$$\Delta \mu\text{V}/^\circ\text{C at } 14.0^\circ\text{C(B)} = 40.26 \pm 0.01 \mu\text{V}$$

$$\Delta \mu\text{V}/^\circ\text{C at } 41.4^\circ\text{C(C)} = 42.37 \pm 0.01 \mu\text{V}$$

Increase in temperature per second as determined from the curves:

A

$$\Delta \mu V / \text{sec} = 0.1320 \pm 0.0010$$

$$\Delta T_m / \text{sec} = 0.003205 \pm 0.000030^\circ\text{C}$$

B

$$\Delta \mu V / \text{sec} = 0.1296 \pm 0.0008$$

$$\Delta T_m / \text{sec} = 0.003205 \pm 0.000025^\circ\text{C}$$

C

$$\Delta \mu V / \text{sec} = 0.1412 \pm 0.0006$$

$$\Delta T_m / \text{sec} = 0.3333 \pm 0.000016^\circ\text{C}$$

Results:

Inserting the parameters given above into Equation II in text, the following dose rates are found:

$$D_r = 4.03 \text{ Mrads/h (A)}$$

$$D_r = 4.10 \text{ Mrads/h (B)}$$

$$D_r = 4.20 \text{ Mrads/h (C)}$$

Correcting the (A) and (B) results to stationary conditions (See Figure 11) gives the following sets of results:

$$\underline{D_r = 4.18 \pm 0.04 \text{ Mrads/h (A)}}$$

$$\underline{D_r = 4.21 \pm 0.03 \text{ Mrads/h (B)}}$$

$$\underline{D_r = 4.20 \pm 0.02 \text{ Mrads/h (C)}}$$